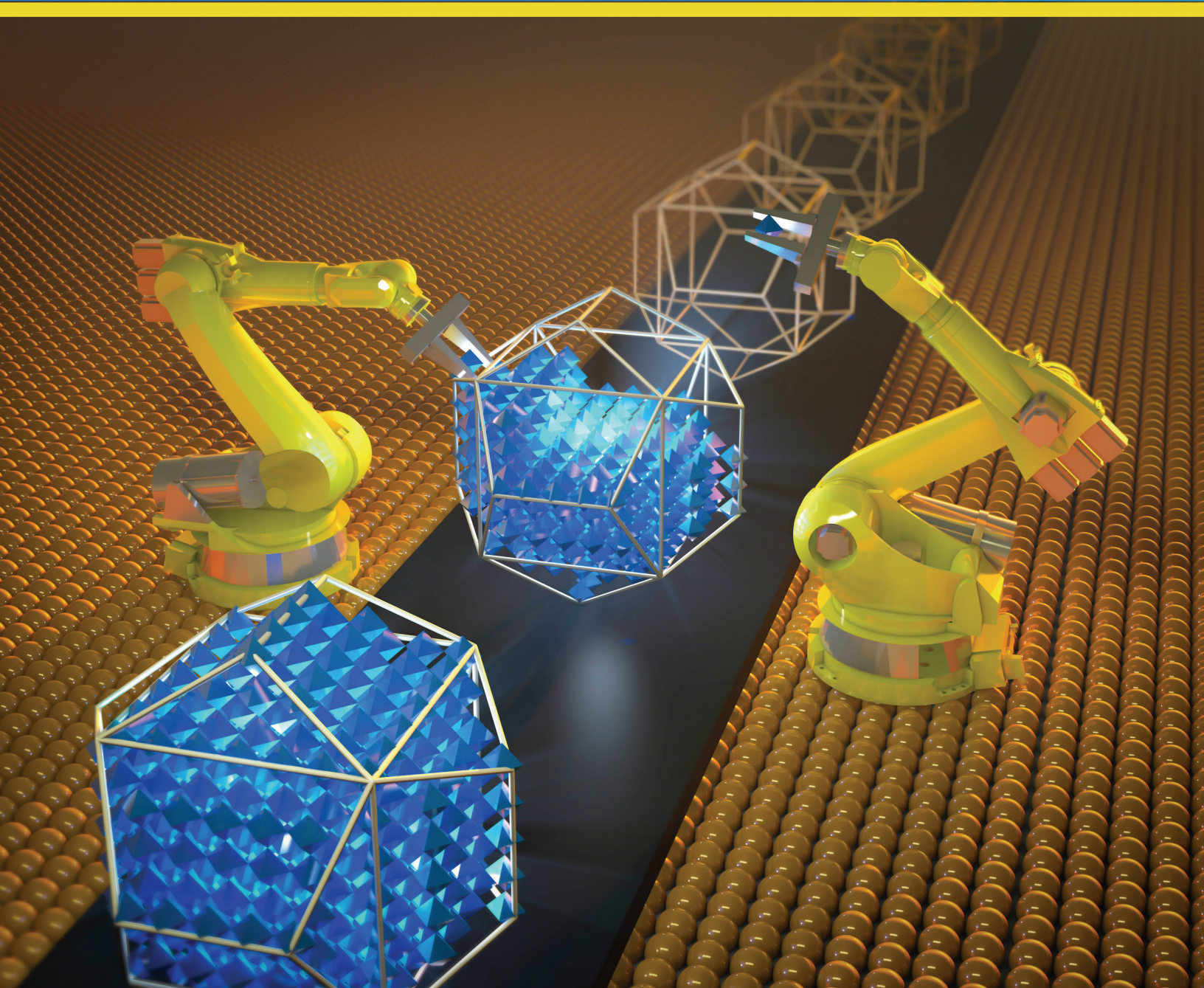


Basic Research Needs for Transformative Manufacturing



*Report of the Basic Energy Sciences Workshop on
Basic Research Needs for Transformative Manufacturing
March 9 – 11, 2020*

The artwork on the cover is a graphic representation of autonomous synthesis of atomic-scale building blocks for novel materials with unparalleled structures and functions. To revolutionize manufacturing, fundamental advances in precision synthesis, atom and energy efficiency in chemical processes, operando characterization, multi-scale modeling, validation, and methodologies for simultaneously achieving multiple performance objectives are needed.

Image courtesy of Oak Ridge National Laboratory.

Basic Research Needs for Transformative Manufacturing

Fundamental science to revolutionize manufacturing

Report from the US Department of Energy, Office of Basic Energy Sciences Workshop
March 9 – 11, 2020
Rockville, Maryland

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Abbreviations

0D	zero-dimensional
1D	one-dimensional
2D	two-dimensional
3D	three-dimensional
AFM	atomic force microscopy
AI	artificial intelligence
AM	additive manufacturing
AMO	DOE Advanced Manufacturing Office
aPPO	apactic polypropylene oxide
BES	DOE Office of Basic Energy Sciences
BRN	Basic Research Needs
CAMERA	Center for Advanced Mathematics for Energy Research Application
CT	computed tomography
DFT	density functional theory
DOE	Department of Energy
DPD	dissipative particle dynamics
EDS	energy-dispersive x-ray spectroscopy
EJ	exajoule
FEL	free-electron laser
fs	femtosecond
GHG	greenhouse gas
HEDM	high-energy diffraction microscopy
HPC	high-performance computing
HTE	high-throughput experimentation
iPPO	isospecific polymerization of propylene oxide
IR	infrared
LED	light-emitting diodes
Li	lithium
MAS	magic-angle-spinning
ML	machine learning
MOF	metal-organic framework
MS	mass spectroscopy or microsecond
NIST	National Institute of Standards and Technology
NO _x	nitrogen oxides
NLS	National Synchrotron Light Source
PCAST	President's Council of Advisors on Science and Technology
PDF	pair distribution function
PRD	priority research direction
ps	picosecond
R&D	research and development
s	second
SAXS	small-angle x-ray scattering
SEM	scanning electron microscopy/microscope
SLM	selective laser melting
ssNMR	solid state nuclear magnetic resonance
TEM	transmission electron microscopy/microscope
YAG	yttrium aluminum garnet

Executive Summary

Manufacturing is central to the nation's prosperity and security—it currently represents about 12% of the gross domestic product, provides nearly 13 million jobs, and accounts for about 25% of energy use. The economy relies heavily on numerous manufacturing subsectors that face common challenges, including how to best take advantage of available process data or inexpensively obtain needed additional data; a lack of physics- and chemistry-based models across length and time scales; a need for more sustainable processes; and supply chain constraints in a global marketplace. Advances in basic science are required to best meet these challenges and transform manufacturing. For example, how could understanding the issues of scaling from molecules to materials lead to the manufacturing of better, more economical batteries with higher capacity or reduce the energy consumption for industrial chemical production? Additionally, entirely new approaches to manufacturing and new products are needed and often arise from basic scientific research.

To identify fundamental scientific opportunities and determine priority research directions (PRDs) that could accelerate innovation to transform manufacturing in the future, the US Department of Energy (DOE) Office of Science, Office of Basic Energy Sciences (BES), held a workshop on Basic Research Needs for Transformative Manufacturing in March 2020. DOE's Advanced Manufacturing Office (AMO) within the Office of Energy Efficiency and Renewable Energy has long worked with industry to advance its mission to catalyze the research, development, and adoption of energy-related advanced manufacturing technologies and practices to drive US economic competitiveness and energy productivity. BES worked with AMO to identify technological barriers and manufacturing issues and to produce a factual document about the current state of manufacturing for discussion by invited workshop participants. (The factual document is posted along with this report at <https://science.osti.gov/bes/Community-Resources/Reports>.) Along with leading scientists and engineers from academia and national laboratories, industry representatives served as keynote speakers to provide additional perspectives.

This report is the first US government examination of basic energy science needs for manufacturing. The identified PRDs provide a basic science strategy that underpins applied technology research. To transform manufacturing, fundamental advances in synthesis, processing, modeling, operando characterization, and validation are needed. As manufacturing processes become increasingly data driven and fully networked, integration of autonomous sensing and control will enable greater productivity and competitiveness. Finally, co-design will tackle data, control, and design across components, delivering multiple system-level performance criteria simultaneously. Research based on these priorities will lay the scientific foundation to go beyond incremental improvements to create new, transformative technologies for manufacturing that are energy efficient and sustainable.

The workshop was attended by more than 140 leading national and international scientific experts from academia, national laboratories, and industry. In a first for this workshop series, 40% of the participants attended virtually because of the emerging global pandemic. The five topical and one crosscutting panels identified five PRDs that should serve as the foundation for future DOE basic research to transform manufacturing. These PRDs are highlighted in the following paragraphs, along with a summary of the underlying critical opportunities for each one.

Achieve Precise, Scalable Synthesis and Processing of Atomic-Scale Building Blocks for Components and Systems

Innovations that enable precise and scalable synthesis and processing will accelerate the transition from current manufacturing methods to new paradigms for creating unparalleled structures and functions. Application-specific materials with unprecedented performance at manufacturing scale will emerge from

targeted synthesis and processing of building blocks, components, and systems that are precisely controlled at the atomic scale.

Integrate Multiscale Models and Tools to Enable Adaptive Control of Manufacturing Processes

Linkages between small-scale physics and chemistry and macro-scale nonequilibrium processes and component performance are not fully understood, limiting the achievable precision and functionality of products. A combination of multiscale modeling, in situ diagnostics, and an online decision-making framework is needed to realize adaptive manufacturing processes and guarantee component qualification.

Unravel the Fundamentals of Manufacturing Processes Through Innovations in Operando Characterization

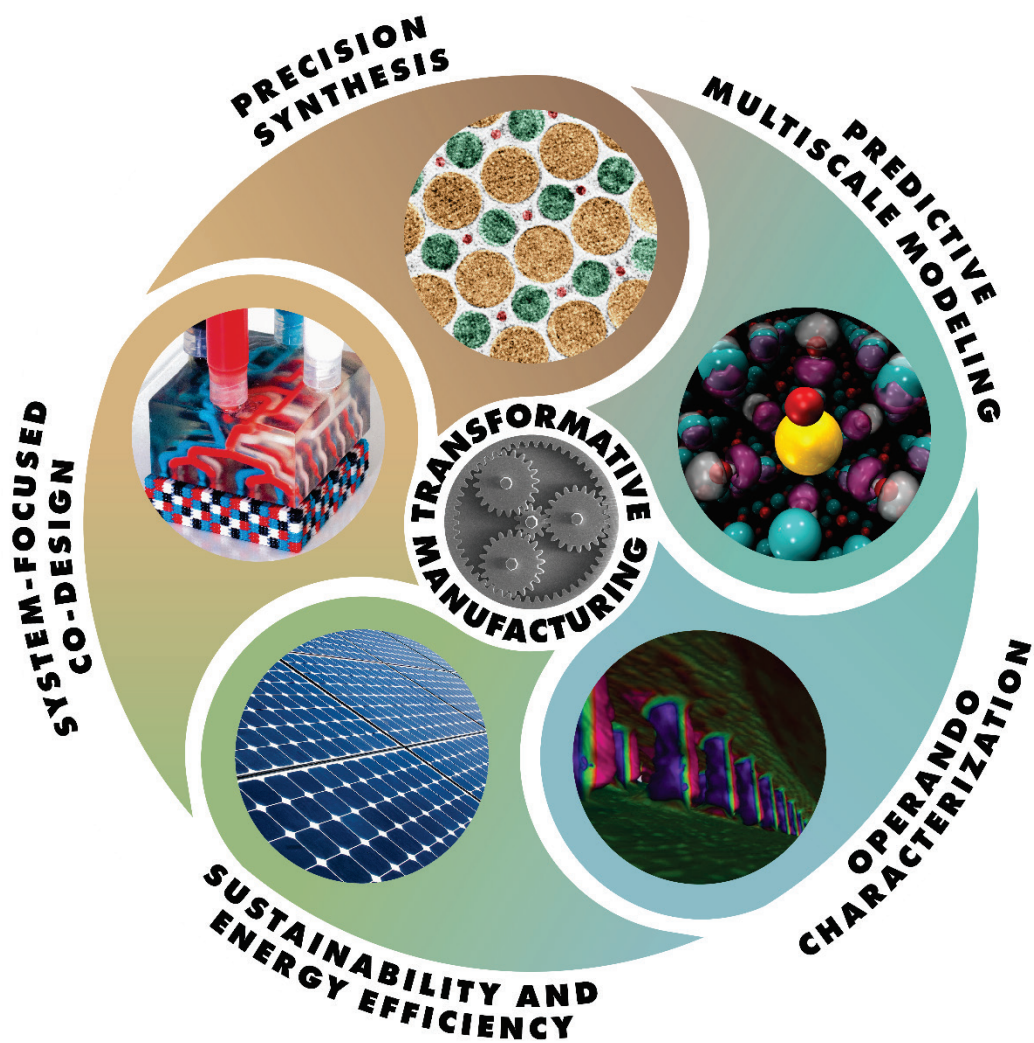
In 21st century manufacturing, many processes are still practiced as “art” instead of science. Frequently, there is insufficient fundamental understanding to tailor and control materials and processes so that they perform exactly as desired, with minimum energy consumption and maximum efficiency. Operando characterization—direct visualization and characterization under actual manufacturing conditions—will provide the knowledge needed to transform the science of manufacturing.

Direct Atom and Energy Flow to Realize Sustainable Manufacturing

Sustainable manufacturing requires localizing energy delivery and directing atom- and energy-efficient chemical and materials processes. Synergistically using diverse forms of energy (e.g., electrical, thermal, radiative, and mechanical) coupled with understanding phenomena across length scales would enable sustainable, high-efficiency processes. Scientific advances could support the design of circular feedstocks that minimize waste and reduce the use of critical materials in existing and future chemicals and materials, moving toward resiliency.

Co-design Materials, Processes, and Products to Revolutionize Manufacturing

Co-design is a paradigm that provides scientific foundations for the creation of new materials, chemical processes, or systems by addressing the ubiquitous manufacturing challenge of simultaneously satisfying multiple performance objectives. Exciting opportunities exist to meet this challenge with new approaches integrating predictive modeling and experimental data with system resiliency, circularity, and operability. Doing so will enable the navigation of the near-infinite range of possible designs to identify inherently resilient systems.



The image is courtesy of Oak Ridge National Laboratory and Argonne National Laboratory, and portions are reprinted with permission from the American Chemical Society and Springer/Nature.

1. Introduction

Manufacturing plays a central role in how we live and work. From the cars we drive to our cell phones and computers, as well as our homes and clothing—all rely on manufacturing innovations. Advances in manufacturing have been forged over centuries of inventions, yet key challenges remain. Now, perhaps more than ever, basic science is poised to transform manufacturing. Emerging opportunities are possible as a result of recent scientific advances in the complexity of systems that can be studied, including, among others: characterization tools that probe materials and chemical processes with unprecedented temporal and spatial resolution, enabling in situ and operando analysis of devices and processes; new chemistries and materials that are ushering in device miniaturization; artificial intelligence and robotics that are speeding up discoveries. With energy costs exceeding \$150 billion a year¹ for manufacturers in the United States, new ways to both reduce costs and ensure a circular economy are also needed. Figure 1 is a Sankey diagram showing the sources of energy and energy consumption by US manufacturing.

U.S. Manufacturing Sector (TBtu), 2014

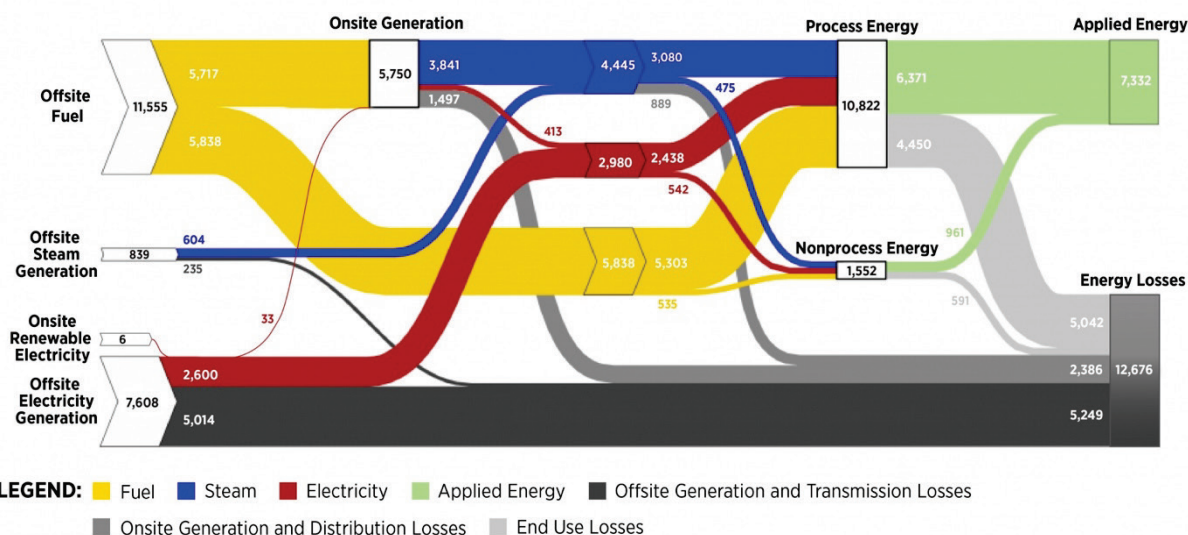


Figure 1. The US manufacturing sector Sankey diagram shows how total primary energy is used by US manufacturing plants. It is based on the US Energy Information Administration's manufacturing energy consumption survey data for 2014 and updated assumptions as of May 2019. Source: Image provided by DOE Advanced Manufacturing Office, [Static Sankey Diagram Full Sector Manufacturing \(2014 MECS\)](#).

The President's Council of Advisors on Science and Technology (PCAST)² defined manufacturing as

a family of activities that (a) depend on the use and coordination of information, automation, computation, software, sensing, and networking, and/or (b) make use of cutting edge materials and emerging capabilities enabled by the physical and biological sciences.

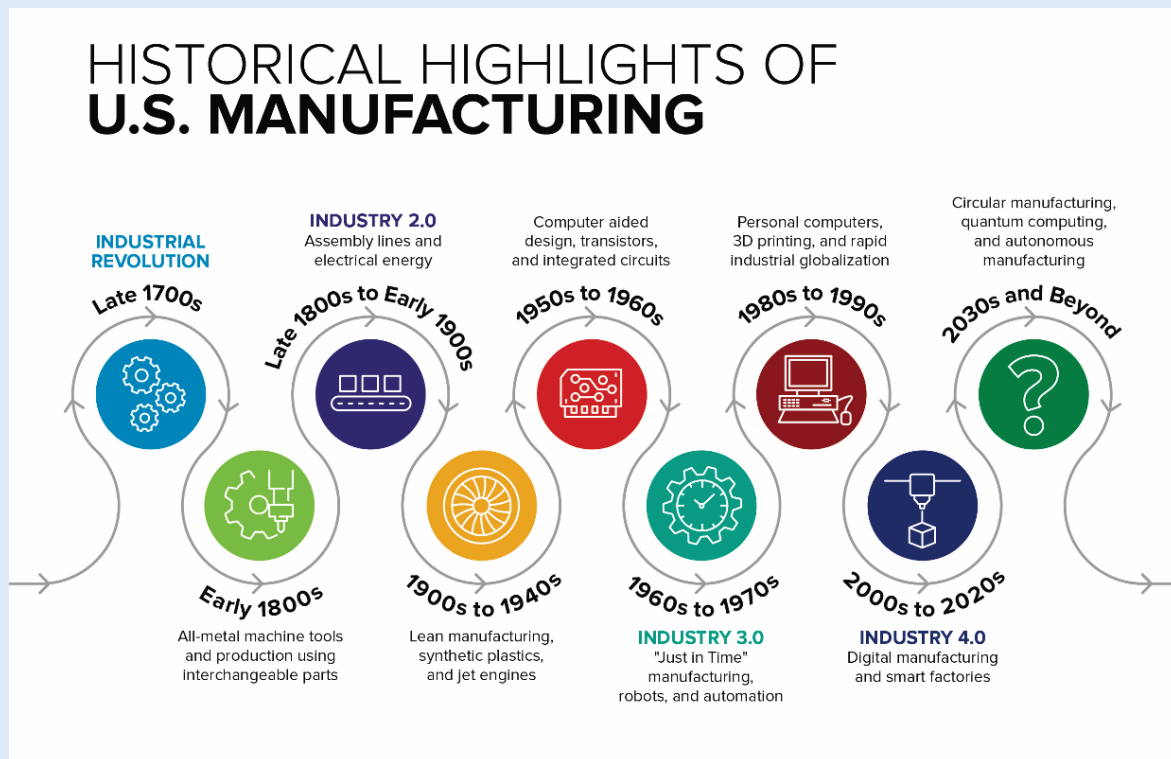
This workshop was designed to capture the highest-priority fundamental research directions that could lead to transformative impacts on manufacturing, and the priorities laid out build upon this definition.

The breadth of basic science provides many possible pathways to transform. For example, new methods that analyze vast amounts of process information, enhance automation, model chemical reactions and material syntheses, and enable networking and data mining are opportunities for transformation. These new methods will be enabled by further advances in data science, computation, robotics, and high-

performance computing. The design and discovery of new characterization tools and the integration of current capabilities with these new tools could be particularly beneficial to manufacturing.

Sidebar: A Short History of US Manufacturing

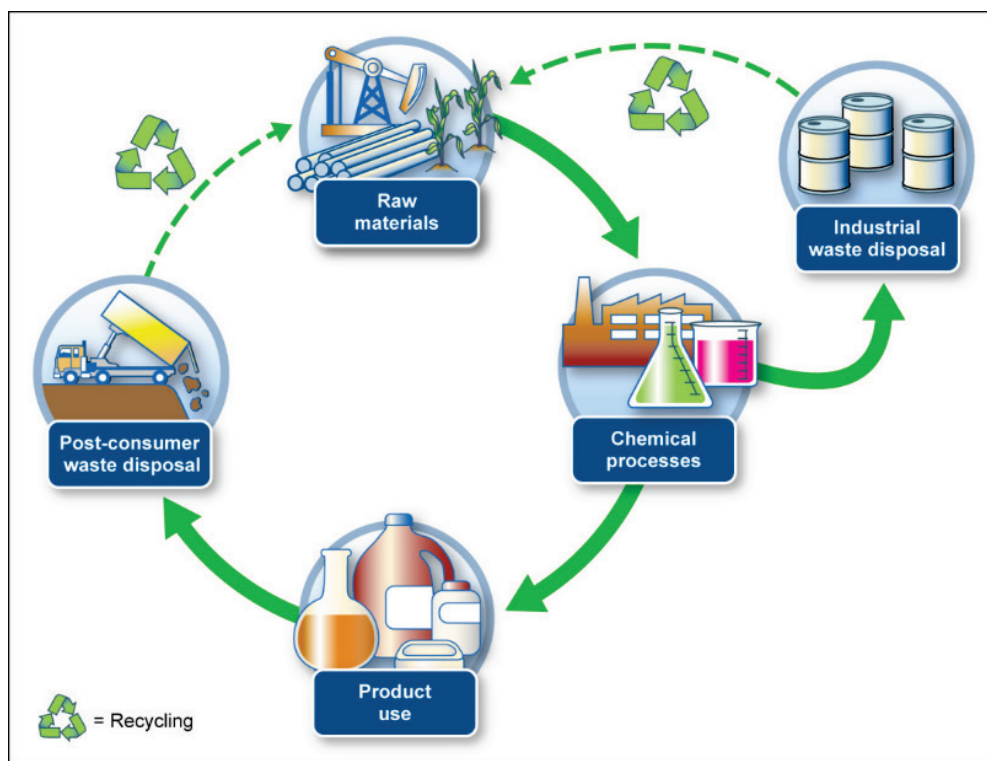
The first known use of the word “manufacture” dates back to 1567. The word derives from Latin *manu factus*, meaning “made by hand.” Clearly, manufacturing has come a long way since then. US manufacturing origins include an automated grain mill that used conveyor belts, dating back to 1787. This era represents what some call “Industry 1.0.” The “American System of Manufactures” has its origins in the 1800s and was named to contrast it with the English system; it refers to the prevalence in US industry of production of goods using a sequential series of operations performed using purpose-built machinery to make interchangeable parts³ — the beginning of Industry 2.0. The 1900s brought about dramatic advances as assembly lines were introduced and the integrated circuit was invented. In the 1970s, automation of manufacturing lines become pivotal for beginning Industry 3.0. And, most recently, 3D printing and smart manufacturing has ushered in Industry 4.0. US manufacturing today is made up, predominantly, of small businesses with only about 4000 of the roughly 250,000 businesses having more than 500 employees. Manufacturing today is adaptable and lean and ready for innovation. Moving forward, the gaps that have prevented the seamless movement of basic science innovations to market and manufacturing challenges will be overcome through new research. New products will be designed and new manufacturing methods will be developed through discovery science.



Source: Image provided by Argonne National Laboratory.

The discovery of methods, processes, and materials as yet unvented could revolutionize manufacturing processes. Recent examples of methods currently revolutionizing manufacturing include additive manufacturing, which allows distributed manufacturing of customized products, and artificial intelligence, which is driving manufacturing autonomy. Basic science can provide new capabilities to advance what can be manufactured and how it can be manufactured.

Other approaches include further incorporating sustainability and circular economy innovations into manufacturing. Opportunities here include finding ways to incorporate sustainability into product design and manufacturing processes, ensuring facile reuse of products at end-of-use, and developing new capabilities needed to further enable the use of recycled feedstocks. Considerable opportunities exist for basic scientific innovations that will accelerate the adoption of a circular economy. Figure 2 illustrates opportunities for sustainable chemical manufacturing.



Source: GAO. | GAO-18-307

Figure 2. Life cycle of chemical processes and products. *Source:* Image provided by Government Accounting Office, <https://www.gao.gov/assets/690/689951.pdf>.

In today’s global manufacturing market, existing sources of raw materials, and their corresponding supply chain issues, underscore the need for scientific discoveries of alternative raw materials and of ways of processing them into products. Also needed are new routes for processing chemicals and materials that reduce reliance on foreign sources of feedstocks.

Basic science is needed to overcome current challenges in moving discoveries to market. Bridging common “valleys of death” that inhibit moving basic science innovations to market, such as those shown in Figure 3, is vital. There are long-standing challenges, for example, in scaling production from the laboratory benchtop to batch and continuous reactors. Another current manufacturing challenge stems from the incomplete understanding of device and component interoperability that can lead to performance issues; opportunities abound for improved integration at all stages of manufacturing, including the co-

design of products, wherein the multiple phases of research and development (R&D) are considered in an integrated way.

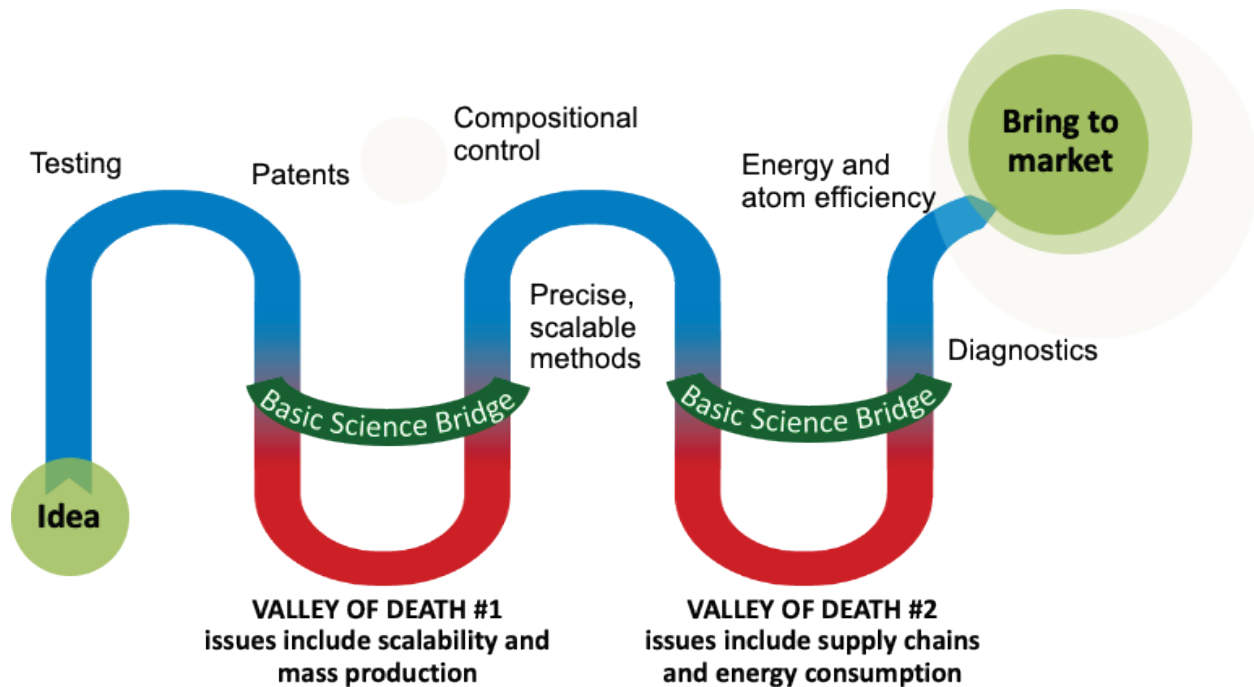


Figure 3. Basic science is the key to overcoming current manufacturing challenges in moving discoveries to market by bridging common “valleys of death.” Source: Image provided by Argonne National Laboratory.

In every sense of the PCAST definition of manufacturing, this workshop report provides a framework for furthering knowledge of the physical sciences to establish capabilities and provide cutting-edge results that enable such coordination from atoms and molecules to products, and from test tubes to tankers.

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2. Report to the President on Ensuring American Leadership in Advanced Manufacturing; President’s Council of Advisors on Science and Technology. Washington, DC, 2011.
3. Hounshell, D. A. From the American System to Mass Production, 1800–1932: The Development of Manufacturing Technology in the United States; Hopkins University Press: Baltimore: 1984.

2. Workshop Organization

This was the first workshop of its kind to examine how basic energy science can drive manufacturing forward and innovate new ways to manufacture goods. Rather than organizing the workshop around the subsectors of manufacturing, such as chemical manufacturing, primary metal manufacturing, machinery and so on,¹ the workshop's six panels were structured by themes that predominantly crossed multiple subsectors. The thought was that doing so would help reveal opportunities for discussions about fundamental research needs that could impact multiple manufacturing areas, and thus help identify the highest priorities for fundamental research.

The following are the workshop's six panels:

1. Precision Synthesis Science
2. Processing and Scale-Up Science
3. System Integration Science
4. Sustainable Manufacturing
5. Digital Manufacturing
6. Crosscutting Topics

Panel 1 considered precision synthesis, deposition, assembly, and deterministic organization in synthetic molecules and materials, ranging from atomic and molecular scales, to low-dimensional and nanostructures, through the meso-scale to the macro scale. This panel considered experimental, computational, and especially combined experimental and computational approaches in precision synthesis science. The themes included, but were not limited to, sequence-specific molecules and polymers, atom-specific low-dimensional and bulk inorganic materials, deterministic assembly of architectures and functional systems, and materials genome and machine learning strategies for materials design and optimization.

Panel 2 centered around the often-complicated processing steps considered acceptable in prototyping to enable volume manufacturing, by either scaling up or numbering up. Moving technology from carefully controlled laboratory settings into industrial processes remains a significant challenge. This panel discussed the state of the art in processing and scale-up science and considered how processing may evolve in a future state with new raw material and energy inputs.

Panel 3 focused on new technologies that require the integration of multiple devices or components that function in complex physical environments for economic benefits. The panelists discussed common classes of "show-stoppers" that emerge in the later stages of R&D that can be addressed if deeper understanding of key basic science issues is available. Looking at the changing energy-generation landscape, impacts were considered for manufacturing and the development of phenomena and approaches that can drive the discovery of new hybrid, integrated, or decentralized processes.

Panel 4 reimagined the landscape of opportunities in sustainable manufacturing, where innovative concepts, materials, and processes make possible substantial reductions in resource and energy consumption and mitigate environmental impacts. Based on overall energy intensity, manufacturing processes for high-volume chemicals, polymers, and structural and building materials are likely to benefit the most from new discoveries that lead to breakthrough innovations. Additional understanding of how to effectively de-manufacture materials at the molecular level for expedient recovery and reuse in manufacturing supply chains was considered.

Panel 5 focused on opportunities relevant to digital manufacturing. Digital manufacturing involves the application of computational systems to manufacturing process streams from conception and design to

final product. Discussions included identifying and exploring the fundamental scientific questions, underlying physics, and existing gaps, which—if bridged—can advance and solidify digital manufacturing with true scientific underpinning.

Panel 6 was aligned across the other five panels. It sought to identify crosscutting themes that will enable seamless integration of characterization, sensors, process design, computer simulation, algorithm development, machine learning, and advanced analytics and enable manufacturers to be successful in an increasingly competitive economic environment. For many high-tech manufactured products, the precision required is not possible through human ability, so automation and robotic technologies will be increasingly needed. Opportunities for scientific innovation as a result of these areas were also discussed.

Leading experts were invited from across academia, national laboratories, and industry to be a part of each panel. To provide a common understanding of the current state of manufacturing, program managers and experts funded through DOE’s Advanced Manufacturing Office (AMO) assembled a document about the current state of manufacturing; a draft version of this document was provided to all panelists before the workshop, and it is available through the DOE Office of Science website. During the workshop, the panels met separately to discuss their particular panel focus area and came up with possible priority research areas. Together, all the panels identified common priority themes that emerged from across the panels and that led to the Priority Research Directions (PRDs) discussed herein.

References

1. US Bureau of Labor Statistics. *Industries at a Glance*. <https://www.bls.gov/iag/tgs/iag31-33.htm> (accessed December 1, 2020).

3. Priority Research Directions

The workshop panelists identified five PRDs, listed in Table 1, to enable transformations and significant advancements in manufacturing. In the sections that follow, each PRD is discussed in depth.

Table 1. List of Priority Research Directions and key questions.

1. Achieve precise, scalable synthesis and processing of atomic-scale building blocks for components and systems
<i>What are the mechanisms needed for manufacturing multiscale, atomically and molecularly precise materials? How can basic research uncover structure–function relationships across multiple scales in components and systems? How can chemical processes readily be scaled from laboratory results?</i>
2. Integrate multiscale models and tools to enable adaptive control of manufacturing processes
<i>What are the frameworks required to model, monitor, and ultimately control manufacturing processes that tightly couple physics and chemistry across scales? How can complex multiscale models be translated to fast surrogate models for process control?</i>
3. Unravel the fundamentals of manufacturing processes through innovations in operando characterization
<i>How can manufacturing processes and products be “visualized” at the atomic level, in real time, and under operating conditions to reveal the intricate details of underlying physical or chemical events? How can these insights be used in control schemes that inform decision making?</i>
4. Direct atom and energy flow to realize sustainable manufacturing
<i>What are the methodologies to achieve atom and energy efficiency for sustainable manufacturing? How can science enable adaptive and resilient manufacturing across scales to exploit renewable or recycled feedstocks?</i>
5. Co-design materials, processes, and products to revolutionize manufacturing
<i>How can bottom-up scientific discovery be combined with top-down system-focused design to identify new and efficient manufacturing modalities? What new approaches will allow the control of matter in the presence of impurities and/or nonequilibrium states? How can science enable multiple performance objectives to be achieved simultaneously for complex, multicomponent processes?</i>

Summary

Manufacturing plays a vital role in the national economy, and manufacturing innovations impact lives on a daily basis. Even with tremendous advances, many manufacturing challenges remain. The opportunities for new materials and chemical processes to be discovered and moved to market are endless, and they will be enabled by scientific innovations that provide revolutionary advances in manufacturing methods. These innovations require multidisciplinary approaches that fully integrate modeling, data analytics, digital assembly, and operando characterization not only to revolutionize manufacturing, but also to decrease life cycle energy, reduce resource requirements, and lessen economic impacts.

This workshop report discusses five PRDs that comprise fundamental science strategies to accelerate innovation and solve industry’s toughest challenges in manufacturing. While distinct, the five PRDs identify basic research needs that share common themes, including the need to bridge fundamental understanding across length and time scales, to precisely and adaptively control syntheses and other manufacturing processes at the atomic and molecular scales, and to develop and validate high-speed computational, characterization, and analysis tools that control and optimize manufacturing processes in real time. The realization of these common scientific themes will transform manufacturing at a faster pace.

PRD 1: Achieve Precise, Scalable Synthesis and Processing of Atomic-Scale Building Blocks for Components and Systems

Summary

Innovations that enable precise, scalable synthesis and processing will accelerate the transition from current manufacturing methods—which form, for example, materials with limited atomic precision—to new manufacturing paradigms that result in materials with unparalleled structures and functions arising from precisely placed atoms and macromolecules made through precise molecular assembly. Application-specific products with unprecedented performance at manufacturing scale will emerge from targeted synthesis and processing of building blocks, components, and systems that are precisely controlled first at the atomic or molecular scale, and subsequently at the nano-, micro-, and macro-scales. While molecules are already precise, and the value of such precision is absolutely clear in pharmaceuticals and materials such as liquid crystals, such precision is not present in most manufactured products. If precise synthesis could be realized at manufacturing scale, the benefits would be unprecedented. Autonomous synthesis and processing in manufacturing offer opportunities for faster discovery of optimal pathways to products with improved efficiency and carbon intensity through precise control of materials and reactants, as well as operating conditions. Materials and complex molecules with unparalleled properties for energy storage, harvesting, and use, as well as materials for future computing, materials with remarkable mechanical properties, new catalysts, water purification membranes, and materials with exceptional thermal properties would become available, to name some applications. Background for this PRD can be found in previous Basic Research Needs reports, including the report on synthesis science¹ for a discussion of strategies to form precise atomic-scale building blocks. While applications in quantum information science and microelectronics are not the focus of this BRN report, the BRN roundtable reports on quantum information science^{2,3} and the BRN reports on quantum materials⁴ and microelectronics,⁵ provide examples of where manufacturing of precise atomic-scale building blocks could impact these important areas. The BRN report on catalysis⁶ notes also areas that precision synthesis could impact.

Precise, scalable synthesis and processing of atomic-scale building blocks requires fundamental understanding of chemical and physical forces and synthetic methods that will serve as first steps for the synthesis and assembly of multi-component, multiscale structures. Fundamental understandings of chemical and physical forces and the resultant interactions within and between components also will enable the discovery of components and systems containing multiple, unconventional combinations of desirable functions (Figure 4). These hierarchical systems have architectures that go beyond pure and periodic structures to embrace compositional and structural complexity and to intentionally introduce defects and interfaces to add function. Energy applications^{5,7,8} often require considerable volumes of material, and the precise synthesis and processing methods discovered must be scalable; this is key to making an impact. Research will yield new theoretical/computational tools and experimental techniques to design and characterize multiscale structures and properties of functional organic and inorganic building blocks relevant for larger-scale manufacturing designed to meet the energy challenges of the 21st century.

This PRD seeks to address key questions that include the following: *What are the mechanisms needed for manufacturing multiscale, atomically and molecularly precise materials? How can basic research uncover structure–function relationships across multiple scales in components and systems? How can chemical processes readily be scaled from laboratory results?*

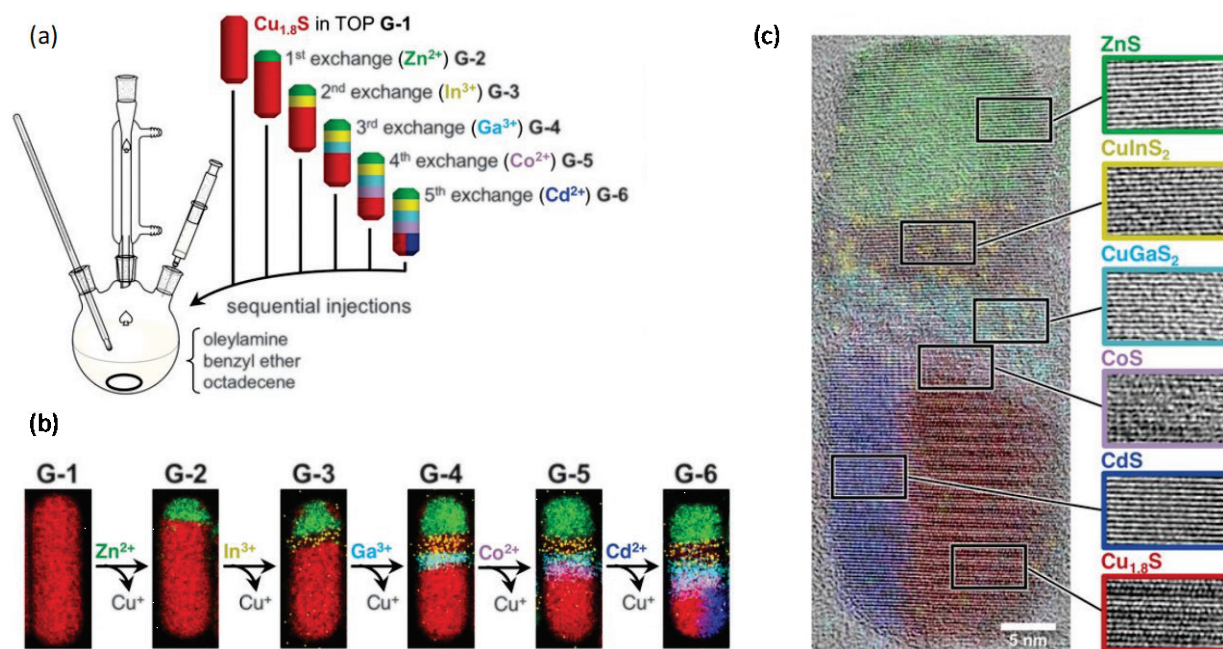


Figure 4. Rational construction of multiple generations (G) of nanorods synthesized by cation-exchange reactions. (a) Schematic showing the reaction setup and injection sequence. (b) Scanning transmission electron microscopy–energy-dispersive x-ray spectroscopy (EDS) element maps for each nanorod generation. The elements Cu, Zn, In, Ga, Co, and Cd are shown in red, green, yellow, teal, purple, and blue, respectively. (c) High-resolution transmission electron microscopy image with overlaid EDS map highlighting the crystallinity of each material within the G-6 nanorod. Source: From [B.C. Steimle et al. Science 367, 418, 2020](#). Reprinted with permission from AAAS.

Scientific Challenges

The first scientific challenge is the need to design multi-functional and multi-structural materials that are precise across multiple scales, including those with complex organization. The complex functions of energy production, conversion, storage, and use are enabled by hierarchical design, often involving heterogeneous materials with organic, inorganic, or mixed components (see Figure 5). For example, high-efficiency catalysts with uniquely determined locations of multifunctional active sites that are arrayed within constrained reaction spaces and connected by channels for delivery of reactants to drive a serial cascade of reactions would be quite powerful. Systems providing a linked cascade of reactions have already been designed for enzymatic systems⁹ and developing analogous systems in chemistry and materials science is an ongoing challenge. Similarly, nanostructured photonic materials use coupling across length scales to create properties not possible in homogeneous materials.^{10,11} Although remarkable structures have been created for many proof-of-principle demonstrations, major science gaps exist at many levels that limit or prevent their large-scale manufacture. First, there is a need to develop more effective and efficient synthetic and purification strategies to realize the required precision, quality, and purity of fundamental building blocks for both organic and inorganic systems. Second, new insight is required for the design of fundamental building blocks for the efficient assembly of predictable equilibrium and nonequilibrium hierarchical structures with targeted functions. Third, manufacturing often imposes additional constraints such as cost, product life-cycle, resource impact, and safety that require the development of new science beyond the discovery phase of synthesis and processing to realize impactful hierarchical structural and multifunctional materials systems at scale.

The second scientific challenge is to avoid or overcome undesirable states through pathway engineering. The energy landscape and the synthesis reaction pathway is often complex and characterized by numerous local energy minima. A key question is what block design modalities (i.e., shapes, interactions,

anisotropic characteristics, and chemical selectivities) and what processing principles (i.e., confinement, equilibrium, kinetic effects, active control, time- and chemistry- synchronized processes, external stimuli) can be used for steering system formation toward the targeted structure? The discovery of underlying principles and establishment of practical methods will enable the manufacture of nanomaterials and chemicals with prescribed organization at multiple length scales. Already well known in biological systems,^{12–14} pathway engineering has recently been explored in all-organic and organic-inorganic hybrids^{15–21} and nanoparticle lattices²² to drive systems to the desired states. Use of information-rich materials and steered processes will allow control of material formation.

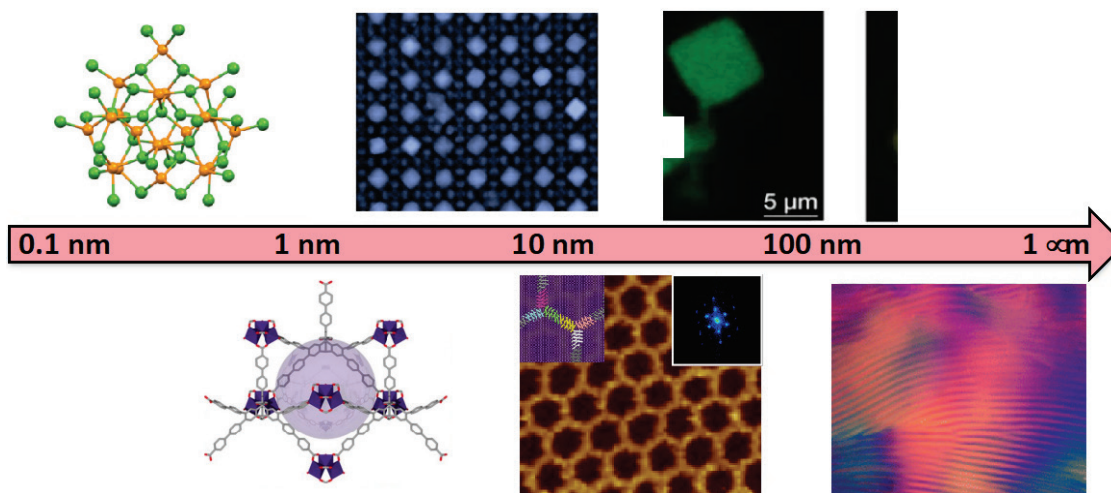


Figure 5. Hierarchical design from atomistic to micron scales generates novel architectures and functions. At the shortest length scale, atomically precise clusters—shown here for In_{37}P_2 —are stabilized by organic ligands (not shown), while complex colloidal liquid crystal phases exhibit features at the micron scale. Highly porous metal organic framework compounds, nanoparticle superlattices, self-assembled protein lattices, and DNA origami-nanoparticle hybrid materials. Ordered three-dimensional nanomaterials using DNA-prescribed and valence-controlled material voxels span the intervening scales. Sources (L to R, top to bottom): • J.L. Stein et al. Cation exchange induced transformation of InP magic-sized clusters, *Chem. Mater.* 29, 7984–7992, 2017, <https://pubs.acs.org/doi/10.1021/acs.chemmater.7b03075>. Used with permission of the American Chemical Society (ACS). Requests for permission for further use must be directed to the ACS. • L. Rossi et al. *Soft Matter* 7, 64–67, 2011; permission obtained through Copyright Clearance Center. • From Z. Chen et al. *Science* 368, 297–303, 2020. Reprinted with permission from AAAS. • Reprinted with permission from Y. Kang et al., *J. Am. Chem. Soc.* 135, 42–45, 2013. Copyright 2013 American Chemical Society. • Reprinted by permission from Nature Publishing Group: *Nature*, [Controlling protein assembly on inorganic crystals through designed protein interfaces](#), H. Pyles et al. 571, 251–256, 2019. • Reprinted by permission from Nature Publishing Group: *Nat. Mater.* [Ordered three-dimensional nanomaterials using DNA-prescribed and valence-controlled material voxels](#), Y. Tian et al., 19, 789–796, 2020.

A third challenge is the need to manage and design defects and disorder. Defects and disorder in manufacturing associated with either materials or processing can take on many roles; it must be possible to identify and characterize them and manage them accordingly. In semiconductor manufacturing, for example, precision synthesis of very complicated hierarchical and heterogeneous layered materials systems must be carried out with near perfection in each of hundreds of processing steps to produce a functioning system. In other materials, such as steel and other metal alloys, defects and disorder (e.g., phase-separated inclusions, dislocations, grain boundary segregated atoms) are tuning knobs with respect to material properties. For example, in single-site catalysts, which are becoming increasingly important for both hydrocarbon processing and ammonia synthesis,⁶ a major barrier is placing the active atoms into structurally identical sites. This is particularly challenging in systems containing inorganic components. Molecular self-assembly is arguably one of the most promising strategies for high-volume cost-effective

manufacturing at the nanoscale in both hard and soft matter systems, although major challenges remain with management of defects and disorder. In soft matter systems especially, information is encoded into the building blocks to program the materials to spontaneously form regular nanostructures down to molecular length scales with targeted function. However, often the areas or volumes over which materials self-assemble with adequate perfection are quite small (micrometer-scale), while systems with high perfection over macroscopic dimensions are required for industrial relevance. In this case, strategies to mitigate or control defects and disorder include the application of system-biasing external fields or templates in conjunction with self-assembly in processes sometimes referred to as “directed self-assembly.” Controlling defects and disorder in systems with truly macroscopic dimensions, however, remains a significant challenge.

Defects need not have a pejorative connotation and can be desirable. Defects have been used effectively in semiconductor and quantum devices. If manufacturing protocols and the enabling science can be developed to enable control of defects in bulk materials—as is done in the semiconductor industry—the impact on materials for applications ranging from energy storage to separation processes would be significant. Formally, in optoelectronic devices, donors and acceptors are atomic impurities, with low carrier ionization energies, that are introduced to make *n*- and *p*-type semiconductors. Energetically deeper impurities are intentionally created in semiconductors to shorten carrier lifetimes and realize high-speed photodetectors. Atomic- and molecular- defects in crystals have long been prized for the colors they create in minerals, including in precious stones, laser materials, and more recently as spin qubits for quantum information science.^{2,3} In the latter application, defect complexes such as the nitrogen vacancy in diamond serve as quantum emitters capable of photon indistinguishability. Defects (e.g., grain boundaries, dislocations, vacancies) exist in most engineering materials; but in those materials, unlike in advanced semiconductors, there is generally considerable disorder in the chemistry and spatial positioning of the defects. New computational and experimental methods to design, create, and position defects in materials at manufacturing scale, outside the microelectronics and quantum realms, would be powerful. Geometric and topological defects, including symmetry-breaking defects, may enable the realization of optical, acoustic, and mechanical materials with unconventional or superior properties. Development of design and synthesis tools for such applications will be critical for the realization of these new functions.

A fourth challenge is to precisely control the arrangement of atoms and molecules at surfaces and interfaces. The boundary between two materials or between a liquid and a solid, as examples, often either imparts desired functionality or play a deleterious role in system performance. Understanding and synthetically controlling gas, solid, and liquid interfaces of the type seen in various devices or in chemical processing, including separations, remains an important research challenge to practical realization and manufacture of many materials systems employed in energy technologies, for example in hydrocarbon refining.²³ The nature of interfaces in soft, hard, and hybrid materials or liquid and gaseous chemicals—or even between different phases—can determine how energy, charge carriers, and ions move through matter. Whether the interface contains imperfections or inhomogeneities or is atomically coherent, whether it is compositionally abrupt or graded, and whether it is chemically accessible or inert is a key factor in both the initial material performance and the robustness or resiliency of a material or system over time.

In addition to the importance of interfaces in inducing or modulating performance, surfaces and interfaces are at play during the assembly and integration of materials and building blocks, or in directing synthesis operations. Controlling surface chemistry, for example through patterning, is often key to enabling the assembly or processing of components such as block copolymers into more complex architectures and devices.²⁴ Designer surfaces are also key to advanced manufacturing of energy systems—such as catalysts, membranes and quantum devices—with respect to the arrangement of functional motifs with atomic or nanoscale control (see Figure 6). Patterned and selectively reactive and self-limiting surface chemistry forms the basis of atomic layer processing methods, including atomic and molecular layer

deposition and atomic layer etching. Creating designer interfaces in materials will increase in importance with the proliferation of additive manufacturing, a process that by its very nature often introduces large numbers of interfaces into a structure.

A fifth challenge is the need for high-volume, massively parallel, down to atomic-scale positioning of atomic and molecular building blocks, and functional system components. Semiconductor manufacturing represents one of the engineering marvels of the modern world. It offers tools, processes, and concepts for wafer-scale fabrication that should be leveraged when possible or applied to advance scalable precision synthesis and processing for applications outside the semiconductor space. Challenges exist in the further evolution of these tools to reach the single-nanometer or atomic length scale. For example, synthetic methods continue to develop that enable larger atomically precise clusters of atoms; what manufacturing technologies might be leveraged for assembling these building blocks into macroscopic systems? Conversely, in the realm of tool development, can arrays of actuators or parallel scanning probes be developed that allow for the assembly of precise building blocks at the manufacturing-relevant speed, cost, and scale required for applications with lower added value per unit volume than microelectronics? Opportunities exist for the creative use of the most advanced tools as a starting point for directed self-assembly of precise building blocks or in conjunction with selective patterning, deposition, or etching of conformal layers, or self-limiting reactive layers, to produce functional heterogeneous and hierarchical materials systems.

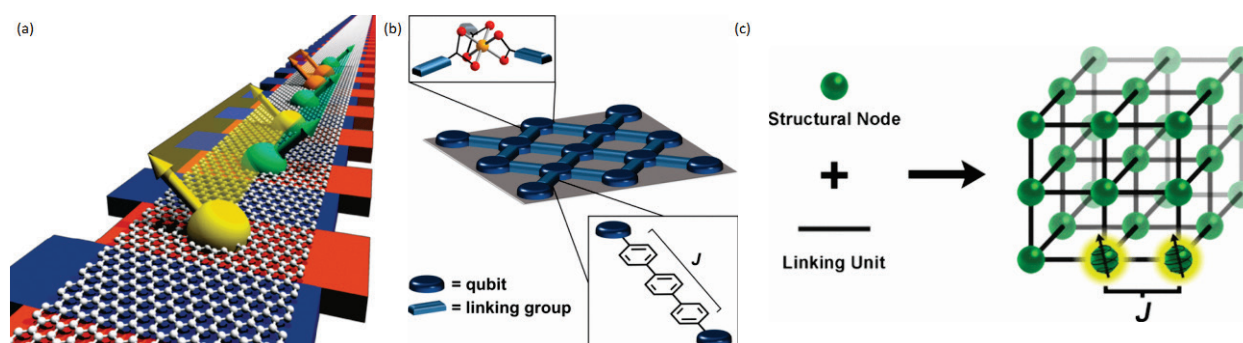


Figure 6. Precise, scalable quantum materials synthesis. (a) Spin qubits along a monolayer graphene nanoribbon with armchair edges. (b) Schematic diagram of an array of surface qubits, showing both a potential linker and metal complex geometry. (c) Qubits (glowing components) can be installed in metal organic frameworks via selection of proper structural nodes or linking moieties. Magnetic interactions (J) between qubits in (b) and (c) are open to synthetic fine-tuning via proper choice of bridging units. Sources: (a) Reprinted with permission from Nature Publishing Group: *Nat. Rev. Mater.* [2D materials for quantum information science](#), X. Liu and M. Hersam, 4, 669–684, 2019. (b–d) M.J. Graham et al. *Chem. Mater.* 29(5), 1885–1897, 2017.

Research Thrusts

Thrust 1. New scalable atom-, electron-, and energy-efficient synthetic routes to precision materials and chemical products

With the transition toward scalable syntheses, there is a need to develop economically viable and sustainable chemistries. For example, in polymer syntheses, much can be learned from concepts related to atom economy developed in organic synthesis. CH-activation reactions are being widely investigated in small molecule syntheses.²⁵ These reactions have translated well for the synthesis of semiconducting polymers in terms of direct arylation polymerizations²⁶ and continue to be further developed for a diverse range of applications. However, challenges remain in controlling the degree of branching and homocoupling. For polymer synthesis in general, moving toward the use of first-row transition metal catalysts or organic catalysts for sustainability, and using photocatalysis and electrocatalysis to enable reactions that are otherwise difficult to achieve at room temperature, will allow for more energy-efficient

processes while achieving greater control over sequence specificity. Of particular relevance are chemistries in which atoms present in the precursors, and the energies required to link the atoms into molecules and polymers are as close to the thermodynamic limit as possible. As an example, consider frontal polymerization (Figure 7),²⁷ a process, in which a self-propagating exothermic reaction wave transforms liquid monomers to fully cured polymers. The energy required to drive the polymerization is the heat liberated by the polymerization, and all of the atoms in the monomer are present in the polymer. Because the heat is generated internally, this approach scales to almost any size. Also, because the heat is generated internally, there is no external equipment that needs to be heated (and then cooled), further reducing the energy consumption of the process. The composition of the resulting polymer is exactly the composition of the monomer, and available functionalities are limited only by that of the monomer.

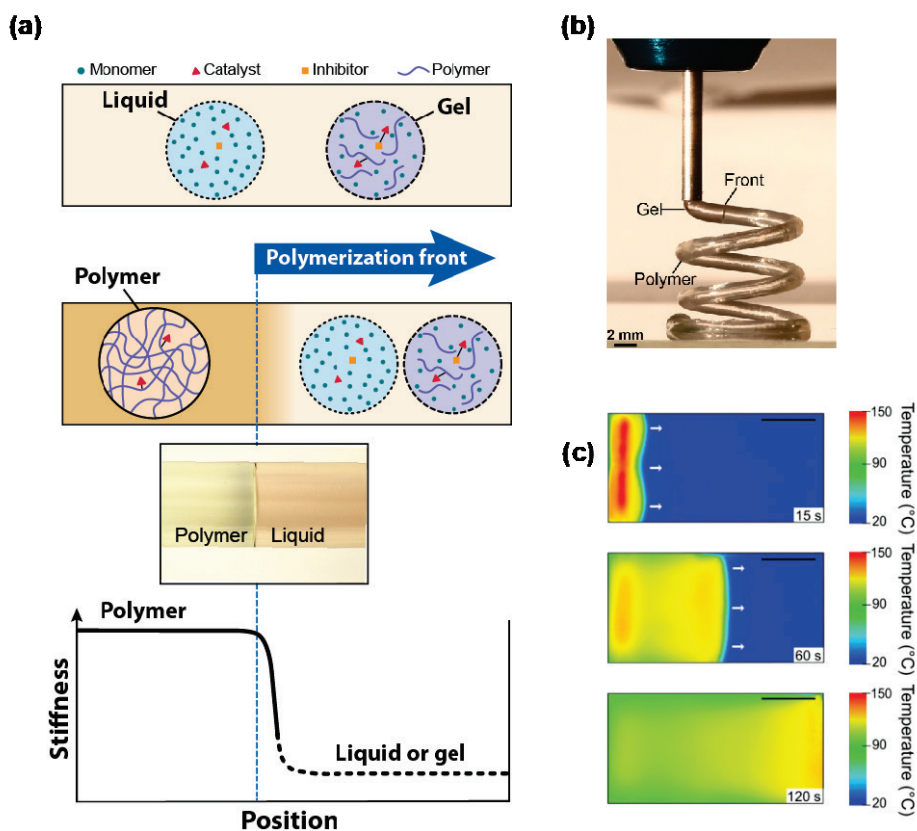


Figure 7. (a) Frontal polymerization uses a propagating thermal wave (formed and maintained by the exotherm of polymerization) to convert a mixture of monomer, catalyst, and inhibitor from a low-viscosity liquid or gel into a stiff solid polymer. (b) A prepolymer gel is extruded from a printing nozzle at the same rate at which the polymerization front propagates through the gel. Behind the front, a stiff self-supporting polymer is present. (c) Thermal imaging of a polymerization front passing through the prepolymer. Scale bar is 4 mm. Source: Adapted with permission from Nature Publishing Group: *Nature*, [Rapid energy-efficient manufacturing of polymers and composites via frontal polymerization](#), I.D. Robertson et al., 557, 223–227, 2018.

Classical electrochemical processing is widely practiced in industry, where the ability to electroplate thin metal films from aqueous solutions onto both planar and curved surfaces, or to electrochemically refine materials provides considerable value. More recently, it has been recognized that materials such as LiCoO_2 and silicon for battery cathodes and anodes, respectively,^{28,29} can be electrodeposited into precise forms, including those with tight control of crystallographic orientation, at temperatures far below typical solid-state processes used for such materials. Electrosynthesis of chemical compounds also is an area of

growing interest and importance due to the potential to reduce energy consumption and waste, as well as to access otherwise difficult to synthesize compounds.³⁰ In electrochemically driven systems, efficient use of electrons is paramount. Electrons not participating in the reaction of interest are both wasted, and can drive undesirable side reactions. There remains a need to generalize these concepts in atom, electron, and energy efficient routes to broad classes of precise materials.

Thrust 2. Synthesis of hierarchical structures across multiple scales, in complex organizations, and at interfaces with targeted functions

Modern molecular and material systems are complex and are rarely composed of single building blocks because of the need to satisfy multiple, stringent, and sometimes competing performance demands. For example, many energy materials derive performance benefits from combining large surface areas, to maximize locations for desired transformations, with high atomic deposition rates to maximize the frequency at which reactants interact with the material. Biological materials encode both of these performance advantages through the use of structural hierarchy across length scales.^{31,32}

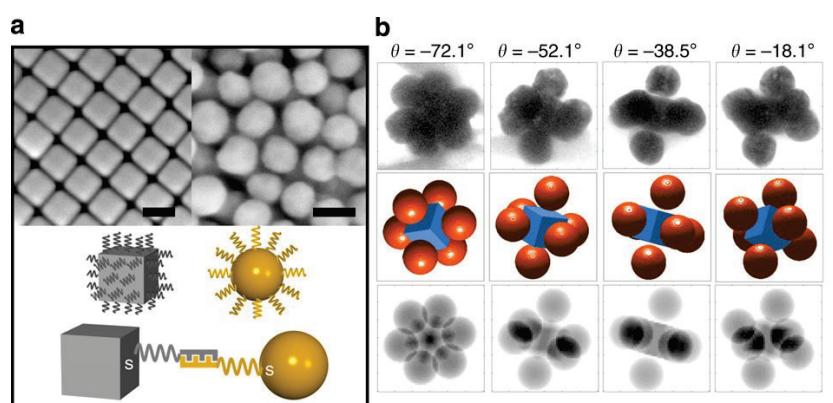


Figure 8. (a) (Top) SEM images of (left) cubes (46 nm in edge length) and (right) spherical nanoparticles (46 nm in diameter). (Bottom) Schematic of the DNA functionalization and assembly of cubes and spherical nanoparticles. (b) Comparison of (top) TEM images with (middle) 3D reconstruction models and (bottom) the projections of the modeled structures at a few selected tilt angles to reveal the 3D structure of the assembled clusters (left to right: -72.1° , -52.1° , -38.5° and -18.1°). Source: Reprinted with permission by Nature Publishing Group: *Nat. Commun.*, [Superlattices assembled through shape-induced directional binding](#), L. Fang et al. 6, 6912, 2015.

Important research directions underpinning the manufacture of advanced material systems include developing the synthesis and assembly methods that realize compositional complexity and hierarchical organization with exquisite levels of control of all relevant components (see Figure 8). This requires exploration, theoretically and experimentally, of the chemical and physical forces operable from the molecular to the macro scales and using them in developing suitable processes to define structures across different scales. In partnership with these efforts, research is required to understand the multiple material and scale

structure-function relationships to harness desirable, multiple functionalities. For example, in nanoparticle/polyelectrolyte composites, the percolated nanoparticle domains provide strength, while an independently percolated polymer microphase enables ion transport.³³ Beyond realizing additive functions in composites, mixed material systems open up avenues to couple different, often orthogonal properties to create “homogenized” materials with functions not found in bulk materials. These explorations are just beginning, but an early example is of superparticles assembled from a combination of plasmonic and magnetic nanoparticles that have coupled optical and magnetic properties.³⁴ To illustrate how such control leads to better performance, designed 3D nanoscale arrays have been used to form a chemically active material with an enzyme density that is increased by several orders of magnitude and has a simultaneous enhanced efficiency of the reaction cascade.³⁵ Associated research involves developing characterization methods that probe structure at different scales, and the often buried and high densities of interfaces and their correlation with their functions that may dominate overall system performance. Ultimately the

processes to prepare and assemble these structured materials need to be translated to meet manufacturing scales.

Scientific understanding is needed for the integration of materials into manufacturable device architectures to ensure process compatibility and function. For example, the directed assembly of block copolymers and nanoparticles in templates, described previously, allows the integration of different materials—such as emissive particles in nanophotonic cavities or the patterning of magnetic material for storage—to create devices. Can such processes be applied at manufacturing scale to much broader classes of energy-relevant materials, e.g., for energy storage? Sequential assembly requires orthogonal processes to engineer and differentially functionalize layers. While such processes have been demonstrated for microelectronics, their use in other engineering materials has been limited.

Thrust 3. Spatial and compositional control down to atomic scales in 0D, 1D, 2D, and 3D at manufacturing-relevant scales

Supported by the National Nanotechnology Initiative over the past 20 years, exploratory and open-ended fundamental research in the scientific community has led to both rational and serendipitous discovery, creating the foundation of our understanding of chemistry and physics at the nanoscale and enabling its translation to impact energy science and technology. Now we look to leverage this knowledge to design, synthesize, and fabricate materials and devices with atomic control at manufacturing scales. For example, self-limiting chemical reactions used to grow layers with atomic control in the vapor phase, heralded for allowing the fabrication of robust gate dielectric layers in highly scaled semiconductor devices, are being extended to solution-phase, atomic control of surfaces in chemically synthesized, low-dimensional materials for applications far from microelectronics, e.g. separation membranes.³⁶ Ion exchange interactions at the nanoscale (unlike in the bulk) are kinetically fast at room temperature, allowing rapid, solid-state chemical transformations. Atomic-scale control provides routes to dope nanostructures; create structural, compositional, and functional complexity; and realize metastable phases with single-nanometer critical dimensions. Can chemically specific and self-limiting reactions, and manipulation of the thermodynamic and kinetic landscape, be used to allow more precise spatial and compositional control of polymers and inorganic nanostructures and thus reduce dispersity in their structures and inhomogeneities in their functions even at the molecular level? Can these reactions be applied to a broader material set; and is it possible to maintain the precision, yet increase the speed of slow, sequential atomic layer processes to allow rapid growth of 3D and hierarchical structures?

Physical, mechanical, optical, electrical, and magnetic forces have enabled the manipulation of low-dimensional materials, even the manipulation of individual atoms. For example, the extension of single electrochemical devices to parallel arrays of scanning probes and actuators with three degrees of freedom is aimed at translating atom-level control on surfaces to manufacturing scales. Laser cooling and magnetic field gradients have been used to cool, confine, and trap isolated atoms and ions and are a leading platform for quantum computing.^{2,3} However, parallelization is similarly needed to scale to hundreds, thousands, and perhaps even millions of interconnected traps. These multiple forces are also the basis of operation of many energy technologies, including optomechanical and electromechanical devices and designs for smart windows, water treatment, and environmental sensors.

Thrust 4. Design rules and processes for manufacturing

To impact manufacturing, the synthesis of atomically and molecularly precise building blocks, components, and systems must be undertaken and must account for both function and manufacturing constraints. Successful design rules include, for example, (1) synthesizing materials and chemicals from abundant and Earth-friendly building blocks, (2) processing functional materials in large volumes without uncontrolled defects when needed for their end use, (3) positioning materials with both spatial precision and controlled orientation and morphology, and (4) simplifying chemical reaction pathways

with enhanced yields. Using such rules will enable manufacturing materials and molecules in which the flow of ions, molecules, electrons, photons, and phonons is controlled in unprecedented ways. Other manufacturing constraints related to performance metrics usually not associated in early stages of development—such as lifetime, life cycle, and reliability—enhance the probability of impact. Thus, these need to be addressed as design criteria from the beginning. Advancing these scientific thrusts will enable manufacturing processes that are fully optimized with respect to these constraints. Safety considerations are also important for manufacturing processes. For example, new materials for coating applications are infinitely more attractive if processing is water-based than organic solvent-based. In devices such as lithium-ion batteries, replacing flammable and toxic liquid electrolytes with solid electrolytes may offer distinct processing and recycling advantages. Incorporating manufacturing constraints into all aspects and all stages of scientific discovery, rather than treating these constraints as afterthoughts, promises to result in vastly improved technological outcomes for discoveries in the scalable synthesis and processing of precise atomic-scale building blocks for components and systems, and to shorten design cycles.

Thrust 5. Translating fundamental understanding of science of assembly across length scales

Research advances motivated by this PRD will provide fundamental understanding of how collective properties emerge and are structured across multiple length scales, and strategies for the purposeful design of emergent properties for technology. For example, heterogeneous superlattices constructed from functional nanocrystal building blocks (e.g., metallic, semiconductor, magnetic) have been shown to support collective phenomena when the internal architecture is appropriately controlled (see the sidebar). Assemblies of these and other component types have displayed coherent vibrational states,³⁷ tandem catalysis,^{38–40} enhanced magnetism,^{41,42} optical responses,^{43–45} and robust mechanical properties.⁴⁶ While there have been previous self-assembly demonstrations of binary^{47–49} and ternary⁵⁰ superlattices, the discovery of manufacturable methods for creating 3D lattices, across many length scales, with desired emergent properties, would be a very significant technological advance.

Science and Technology Impact

Improved models, theory, and simulation tools for manufacturing science. Advances in scalable synthesis and processing of materials building blocks, components, and systems with atomic and molecular precision will enable and be enabled by transformational developments of theory, simulation, predictive modeling, and materials databases of great relevance for manufacturing. Non-incremental extensions of ab initio theoretical frameworks require validation but often are tested on a limited number of systems. Predictive models often also depend on empirical parameters that are determined based on limited data. Access to larger and richer data sets, including observables at multiple length scales, will both increase the physical content of the models and better leverage machine learning approaches. While examples of harnessing reaction data to guide synthesis exist, opportunities remain to extend these models to larger length scales or heterogeneous materials systems. Improved chemistry- and physics-based models can lead to entirely new understanding of complex molecular and material syntheses in terms of their constituent building blocks. They also will accelerate and better integrate the theory-characterization-synthesis paradigm for rapid development of heterogeneous systems.

Realization of specific hierarchical, multi-functional structured materials and systems. Progress in the research directions outlined in this PRD will result in foundational knowledge that can be used to predict and then direct the synthesis and assembly of precise, often hierarchical and multicomponent material systems with explicitly targeted functionalities. To have the greatest impact on technology, next-generation materials not only must be precise but also likely must concurrently provide multiple functions, often with competing demands. For example, catalytic membranes must exhibit high porosity, high mechanical strength, chemical activity, and thermal stability. Materials of importance for both energy storage and separation processes often consist of composites containing separate ion and electron

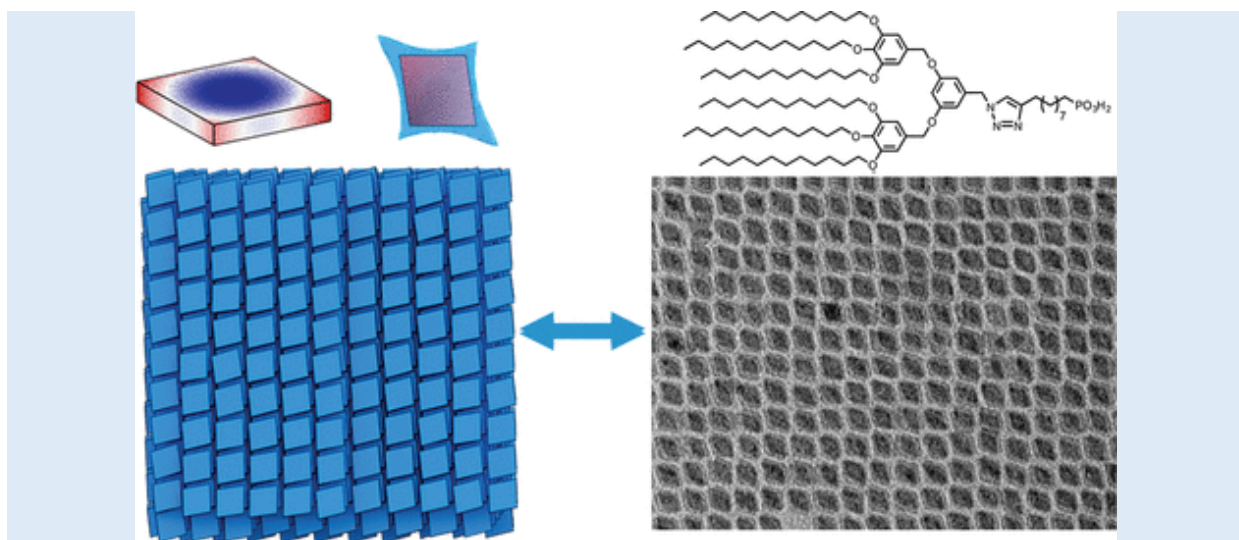
conducting components.⁵¹ In both these examples, material systems in which one material can provide high performance across multiple unrelated properties will lead to performance enhancements and potentially reduce manufacturing complexity. This PRD will inspire researchers to answer important questions of significant relevance to manufacturing, including these: What are the optimal synthesis and assembly methods to achieve specific and targeted structures? How can desired synthesis pathways to multiscale material organization be established and carried out? What is the relationship between material's multiscale architecture and its desired cross-functionality?

Sidebar: Colloidal Nanocrystal Superlattices

Colloidal nanocrystals are fragments of inorganic crystalline solids that have organic (i.e., molecular, DNA, dendrimeric, or polymeric) or inorganic ligands coordinating their surfaces. Wet-chemical methods provide exquisite control for synthesizing nanocrystals tailored in size from ~2 to 200 nm in diameter, which can be truly molecular for the smallest size species, or precise to within atomic roughness for larger nanocrystals. Synthesis routes also allow control of the shapes of nanocrystals (e.g., to form spheres, cubes, rods, and polygonal platelets) and internal structure (e.g., to introduce dopants and to achieve complex core-shell and Janus heterostructures). The library of nanocrystals includes elemental, binary, ternary, and quaternary compositions that can be metallic, semiconducting, insulating, magnetic, luminescent, and phase change materials. The ligands at the nanocrystal surface mediate their synthesis, providing size and shape control, and sterically or electrostatically stabilize the nanocrystals in solvents.

The precision synthesis of nanocrystals and their colloidal stability enable their self-assembly and directed self-assembly to form artificial superlattices. These structures can be crystalline, liquid crystalline, quasi-crystalline, or glassy. Many nanoscale superlattices adopt structural analogs of atomic solids (e.g., NaCl, MgZn₂, CaCu₅, and NaZn₁₃; AlMgB₄; nematic and smectic; and quasicrystalline AB₄, AB_{3.84}, AB_{7.7}) with examples of packings not previously observed in bulk atomic systems. The motif depends on the sizes and shapes of the nanocrystals, the ligand chemistry (e.g., charge, steric, bonding interactions) and surface coverage, and the chemical affinity of the nanocrystals to liquid or solid substrates upon which they are assembled.

The physical properties of superlattices can be tuned by the type (i.e., precise size, shape, and composition), number, and arrangement of nanocrystals and by the distance and composition of the medium between the nanocrystals. For example, by tailoring the length and composition of intervening ligands, the strength of quantum mechanical coupling can be tuned, leading to the emergence of unique physical properties (e.g., optical, electrical, magnetic, mechanical, thermal). Tailoring the ligand and thus the distance between metal nanocrystals can cause the nanocrystal assemblies to undergo an insulator-to-metal transition and allow tuning of optical dielectric functions. Selection of the ligand chemistry on semiconductor nanocrystals has been used to tailor the energies, types, concentrations, and lifetimes of carriers in the construction of efficient solar photovoltaic devices. Combinations of nanocrystals with different or orthogonal physical properties promise the design of materials with exotic or multiple functionalities to achieve, for example, adaptive materials that transduce stimuli-responses in multiple domains.



Simulation and experimental transmission electron microscope (TEM) image of a multi-layer assembly of GaF₃:Yb/Er rhombic nanoplates capped with dendrimeric ligands. The bulky dendrimeric ligands create coronas around the plates and create a directionally offset architecture in the multi-layer assemblies. The rhombic plates in the TEM image (right) have dimensions of 16.6 and 19.8 nm on their short and long sides, respectively. Source: Reprinted with permission from K. C. Elbert et al. *ACS Nano* 13, 14241–14251, 2019. Copyright 2019 American Chemical Society.

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PRD 2: Integrate Multiscale Models and Tools to Enable Adaptive Control of Manufacturing Processes

Summary

With the advent of new, advanced digital manufacturing technology, the need to develop models that accurately describe manufacturing processes, product performance, and materials is pressing. Reliable, computationally non-taxing physical models are essential to enable rapid advancement toward science-based manufacturing. Increasingly powerful computational resources are now available and can be leveraged toward this goal by linking models across length and time scales as shown in Figure 9. The current manufacturing enterprise in the United States relies upon highly skilled and experienced operators, for whom controlling the manufacturing process can be more of an art than a science. A combination of multiscale modeling capabilities, in situ diagnostics, online decision-making frameworks, and operator experience is needed to realize adaptive, automated manufacturing processes with built-in component qualification.

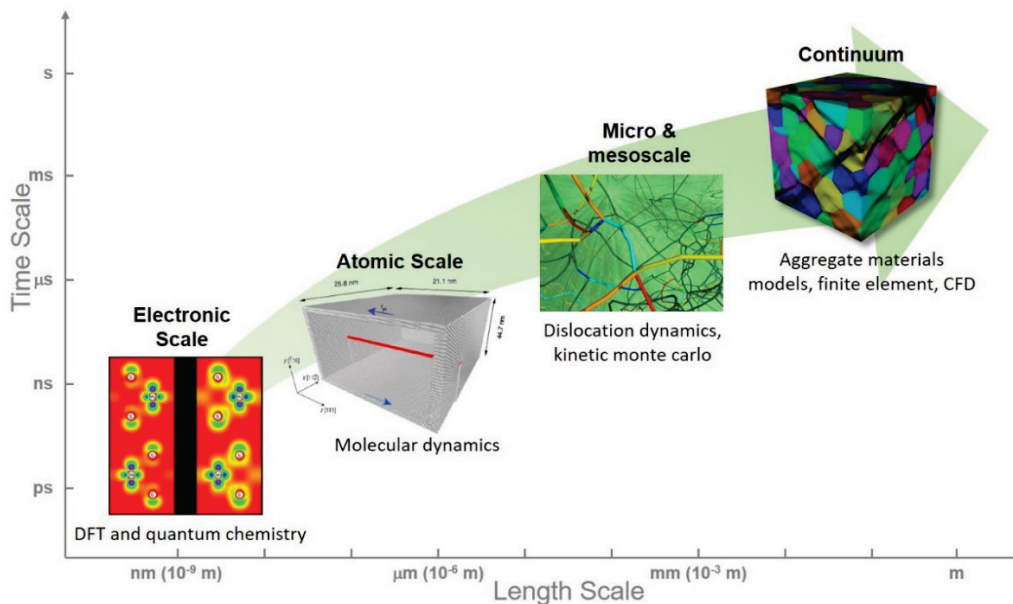


Figure 9. Multiscale modeling consists of linking models across orders of magnitude on temporal and spatial scales. *Source:* Image provided by Lawrence Livermore National Laboratory.

Several fundamental scientific advances are needed to achieve this vision:

1. Multiscale physics-based models that can capture and predict the parameter space of specific manufacturing processes and material characteristics during fabrication.
2. Fast and accurate in situ diagnostics and characterization tools with high spatial and temporal resolution.
3. In-process decision making frameworks that are informed by multiscale models in real time, and in situ diagnostics and use of data science techniques to enable adaptive manufacturing processes.

The state of the art in modeling and simulations of manufacturing processes relies upon high-performance computing (HPC) resources to capture the multiphysics, multiscale, and dynamic environment of many emerging processes. Great strides have been made to date in, for example, modeling laser-matter interactions and melt-pool dynamics for selective laser melting (SLM) in metal powder-bed additive manufacturing (AM) shown in Figure 10. Since these high-fidelity simulation examples require massive computing resources and exceedingly long execution times, they are often impractical for manufacturing. Multiple models with ambiguous input/output compatibilities are often relied upon to span the required size scales.¹⁻³ Coarse models that are faster and more practical have been developed at the expense of inaccurate physics and/or unrealistic boundary conditions.

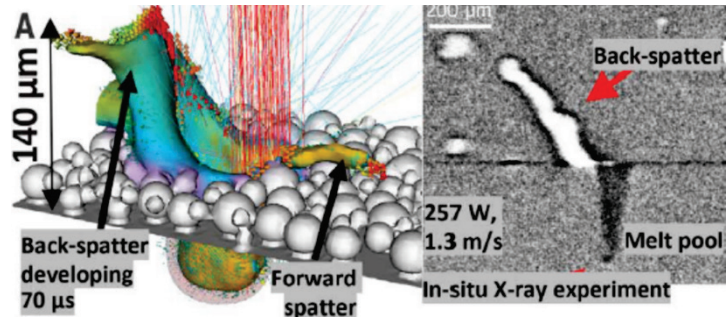


Figure 10. (a) A model of a laser interacting with metal powder to create a melt pool in SLM processing. This HPC simulation includes ray tracing of the laser, phase changing and consolidation of the metal particles, and the dynamics of fluid flow. (b) Data from an in situ x-ray experiment of a SLM system showing the melt pool and back-spatter of material, qualitatively similar to the model. Source: From [S.A. Khairallah et al. Science 368\(6491\), 660-665, 2020.](#)

Modeling and simulation have provided significant advancement in the scientific understanding of AM polymeric systems. Atomistic models and molecular dynamics simulations use ensembles of the most basic possible constituents to understand the fundamental behavior of these materials. These models require enormous computational costs, limiting their output to irrelevantly small time and spatial scales. Coarser models and stochastically based simulations, such as dissipative particle dynamics (DPD), can provide information over tens of microseconds and hundreds of nanometers, such as a simulation of spider silk in tension, shown in Figure 11.⁴ They represent an improvement over atomistic methods, leaving a significant challenge for any model or combination of models to span the manufacturing-relevant time and length scales, up to the continuum regime at which most production systems operate. Typically, these costly and complex models only inform the macroscale process simulations, rather than explicitly couple to them; the latter would enable more realistic representations. Developing and using simulations that can describe complex processes and materials is crucial for understanding the fundamental underlying science, but moving toward efficient lower-order models for in-process control is a long-standing goal.

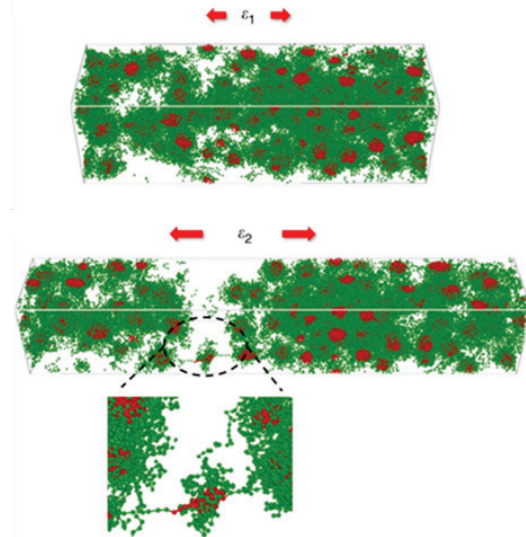


Figure 11. An example of a DPD simulation showing spider silk at various tensile strains and ultimately failing. Green represents amorphous structure (β_1 -helices and β -turns), and red corresponds to crystalline regions. Source: [L. Pan et al. Nat. Commun. 11, 1332, 2020.](#) Licensed under a [Creative Commons Attribution 4.0 International License.](#)

Modeling and simulation alone cannot result in science-based manufacturing. Measurements of process parameters, characterization of materials in real time, empirical data, and in situ diagnostic capabilities are also essential. To exemplify this reality, an image of the melt pool and back-spatter of particles in an SLM process, shown in Figure 10 (right hand side), was generated using a special in situ x-ray setup. It

required the construction of a custom-designed, simplified SLM system that was installed in a large-scale, synchrotron beamline; it does not even represent a functioning manufacturing system. These types of experiments are important for the validation of models and to gain intuitive understanding of the underlying physics; but ultimately, on-machine diagnostics are needed to realize the vision of online decision-making frameworks in manufacturing systems.

Statistical process control and monitoring of machine stability are currently possible within some manufacturing systems. Direct observations and in-line measurements of the actual components being fabricated remain limited, especially those capable of real-time defect detection during manufacturing.^{5,6} The processes equipped with in situ diagnostics for material and component observation are typically restricted to visible and infrared cameras with marginal spatial resolution and inadequate speed, and do not benefit from in-process decision making. Most of the detailed characterization and metrology are conducted post-fabrication and typically are time consuming, are costly if conducted nondestructively, or cause sample damage otherwise.

Applying data science techniques to fabrication processes will facilitate synergy between modeling and simulations and in situ diagnostics, which enable *adaptive* manufacturing, capable of in-process decision making. It will allow, for the first time, the fabrication of materials, components, and chemicals that are qualified upon manufacturing completion, thus reducing and eventually eliminating the need for post-fabrication inspection, metrology, and characterization. Machine learning algorithms and other data fusion methods are needed to analyze and to synthesize the potentially enormous and orthogonal data streams to understand the genesis of defects and to mitigate or eliminate these defects on-the-fly through intelligent process parameter changes.

The impact of an adaptive, automated, and intelligent manufacturing ecosystem will be extraordinary. The ability to adjust process parameters on-the-fly in response to built-in in-process intelligence will result in the high-throughput production of qualified components at significant cost and time savings that stem from the elimination of post-fabrication characterization and inspection. It also will bring the vision of “first time right” manufacturing and near 100% production yield to reality.

This PRD seeks to address the following key questions: *What are the frameworks required to model, monitor, and ultimately control manufacturing processes that tightly couple physics and chemistry across scales? How can complex multiscale models be translated to fast surrogate models for process control?*

Scientific Challenges

A primary challenge is that advanced manufacturing processes span multiple scales and multiple physical phenomena that must be coupled to effectively simulate their emergent complex and nonlinear behavior. Today’s physics-based models involve sophisticated numerical procedures, with arrays of interdependent equations and variables that naturally lead to large-scale, computationally intensive modeling. Gaining insights into parameter sensitivity and into uncertainty propagation through different physical regimes and length scales is crucial; current approaches rely on modeling everything to the highest fidelity, yet are unable to deterministically capture all the details. Ultimately, fast and accurate surrogate models are needed to impact material and component manufacturing in-process and understand the underlying physics, with highly refined large-scale models being used to develop these surrogates. In-depth understanding of the manufacturing process sufficient to generate reduced-order, fast, surrogate models also remains a barrier to realizing this vision.

The drive to achieve adaptive control and in-process decision making necessitates having the ability to conduct in situ measurements, such as geometric tolerance, temperature, defect detection and distinction, chemical composition, and more. The relevant length and time scales associated with these measurements

are process specific and can be challenging to achieve. For example, in metal powder-bed AM, extreme temperatures in excess of 3000° C and thermal ramp rates approaching 10,000° C/sec are not uncommon. In addition, the laser spot size is typically ~50 μm, and the corresponding melt pool can be as small as 100–200 μm across. The melt pool also can be moving at tens of millimeters per second; and the consolidated part's feature size can range from ~200 μm to centimeters with a surface roughness of <50 μm, all of which are relevant quality metrics. Micro-scale defects, such as voids and pores with diameters of 5–10 μm, can substantially deteriorate material strength and integrity. Some aspects of the laser-induced melting processes can be observed in situ now, as shown in Figure 10, at the cost of building a simplified manufacturing system inside of a large synchrotron beamline; however, such a system would not be broadly deployable and could not be integrated into an in-process decision making framework. Other manufacturing processes and materials have their own specific size, timescale, and other requirements for in situ diagnostics.

Another challenge associated with achieving adaptive control of manufacturing processes is data fusion and in-process decision making. It is likely that multiple, orthogonal sensing modalities will be needed to understand the emerging manufacturing processes; and given the necessary speed and resolution, the generated amounts of data have the risk of being extremely large. How to synthesize and mine these data sets in an efficient way to generate actionable decisions currently is largely unknown. It is not even known which measurements are most relevant and what combinations of parameters may reveal defects or predict upcoming manufacturing errors. While machine learning and other data science methods are advancing rapidly, how to apply these techniques to emerging manufacturing systems and materials remains an open question. Figure 12 shows an example of a simple and early-stage demonstration of this concept, in which image analysis and high-speed video capture were used to train a machine learning algorithm for process control of a microfluidic assembly process.⁷ Combining the large data sets generated continuously through the process with the in-process models to alter the process parameters based on geometry of components is a grand challenge.

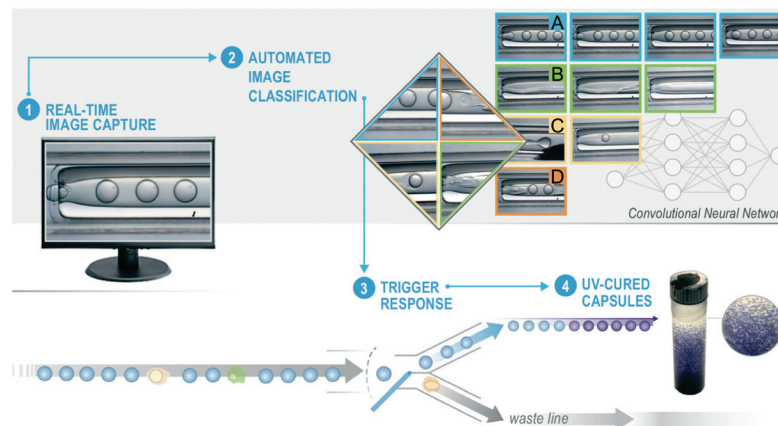


Figure 12. An example of an automated microfluidic assembly sorting system. (1) A digital camera captures images of microencapsulation events in real time. (2) An image classification algorithm assesses whether microencapsulation is in the desired fluid dynamic dripping state (A) or the undesired wetting (B), jetting (C), or rupturing (D) states. (3) The predicted class of microencapsulation informs a valving system that sorts desired and undesired events. (4) Sorted final microcapsules and non-dripping events accumulate in collection and rejection jars, respectively. Source: [A. Chu et al. *Lab on a Chip* 19\(10\), 1808-1817, 2019.](#)

Research Thrusts

Thrust 1. Validated multiscale, multiphysics models

To better understand the existing and emerging manufacturing processes, detailed and accurate process models are required. These multiscale, multiphysics models must be computationally efficient and physically sound. Most process models used today fall into two categories: (1) computationally practical models that do not accurately represent the physics owing to assumptions and simplifications incorporated into the model to reduce computational cost and (2) high-fidelity models that accurately capture the physics but are so computationally intensive that they require HPC resources and significant time (days to weeks) to converge to a solution. To truly understand the physics of the relevant processes, a detailed model spanning multiple length scales is needed, and HPC codes and hardware are required. These simulations are too expensive to be used in an in-process adaptive control scheme; instead, they can inform surrogate models and guide relevant diagnostics and measurements because they elucidate the relevant physics. Even the existing most-complex process models have difficulty spanning size scales from molecular levels to the macroscale. Synthesis and processing are closely coupled to the actual manufacturing process in today's advanced fabrication methods. The difficulty of spanning these scales while combining normally disparate physics remains to be overcome, and any models that could do so would constitute a significant advancement in the understanding of manufacturing.

Thrust 2. High-speed, in situ diagnostics and characterization tools

Diagnostic analysis and characterization for synthesized molecules, fabricated materials, and full components from emerging and advanced manufacturing processes are critical for scientific understanding and model validation. This thrust focuses on specific needs for high-speed, in situ tools to enable adaptive control in manufacturing; additional needs for characterization tools are laid out in PRD 3. To realize the vision of adaptive and intelligent on-the-fly control, more sophisticated and advanced diagnostic tools are required. Sensors that can accurately conduct in-process metrology, temperature measurement, chemical analysis, and other measurement modalities are required. These measurements must be time-efficient, on the order of the smallest timescales associated with the process itself, and must be analyzed similarly fast. Additionally, these sensors, diagnostics, and characterization tools must have excellent spatial resolution, on the order of the smallest relevant length scales of a given manufacturing process, i.e., microns or even hundreds of nanometers. The acquired data must be stored, moved, synthesized, and analyzed at sufficiently high rates to allow for corrective action by the intelligent manufacturing system, which is enabled by these diagnostics coupled with data science algorithms and fast surrogate models.

Thrust 3. Data processing and fusion

Data analysis and advanced modeling represent key steps in enabling in situ evaluation and qualification, i.e., interpretation of the measured data and using the findings to develop useful databases. Advanced manufacturing is a complex multidimensional process, so the data needed to evaluate the quality of the output cover multiple length scales and different phases of the material. Therefore, the measured data would be obtained from a variety of sensors and as multimodal signals. For online monitoring of advanced manufacturing, one important area of development is the fusion of the process-, chemistry-, and material-specific data that are heterogeneous and multidimensional.

For example, the genesis of materials created through many advanced manufacturing methods is that they are derived from a *photoinitiation-based chemical reaction*, with post-processing that requires chemical synthesis and photoresist development. Of particular importance is the need to recognize the significant deviations in the (commonly overlooked) attainable chemical compositions and microstructures of additively manufactured materials compared with those of the same materials that are conventionally produced; the importance of developing a deep understanding of the parameter space of chemically

produced materials cannot be overstated. Vat photopolymerization, a subset of AM techniques that achieves spatial patterning via photopolymerization of a liquid photopolymer in a vat, stands out because of its high print resolution and rapid fabricate speeds.^{8–12} Incorporating dynamic covalent chemistries^{13,14} or supramolecular chemistries¹³ enables the design and fabrication of self-healing materials, ceramics, and—less commonly—metals by placing them in the framework of an “in situ chemical reactor,” as shown in Figure 13.

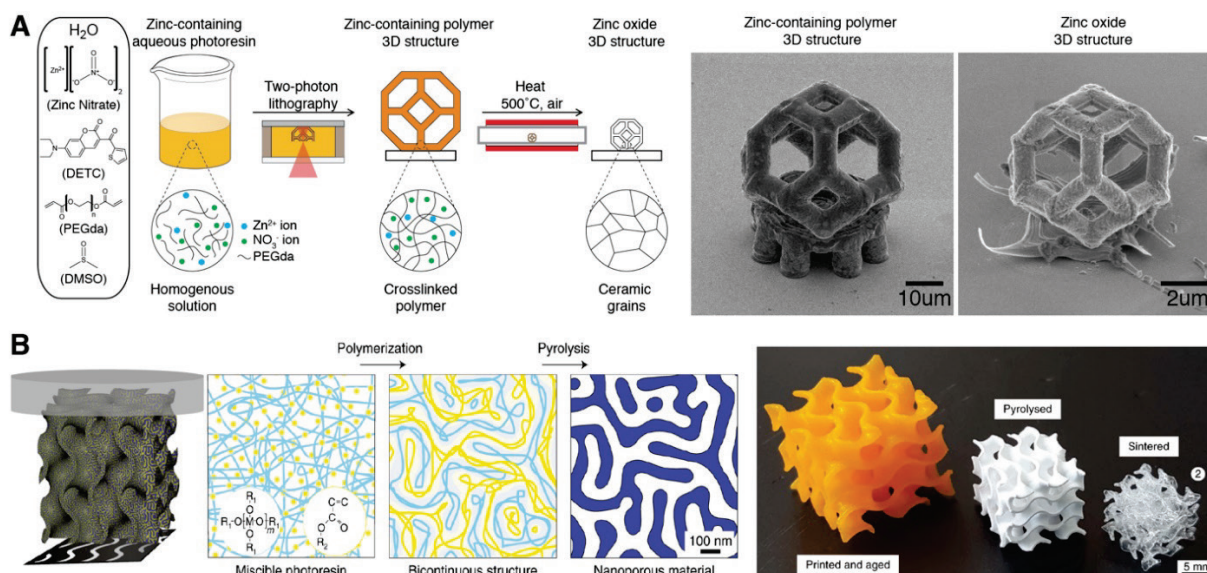


Figure 13. “Chemical reactor” framework for ceramic vat photopolymerization. (A) Zinc oxide microstructures fabricated via the use of zinc nitrate containing aqueous photoresins with two-photon lithography. The use of metal nitrates enabled the solution combustion synthesis of metal oxides with the polymer binder acting as the combustion fuel. Source: Adapted with permission from [D. W. Yee et al. Adv. Mater. 31\(33\), 1901345, 2019](#), © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.¹⁵ (B) Photopolymerization-induced phase separation during digital light processing printing results in a bicontinuous structure of polymer and metal alkoxides. Thermal treatment of the 3D printed object results in decomposition of the polymer and condensation of the metal alkoxides to form glass. Source: Adapted with permission by Nature Publishing Group: *Nat. Mater.*, [Three-dimensional printing of multicomponent glasses using phase-separating resins](#), D.G. Moore et al., 19(2), 212–217, 2020.¹⁶

The breadth of chemistries and chemical reactions, as well as concern for defects, drive another important characteristic for in situ evaluation, practical computational cost. Computational hardware requirements and numerical procedures need to be sufficiently affordable to enable online monitoring and decision making while accurately capturing properties of a broad range of chemistries. New computational infrastructure, informatics, algorithms, and numerical analysis are needed to enable the data processing and fusion.

Thrust 4. Fast, predictive models

The current state-of-the-art models are beginning to predict advanced manufacturing and processing physics; they typically require using clusters of HPC resources over many days or weeks. To enable in situ evaluation and redesign of the process and/or a component, reliance on a fast, predictive model in real time is critical. A common approach to understand and predict a system that is not well understood in a time-sensitive application is machine learning. Machine learning typically requires large-scale data sets to train the model offline; and given the range of variabilities and parameters, obtaining the relevant large set of data may not be tractable. It is imperative to invent new methods to develop fast, predictive real-time models for advanced manufacturing, which may leverage heterogeneous data, physics-based models,

machine learning, model fusion, and uncertainty propagation. One way to reduce the modeling cost is to consider only the parameters of interest and importance, which are guided by the in-process decisions. The purpose and application of the model outcome can inform the required degree of accuracy and level of coupling and fidelity, and such intelligent modeling can substantially reduce the required computing resources and cost.

Thrust 5. Adaptive control and decision making

The combination of real-time data acquisition and advanced modeling will enable component quality evaluation during manufacturing. The ultimate goal is to create an adaptive, intelligent process based on the in-process observed data and fast models. This adaptive decision making and process control will be capable of implementing self-directed design and/or process modification during manufacturing. The fundamental concept is to evaluate the quality of a manufactured material, chemical, and component in real time and to continuously adapt to modify the process parameters or designed geometry. As manufacturing, design, and evaluation take place simultaneously and evolve adaptively, advanced manufacturing will take one step closer to the paradigm of “born qualified,” so that the final part is ready and safe to use immediately upon completion. This approach also automatically generates a digital twin that can be employed throughout the life cycle of the part to understand aging and other long-term performance. This will substantially reduce the lead time and reliably certify the resulting products.

Science and Technology Impact

Through the potential scientific breakthroughs elucidated by this PRD, the vision of a science-based manufacturing enterprise in the United States can be realized. Core to this concept is the ability to enable first-time-right manufacturing via a manufacturing framework that includes adaptive control and in-process decision making. This framework would be enabled by a combination of complex multiscale, multiphysics models, fast surrogate models, advanced in situ diagnostics, and algorithms including machine learning methods to synthesize data and make decisions about process parameters and/or component redesign during the manufacturing process.

The payoff from realizing this vision will be improved chemical processes and qualified materials and components that possess optimal properties at the first fabrication and every time thereafter. This payoff will allow for US industry to approach the long-standing goal of near 100% yield on produced parts while reducing the need for and cost associated with the design-fabricate-test iteration, as well as tooling expense and part inspection cost. Similarly, energy consumption during the manufacturing process could more easily be predicted and optimized, further reducing cost. The economic benefit of this concept would be enormous and is difficult to quantify.

With the successful implementation of this vision, new, more complicated designs with extreme tolerances can be physically realized. As advanced design methods emerge, such as topology optimization and statistical exploration of design spaces, higher-performance but more exotic and difficult-to-manufacture structures and components can be conceived. Currently, these designs push manufacturing processes to their limit and often cannot be pursued even in specialized research laboratories. If the framework proposed herein is implemented, these much more challenging components may become commonplace. In addition, it will be possible to know a priori if a structure is manufacturable; and this information can be fed back to the design algorithm for consideration and redesign, eliminating failed builds and detailed postmortem characterization and analysis.

Qualified, on-demand manufacturing of high-value chemicals, materials, and components with advanced designs will benefit nearly all industrial sectors. In the energy sector alone, the impact will be transformative for technologies that include chemical reactors, catalysts, batteries and energy storage devices, photovoltaic systems, metamaterials, and transportation systems. For example, many energy

transport and conversion systems could be redesigned to leverage geometric complexity and new materials enabled by this concept. The cost of manufacturing development also will be radically reduced. As one example, lightweight, energy-absorbing, and thermally and mechanically robust materials are important for fuel-efficient transportation, ranging from ground vehicles to aircraft and spacecraft. Figure 14 shows a possible example of an ultra-lightweight and ultra-stiff architected material that could move from boutique, lab-scale fabrication to truly manufacturable components.¹⁷ This new manufacturing paradigm would open new avenues for creating next-generation chemicals, materials, and systems across countless industrial sectors.

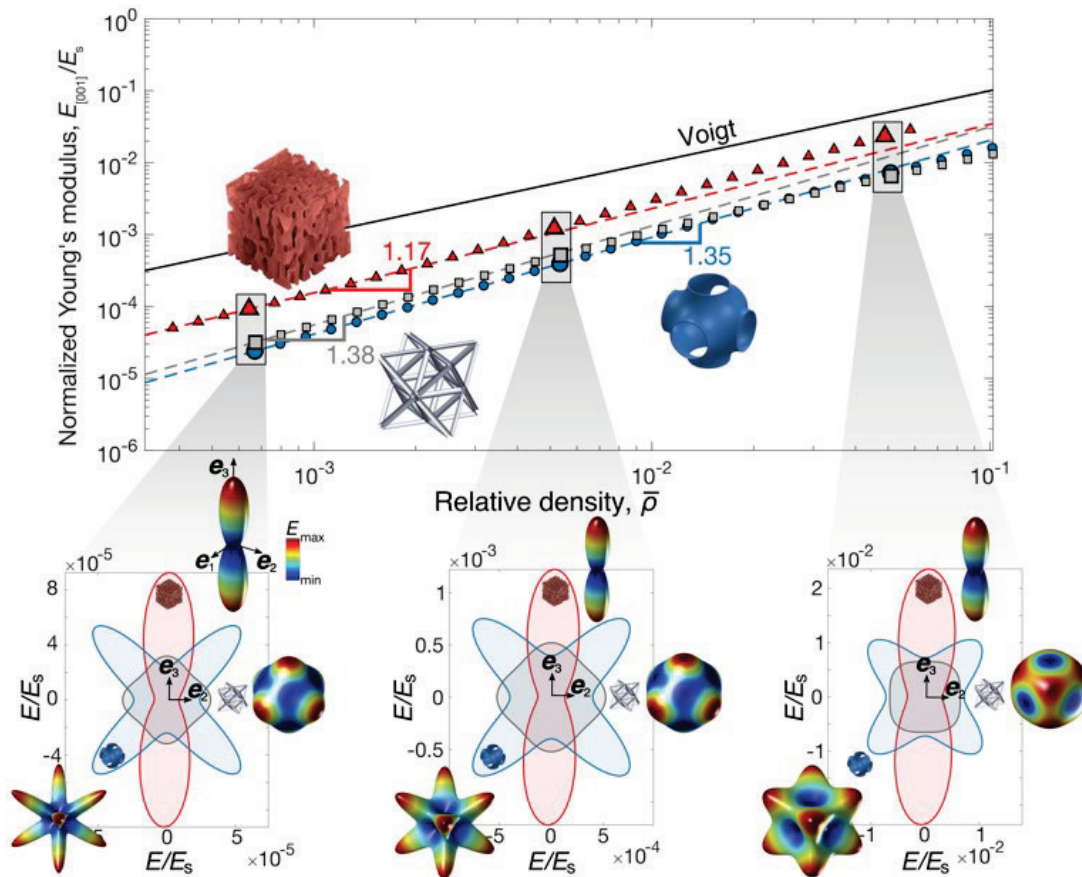


Figure 14. Stiffness scaling and elastic surface variations. Normalized Young's modulus $E_{[001]}/E_s$ as a function of relative density $\bar{\rho}$ for bicontinuous columnar (red), Schwarz primitive triply periodic minimal surface (TPMS; blue), and hollow octet (gray) architectures. Regression fits for the ten lowest relative densities from each architecture are depicted as dashed lines, assuming a relation $E_{[001]}/E_s = C \bar{\rho}^n$, with the corresponding scaling exponent—shown next to each fit. The elastic surfaces for each structure, along with their 2D projections, are shown for three selected relative densities. Close-to-constant anisotropy is observed for the bicontinuous architecture as opposed to the TPMS and octet geometries. Source: C. M. Portela et al. *Proc. Nat. Acad. Sci.* 117(11), [5686–5693](#), 2020. Used by permission.

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PRD 3: Unravel the Fundamentals of Manufacturing Processes Through Innovations in Operando Characterization

Summary

Many manufacturing processes are still practiced as empirical art rather than science, which poses a significant barrier to achieving resiliency, minimizing energy consumption, and innovating new technologies. Frequently, there is insufficient fundamental understanding to tailor and control chemical processes and materials manufacturing so that they perform exactly as desired. In situ characterization (observation of processes under conditions that replicate real-world and real-time conditions) and *operando* characterization (direct visualization and characterization of processes in real time) will provide the crucial knowledge needed to transform the science of manufacturing. To provide critical understanding, state-of-the-art multi-model characterization capabilities, as well as novel inline sensors, are needed to understand chemical and material manufacturing processes across length and time scales, to achieve synthesis control starting at the atomic level, to understand device–component interactions, and to develop appropriate surrogates.

The creation of innovative data infrastructures for in situ and operando characterization will help unleash the full potential of artificial intelligence and machine learning (AI/ML) to translate large data sets into predictive decision-making tools and will have a long-lasting impact on manufacturing. The overarching objective is to better align basic research with the technological needs of manufacturing industries, promote stronger ties between basic and applied research, and cultivate the next generation of scientists and engineers. New research efforts will provide opportunities for close collaboration between researchers in national laboratories, academia, and industry. The transformative effects are likely to arrive both in direct form, i.e., the characterization yields new information on the processes directly, and indirectly, e.g., characterization reveals new possibilities for manipulating the phases present in a process or the microstructure of a material.

This PRD seeks to address key questions: *How can manufacturing processes and products be “visualized” at microscopic to atomic scales, in real time, and under operating conditions to reveal the intricate details of underlying physical or chemical events? How can these insights be used in control schemes for decision making?*

Scientific Challenges

The past decade has seen dramatic progresses in characterization capabilities, most notably the advent of high-brilliance, high-energy synchrotron x-rays and high-flux neutrons. It has become feasible to study materials and chemical manufacturing processes at manufacturing scale in situ and operando. Many research areas, ranging from metal additive manufacturing¹ (AM) to soft matter² and Li-ion batteries,³ are developing or harnessing these capabilities. Two areas particularly important to the chemical industry that could benefit from operando characterization are catalysis,⁴ as illustrated in Figure 15, and separation sciences, according to a recent report of National Academies of Science, Engineering and Medicine.⁵

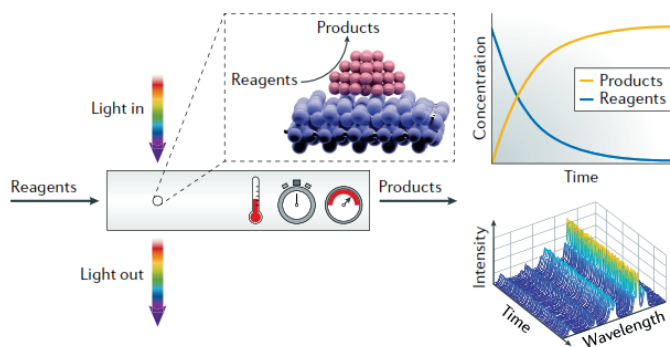


Figure 15. Solid catalysts are characterized in a miniature chamber during operation at the required temperature and pressure. The starting reagents and products formed during the catalytic reaction are recorded. Light from the infrared to the x-ray regime is used as a nondestructive probe to obtain spatially and temporally resolved microspectroscopy data. Source: Reprinted by permission from Nature Publishing Group: *Nat. Rev. Mater.*, [Spatial and temporal exploration of heterogeneous catalysts with synchrotron radiation](#), M. Florian and B.M. Weckhuysen, 3, 324, 2018.

Three challenges for this PRD must be addressed to enable in situ and operando characterization for transformative manufacturing, including developing new tools for unraveling complex manufacturing processes across multiple time and length scales.

The *first scientific challenge* is how to probe microstructural responses and changes at local scale and under process conditions. Some of the hardest problems in the manufacturing industry, for example, include alloy mechanical behaviors related to damage and failure from fatigue, stress, corrosion, cracking, or creep, which could be related to the local chemical environment. The ability to predict what happens as corrosion starts, cracks form, and damage accumulates, exceeds our ability to measure it. For example, what if we could see how hydrogen atoms move around dislocations and crack tips and influence ductility? What if we could visualize the motions of point defects under realistic irradiation conditions and quantify the efficacy of different types of sinks for defects? What if we could quantify strain in thin film structures, say for magnetic devices and energy conversion devices, as they are being grown (deposited)? What if we could map transformation strain with sufficient spatial and temporal resolution to capture diffusionless transformations?⁶ What if we could map out corrosion reactions under various loading conditions at or near crack tips that are buried inside bulk materials?⁷ At present, all these open-ended questions can be answered only partially because of limited spatial and temporal resolution and chemical sensitivity of available in situ and operando characterization tools.

The *second scientific challenge* is how to interrogate manufacturing processes operando through the intelligent creation of surrogates that contain the essential ingredients at production scale but are feasible for basic research in laboratory settings. Another way to articulate this challenge is how to convert large-scale engineering problems into manageable science problems. Many laboratory-scale reactors are used for discovering new catalysts and synthesis routes, and various mechanical or physical tests are normal practices to derive key materials properties in a simplified but well-controlled environment. If we can incorporate operando characterization with these miniaturized reactors and mechanical or physical testing devices, allowing simultaneous and multi-modal characterization, we should be able to accelerate materials discovery or process optimization. What if we could follow the change of oxidation state of a catalyst and the reaction kinetics and product yields for a production process simultaneously. What if we could interrogate the microstructural evolution of a material while it is being tested for strength or fatigue in real time, allowing unambiguous correlation of structure and property? And what if ML could be integrated into this approach and tell us in real time how to change temperature, pressure, or flow rate to maximize yields? The motivation is to create a new, integrated approach of discovery toward maximum efficiency built upon recent advances in characterization, modeling, and machine learning.

The *third scientific challenge* lies in achieving a seamless transition between basic and applied research and determining how to engage manufacturers to create an ecosystem spanning from fundamental understanding to delivering manufacturing *solutions*, as aided by *advanced computing and artificial intelligence*. Diffusion, for example, is critical to a variety of manufacturing processes, including catalysis, separations, phase transformations, thermo-mechanical processing of materials, and transport processes. How could AI/ML be used in combination with characterization at atomic resolution to understand how to measure and control diffusion, not only in academic settings but also in applications in manufacturing processes? Many innovative ideas show promise in simplified model systems but fail to make a meaningful impact on real-world problems because current research is focused exclusively around either basic research or applied research. The weak link can be bridging the two. For example, an imaging technique can work well for a two-phase system such as Al-Cu⁸ but needs to be adapted to more industry-relevant materials for which different contrast mechanisms are required. A desirable outcome would be to bridge the gap between discovery-driven basic research and purpose- or solution-oriented applied research directed toward transformative manufacturing.

Research Thrusts

Thrust 1. In situ characterization under real-world conditions

Engineering materials often must work and chemical manufacturing is often performed under demanding temperature, stress, and other harsh conditions. The objectives of this thrust are to characterize processes and materials in situ: that is to visualize microstructures and to use novel online sensors and analytical tools to follow reactions under flow, catalytic and separation processes, and interfacial phenomena in environments that mimic real-world conditions. For example, in situ methods are needed to understand catalyst deactivation mechanisms to ultimately reduce chemical manufacturing costs and improve efficiency. The complexity of current and future catalyst systems, particularly hybrids, presents formidable challenges in understanding the basic foundations for their performance in response to temperature; concentration; or transients in pressure, current, or voltage that perturb the state of a chemical reaction and dictate its course. We currently lack the means to directly connect local atomic information gleaned from in situ (and operando, discussed in Thrust 2) characterization of primary sites of energy conversion and catalyst activity to both (1) rate constants for elementary processes and ultimately (2) macroscopic system performance, including Faradaic efficiency, selectivity, and turnover. In addition, achieving sustainability requires inline characterization in the areas of post-use plastics and facile separation of plastics, as well as waste treatment and upcycling. Novel tools like single-entity electrochemistry⁹—an emerging, inexpensive, and sensitive method—could be used to observe individual particles, particularly in solution, in systems with natural heterogeneities. In situ characterization can aid understanding of the effects of impurities to reduce the use of ultra-pure reagents in manufacturing for cost reduction. Generally speaking, actual manufacturing processes are often associated with high data rates and demand more sophisticated inline data processing capability and innovative sampling techniques to extract useful information in real time. The following examples are intended to set the scene for a more creative approach to manufacturing-relevant science.

An experimental apparatus that creates an environment mimicking real-world conditions is the prerequisite for in situ characterization. The sample environments currently available are often limited to single variables, such as temperature, and to samples not large enough to represent device performance or manufacturing conditions. For example, recent reports point to the importance of novel catalysts for breaking down (upcycling) consumer plastics into reusable low-molecular-weight products, based on nanoparticles.¹⁰ These products rely on multiple characterization methods, ranging from basic molecular weight measurement, to ¹³C magic-angle-spinning (MAS) solid state nuclear magnetic resonance (ssNMR) spectroscopy, to electron microscopy of the catalyst microstructures. Upscaling such a process from the benchtop to a commercial reactor is highly likely to require new approaches to sampling for true operando characterization. Multi-modal in situ characterization also is required for materials

manufacturing to connect processing, structure, and properties in integrated experiments and eliminate ambiguity when they are measured separately. For example, Persson et al. used in situ x-ray scattering, including pole figure and optical reflectance measurements, during blade coating of semiconducting polymers, in which strong fibril alignment led to anisotropy in carrier mobility as a function of blade speed¹¹ (Figure 16). The Air Force Research Laboratory has designed a sophisticated load frame for combined mechanical testing, synchrotron diffraction, and tomographic imaging. Combining such a load frame with high-energy diffraction microscopy (HEDM) (Figure 17) can enable mapping of the 3D grain structure around a crack,¹² for example, and the relationship between the advancing defects and the microstructure can be investigated through machine learning.¹³ More generally, expansion to multiple variables and incorporation of ML tools would yield important new science.

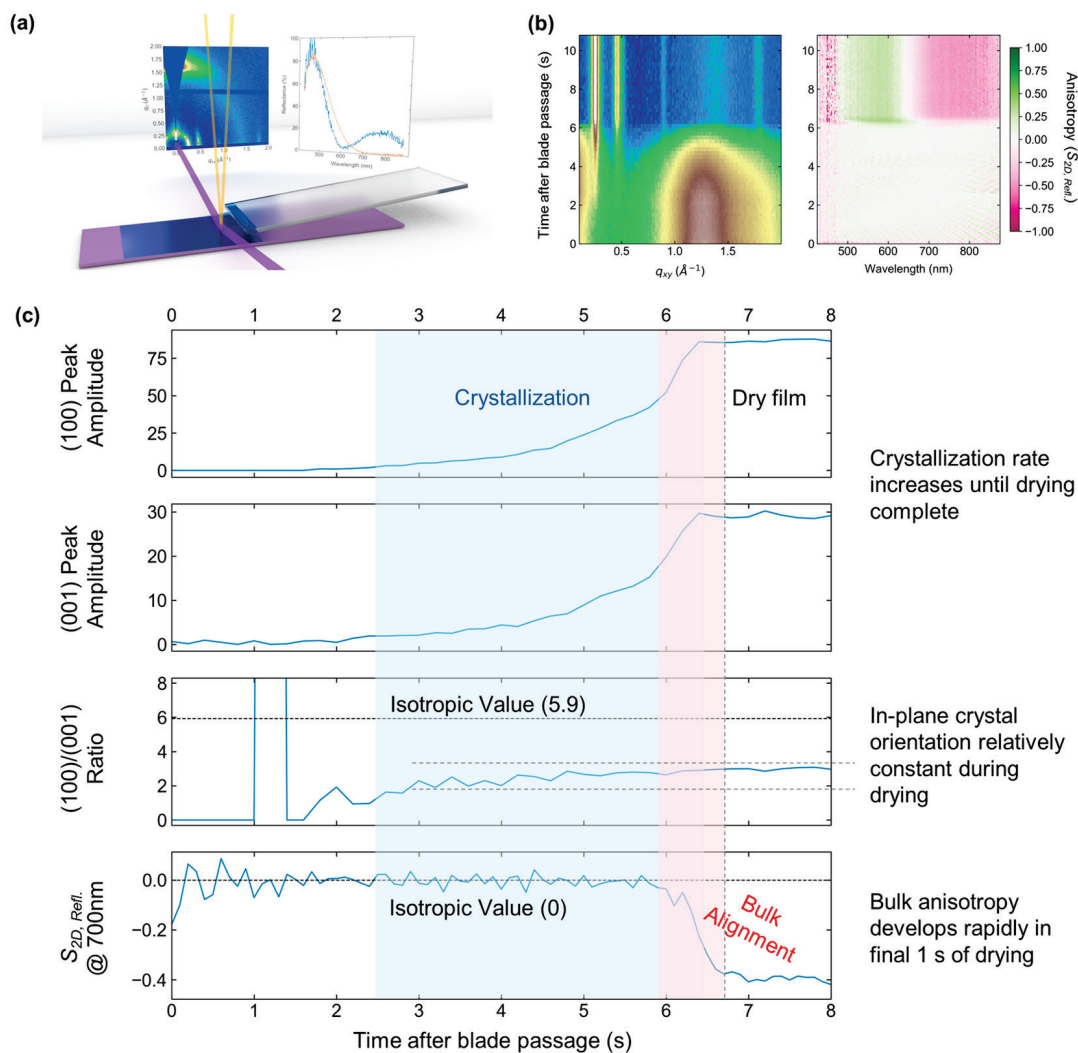


Figure 16. In situ characterization of a blade-coated N2200 thin film (5 mm/s). (a) Schematic of the experimental setup and raw data collection. (b) Raw line cut data from wide-angle x-ray scattering patterns and calculated S_{2D} for all wavelengths of raw ultraviolet-visible (UV-vis) reflectance data versus time after blade passage (vertical axis). (c) Amplitude of the (100) peak and (001) peak from peak fitting of each collected pattern, the ration of the (100)/(001) peak amplitudes, and S_{2D} from UV-vis reflectance at 700 nm versus time after blade passage in seconds. Source: [N.E. Persson et al., Chem. Mater. 31, 4133–4147, 2019.](#)

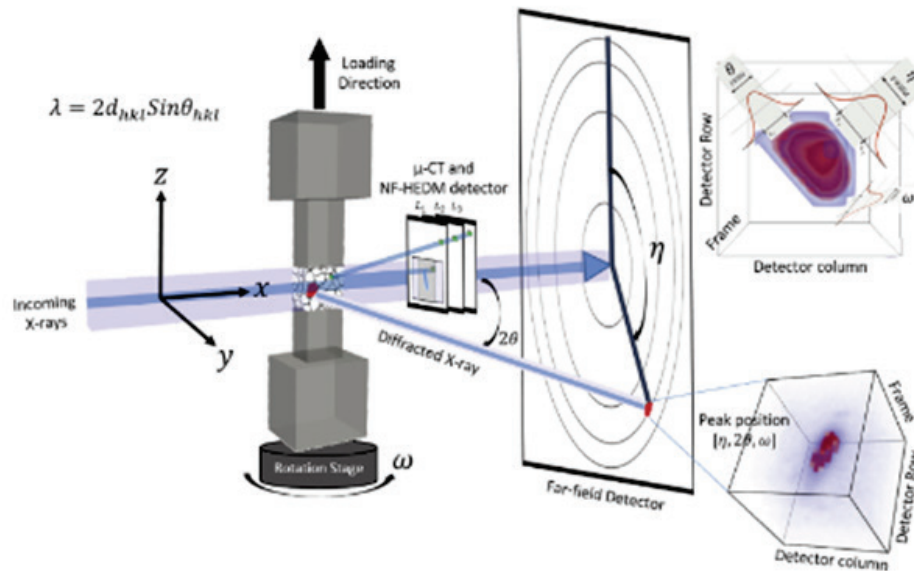


Figure 17. Schematic of an experimental setup featuring a suite of x-ray characterization techniques, which include m-CT (micro-computed tomography, NF-HEDM (near-field high-energy x-ray diffraction microscopy), FF-HEDM (far-field high-energy x-ray diffraction microscopy), and reciprocal space mapping. Source: Reprinted from *Acta Mater.* 179, P. Diwakar et al., [X-ray characterization of the micromechanical response ahead of a propagating small fatigue crack in a Ni-based superalloy](#), 342–359, 2019, with permission from Elsevier.

Figure 18 shows an example of multiple sensing and testing modalities (digital imaging correlation, acoustic emission, and high-speed camera) for monitoring environmental barrier coatings, critical ceramic components for gas turbines, in a relevant engine operating environment including heating, cooling, and a high-temperature/high-pressure air/steam mixed jet¹⁴ at a facility at the University of Virginia. Another example is the Gleeble system, a physical simulator for thermomechanical processing of metals used by academia and industry. With the penetrating power of high-energy synchrotron x-rays or neutrons, the microstructure of materials can be studied during thermomechanical processing, opening up opportunities for better understanding of the metal forming processes. A customized Gleeble system was built for synchrotron application¹⁵ and is being considered by the European Spallation Neutron Source for its new engineering beamline.¹⁶ Similar instrumentation is needed for answering fundamental science questions underpinning manufacturing issues. At present, the development of such complex sample environment lags behind the full light and neutron source capabilities, limiting the full potential of such facilities for more innovative in situ characterization.

Neutron sources have a unique role in advancing manufacturing know-how, because their high penetration power and sensitivity to light elements make it possible to use them to study large industrial components. The Engineering Diffractometer on the VULCAN beamline¹⁷ at the Spallation Neutron Source at Oak Ridge National Laboratory has been used to study the mechanical properties of industrial materials under conditions relevant to manufacturing. Neutron techniques including diffraction and imaging would benefit from further development of spatial resolution, from a few millimeters down to sub-millimeters, as recently demonstrated.¹⁸ They also would benefit from in situ capabilities to handle manufacturing processes involving larger components—complementary to those at synchrotrons—such as residual stress relaxation during heat treatment of industrial components. Neutron powder diffraction

has been routinely used for in situ measurements; and small-angle neutron scattering has been widely used for soft materials, as neutrons provide better contrast for soft materials than x-rays.¹⁹ It is important to connect these capabilities to solve critical manufacturing problems.

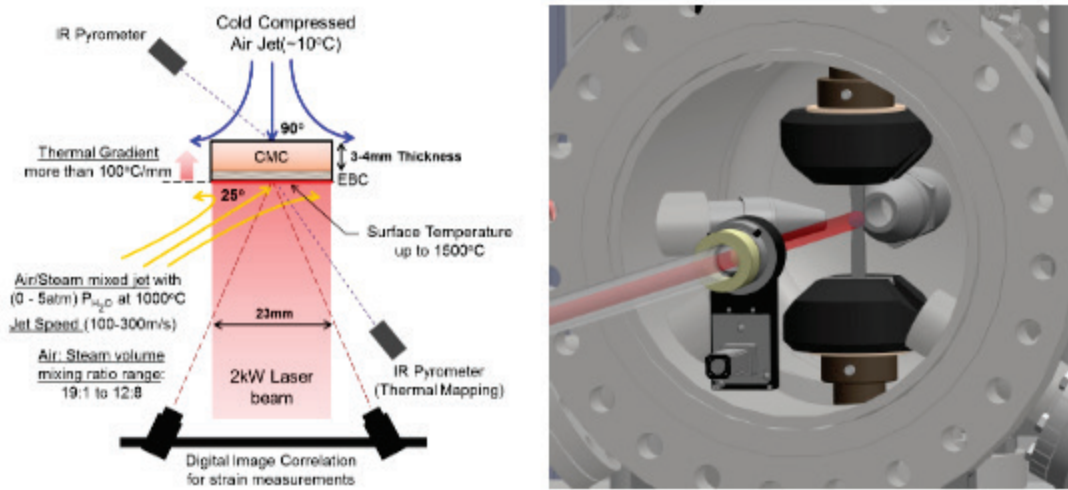


Figure 18. Simulated Engine Materials test facility for thermal barrier coatings and environmental coatings at University of Virginia. Source: H.S. Prabha et al. Laser Steam and Load (LSL) Simulated Engine Materials Testing Facility, University of Virginia website, <https://engineering.virginia.edu/wadley-lsl>.

Manufacturing processes can involve ultrafast events beyond what conventional characterization and even synchrotron-based tools can handle; this is where free-electron lasers (FELs) offer a path forward. An example of using FEL to understand the science behind industrial materials is an effort by Rolls-Royce to follow the shock-driven $\omega \rightarrow \alpha$ phase transformation of zirconium at picosecond time resolution at the Linear Coherent Light Source (Figure 19).²⁰ FELs have also been used to study chemical catalytic processes, and many opportunities exist to apply that understanding to transform chemical manufacturing processes. At present, a major limiting factor for applying FEL techniques to manufacturing problems is its low x-ray energy, which could be overcome by higher-energy FELs.

The scientific challenge is understanding material properties at much higher temporal, spatial, and energy resolutions than is possible today. Higher time resolution is particularly important for studying transient phenomena during processing, such as rapid cooling of laser metal AM; and higher spatial resolution is needed to visualize materials processing down to the finite local scale. In addition, for industry to benefit from in situ characterization, findings should be statistically significant, based not on single experimental observations but on multiple experiments with a high degree of fidelity.

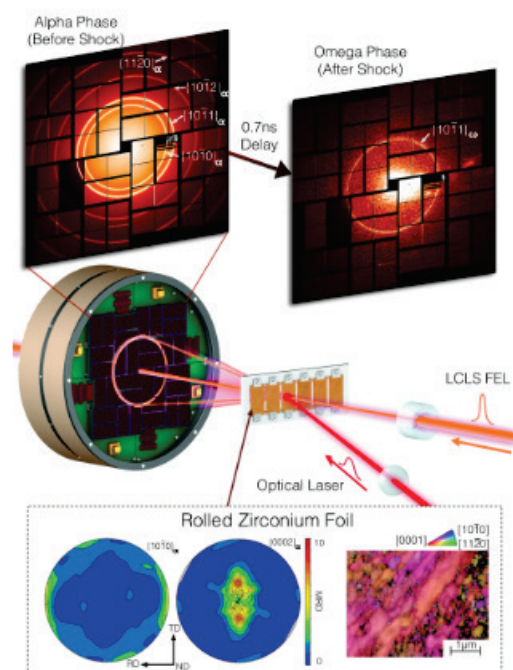


Figure 19. Picosecond dynamics of a shock-driven displacive phase transformation in zirconium. High-pressure solid-state transformations at high strain rates are usually observed after the fact, either during static holding or after unloading, or inferred from interferometry measurements of the sample surface. The emergence of femtosecond x-ray diffraction techniques provides insight into the dynamics of short-timescale events such as shocks. In this laser pump-probe experiment, the response of zirconium to laser-driven shocks, over the first few nanoseconds of the shock event, enabled the transition and orientation relationship to be observed in real time with picosecond resolution. An orientation relationship of $(10\text{-}10_{\alpha})// (10\text{-}11_{\omega})$ was determined that was in conflict with quasi-static annealing experiments $\omega \rightarrow \alpha$ in zirconium and the two pathways proposed for titanium. *Source:* Reprinted figure with permission from T.D. Swinburne et al. [Picosecond dynamics of a shock-driven displacive phase transformation in Zr](#), *Phys. Rev. B* **93**, 44119, 2016. Copyright 2016 by the American Physical Society.

Thrust 2. Operando characterization to elucidate new science for manufacturing processes

Operando methods interrogate both structure and activity in a simultaneous fashion, under operating conditions.²¹ Current efforts in operando characterization relevant to manufacturing often fall short of the appropriate scale. This limitation can be particularly detrimental, for example, for chemical manufacturing when upscaling a working process from laboratory to industrial scale. Given that most such manufacturing processes are taking place in reactors much larger than the benchtop apparatus, how can we create small-scale surrogates to extract information from chemical species under representative conditions? Given the physical limits on probing events at the molecular scale in large systems, this implies a need to advance the science of sampling from manufacturing systems to characterize reactions and materials under relevant circumstances.

An early example of operando characterization can be traced back to 1997 when researchers from DuPont studied Nylon 66 during heat-draw processing, under different temperatures and draw ratios, using synchrotron small-angle x-ray scattering (SAXS) and wide-angle x-ray scattering.²² Fast forward to 2019, and researchers from the Air Force Research Laboratory performed operando x-ray photon correlation spectroscopy (in SAXS geometry) measurements at the National Synchrotron Light Source (NSLS) II during 3D printing of a thermoset nanocomposite²³ (Figure 20) with silicate particles embedded in the extrudate—an epoxy-layered silicate composite ink. They demonstrated that the orientation of the platy particles varied markedly along the extruded features as a function of distance from the nozzle. This study, although limited to a single feature, opened up a significant space of process improvement through optimal control of the alignment of the anisotropic particles as a function of nozzle shape and geometry, as the extruded material exits the nozzle and is deposited on the substrate. Extending this type of

operando probe to more realistic conditions will require collaborative efforts between user facilities, academic researchers, national labs, and industrial users. The report from the 2020 Roundtable on Chemical Upcycling of Polymers²⁴ makes many important points about the challenges of *tertiary recycling*, in which chemical processes are used to break down plastics into smaller molecules that can then become feedstocks. Whatever process is used must be viable at the industrial scale to be effective in dealing with the vast volumes, which brings challenges with respect to catalysts, reagents, enzymes, or biochemical routes. Developing appropriate operando characterization techniques is likely to be crucial in this respect, not least for the manufacture of solid state catalysts, which themselves often need multiscale structures for support. The previous 2017 BRN workshop report *Catalysis Science*²⁵ provides much additional detail in this area but quite naturally focuses on the science of the catalysts themselves, as opposed to issues related to scaling up to industrial practice.

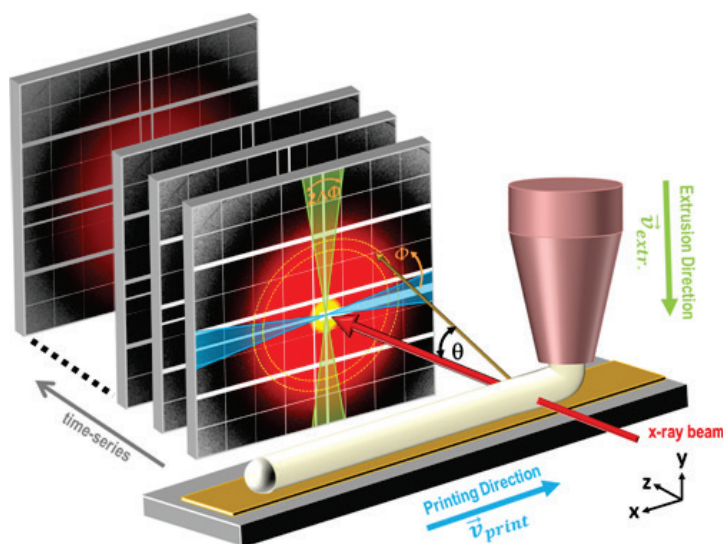
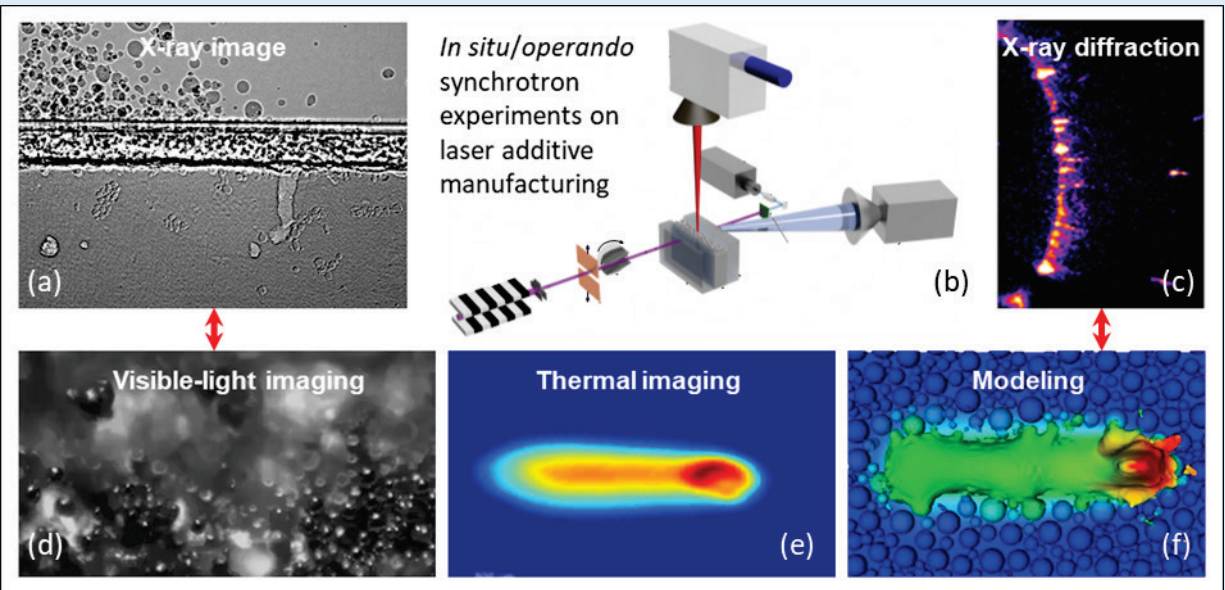


Figure 20. Schematic of the extrusion printing process and the scattering geometry for x-ray photon correlation spectroscopy in SAXS geometry. The material exits the nozzle at the same speed $v = 0.1$ mm/s as the nozzle is moving with respect to the build plate. The print ($\varphi = \pi$) and extrusion ($\varphi = \pi/2$) directions are normal to each other, and the print head and build plate are placed to pass orthogonally to the incoming x-ray beam. The scattered x-ray beam is defined by the scattering angle θ marked with dashed orange lines. The blue and green sections correspond to the extrusion and printing directions, respectively, with widths $2\Delta\Phi$. Source: [K.J. Johnson et al. Langmuir 35\(26\), 8758, 2019.](#)

Operando characterization through ultrafast imaging and diffraction during laser melting AM has gained traction from both academia and industry for obtaining insights into the dynamic process during melting of metal powders (see the sidebar). In this case, the spatial resolution of ~ 1 μm and framing rates up to megahertz are well suited to capturing laser-induced vapor holes (keyholes),²⁶ powder particle ejection, liquid metal spatter,²⁷ pore trapping, pore creation, and hot cracking,²⁸ as well as atomic-scale structures such as phase transformation through diffraction. Such processes are being improved by incorporating multiple path and powder handling capabilities to move closer to the actual 3D printing process. Probing chemical reactions at the atomic scale is an enduring challenge for operando characterization. Novel multi-modal tools need to be identified and explored. For example, while both pair distribution function (PDF) and computed tomography (CT) are well-established techniques, their combination has yielded unprecedented insight by spatial mapping of chemical reactions on an industrial alumina catalyst containing palladium.²⁹ The PDF-CT experiment showed nonreacting metal particles and a more strongly heterogeneous structure in the catalyst than anticipated. Such multi-modal probes point to the potential for time-dependent measurements and their broader applications to novel devices such as an artificial nose based on porous graphene.³⁰ Control of chemical reactions at atomic resolution also can be achieved by using scanning tunneling microscopy, as demonstrated for applications in the semiconductor industry.³¹ In atomic precision advanced manufacturing, for silicon doping, a monolayer of hydrogen atoms on a silicon {100} surface acting as a resist is selectively removed and replaced by phosphine molecules to accomplish the doping. These examples have shown the feasibility of characterizing and manipulating materials at atomic resolution, and much additional effort is still needed.

Sidebar: In Situ/Operando Synchrotron Experiments in Laser Additive Manufacturing.

The advance of laser AM relies on understanding the physics underlying the laser-metal interactions, the driving forces controlling the microstructure evolution, and the mechanisms responsible for the formation of structure defects. The superior penetrating power and brilliance of the high-energy photons at synchrotron facilities have enabled ultrafast imaging and diffraction studies of subsurface structural dynamics, leading to quantitative measurements of the melt pool, vapor depression, porosity, and crack formation, as well as dynamic phase transformation and evolution of microstructures. Such in situ/operando synchrotron experiments at the Advanced Photon Source and the Stanford Synchrotron Radiation Light Source help the manufacturing industry (1) understand how defects are generated and why some materials are difficult to print, (2) develop high-fidelity AM processes simulation models, and (3) validate sensory data for in-process monitoring and closed-loop control.



The experimental setup at the Advanced Photon Source (b) where x-ray and thermal images (a and e) and diffraction data (b) were taken in real time (c) and (d). Source: M.J. Matthews and S.A. Khairallah, Lawrence Livermore National Laboratory.

Operando characterization of the water splitting reaction, among the most desirable ways to convert solar energy to hydrogen as a fuel, has been demonstrated by using potential-sensing electrochemical atomic force microscopy (AFM) on a modified electrochemical reaction cell³² and infrared (IR) spectroscopy to study the catalytic processes at the liquid-solid interface.³³ In the AFM example—carried out in a laboratory setting—a nanoscale conducting tip senses the electrochemical potential across the electrocatalysts on various substrates in the presence of an electrolyte (see Figure 21), allowing direct observation of electrical interfacial properties of heterogeneous electrochemical systems during the water splitting reaction.³⁴

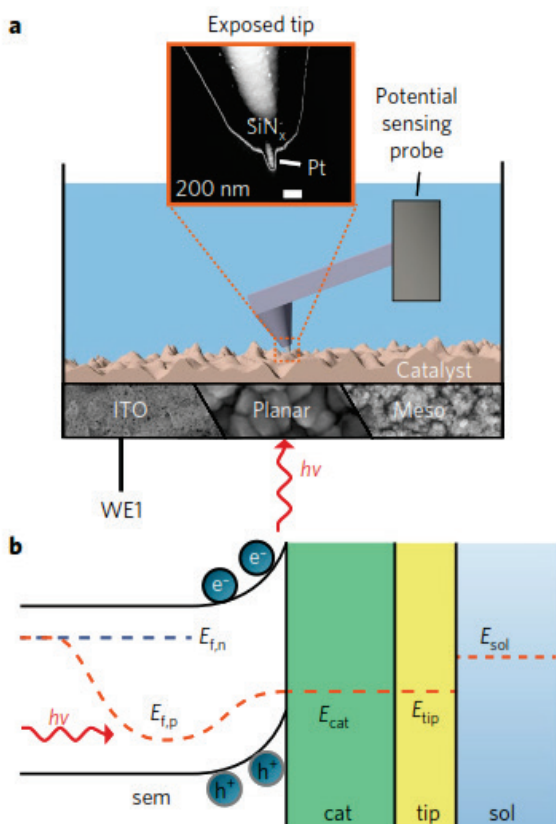


Figure 21. (a) The potential-sensing electrochemical AFM setup with an electrically insulated cantilever (except for the exposed tip) as a potential probe. The inset is a micrograph of the platinum tip. The blue color represents the electrolyte. A wide range of (photo) electrochemical systems can be studied in operando using this cell design. Electrodeposited catalysts on conducting indium tin oxide (ITO), planar hematite, or nanostructured/mesostructured hematite are connected to the circuit via the working electrode (WE1), and the illumination is provided from the bottom with a 405 nm light-emitting diode. (b) Band diagram for an illuminated semiconductor (sem) electrode (for example, hematite) coated with a catalyst (cat) layer. E_{cat} and E_{tip} are the Fermi levels of the catalyst and AFM tip, which are assumed to be in equilibrium. E_{sol} is the redox energy of the electrolyte. The difference between E_{sol} and E_{cat} divided by electron charge q is the overpotential driving water oxidation. Source: Reprinted by permission from Nature Publishing Group: [Nat. Energy, Potential-sensing electrochemical atomic force microscopy for in operando analysis of water-splitting catalysts and interfaces](#), M. R. Nellist et al. 3, 26–52, 2018.

More energy-related scientific areas that can benefit from operando characterization include biogas upgrade, separating methane from carbon dioxide and other gases, and NO_x reduction for reducing air pollution and climate control. To understand moisture tolerance during biogas separation, combined IR spectroscopy and mass spectroscopy (MS) was used operando in a flow chamber containing trace amounts of water. The molecular species on the surface of the molecular sieves were detected by IR, and simultaneously the gas products downstream were monitored by MS.³⁵ In the case of NO_x reduction, synchrotron x-ray absorption and emission spectroscopy at the iron absorption edge was used to investigate the mechanism of NO_x reduction with NH_3 , converting NO_x to harmless N_2 and H_2O in a miniaturized flow cell, aided by iron-containing catalysts.³⁶ A recent article described an approach for separating gaseous species based on flexible metal-organic frameworks subject to mechanical loading.³⁷ Under load, the material absorbs one gas species, e.g., CO_2 , but not another one, e.g., N_2 ; then the load is released, thereby allowing the separated-out gas to be released (Figure 22). The concept is elegant but clearly will require many additional steps of scientific discovery and engineering development to bring it to industrial scale.

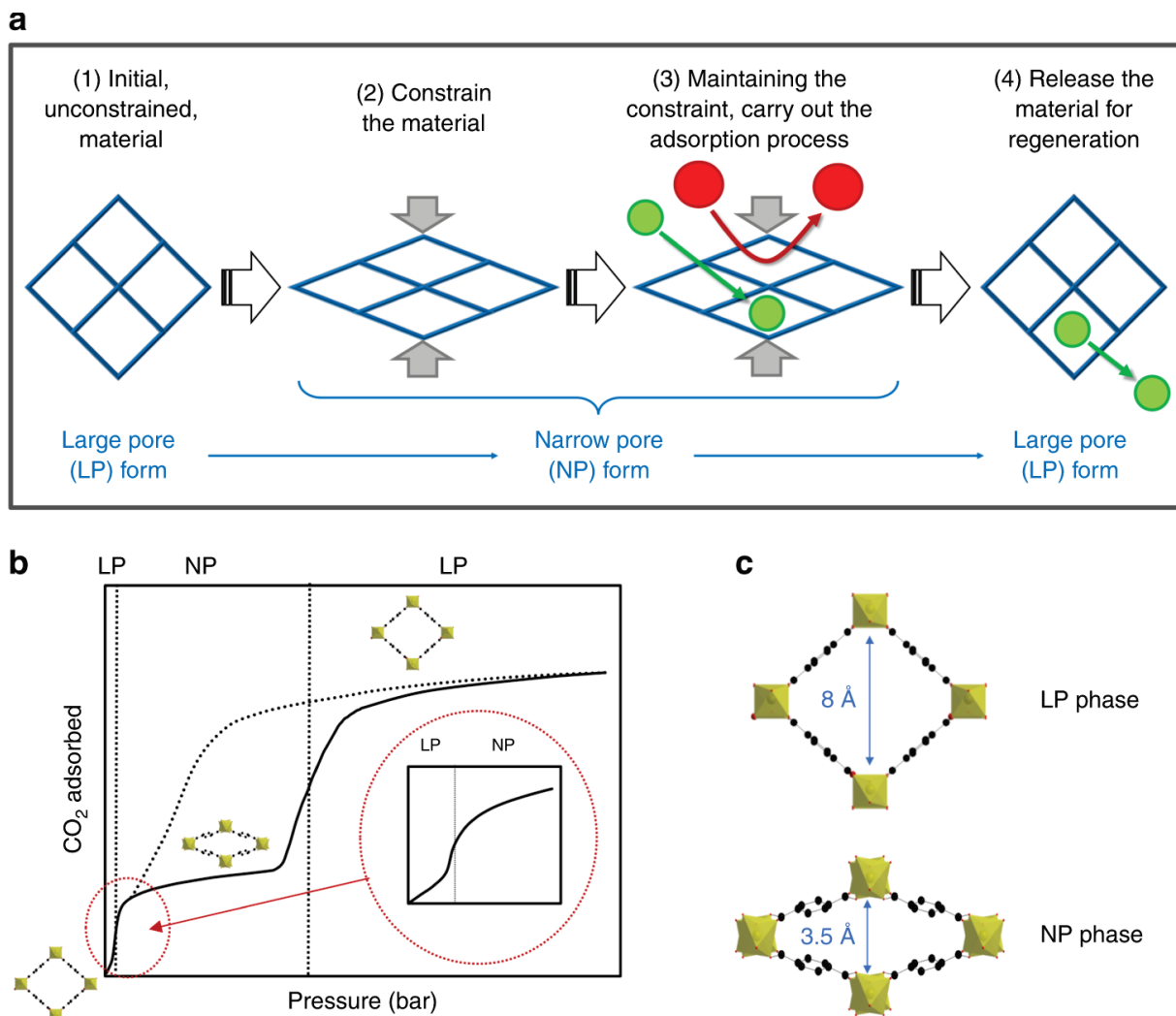


Figure 22. (a) Schematic representation of the concept developed in the present study: control of the breathing behavior of the flexible MIL-53 metal-organic framework (MOF) by application of an external mechanical pressure to provoke a molecular-sieving type of separation, followed by regeneration from the unconstrained MOF after release of the mechanical constraint. (b) CO₂ isotherm (full-line—adsorption, dashes—desorption) of the unconstrained MIL-53 evidencing a two-step process indicative of the framework breathing. Phase transitions between large pore (LP) and narrow pore (NP) phases are indicated by vertical dashed black lines. (c) Evolution of pore size aperture between the LP and NP phases of MIL-53. Source: [Chanut et al. *Nat. Commun.* 11, 1216, 2020](#). Licensed under a [Creative Commons Attribution 4.0 International License](#).

Addressing this thrust involves designing and fabricating professional-grade apparatus that emulate chemicals or materials manufacturing processing conditions for critical areas such as AM, catalyzed chemical synthesis, and polymer processing. This corresponds directly to the oft-stated need for in situ environments permitting observation of processes under real-world/real-time conditions (temperature, pressure, electromagnetic fields, gaseous and fluids) with minimal loss of spectral resolution.³⁸ Note that the large physical size associated with industrial-scale reaction systems, and materials processing in general, poses multiple challenges because it tends to force the use of highly penetrating radiation that often lacks the requisite sensitivity—hence the remarks elsewhere about the need for advances in sampling methods.

Thrust 3. Big data, artificial intelligence, and machine learning

The next important step in advancing manufacturing science is to convert large sets of in situ *and* operando experimental data into new scientific insights and predictive outputs that, in particular, are meaningful to industry. Using AI/ML for controlling measurements and interpreting the data is an important aspect and is highlighted by a recent BES report.³⁹ To provide unified platforms for data acquisition and analysis, BES has initiated a coordinated effort among five light sources and the Center for Advanced Mathematics for Energy Research Application (CAMERA)⁴⁰ and has developed the data acquisition software Bluesky⁴¹ and the data analysis and visualization software *Xi-cam*⁴² at Brookhaven National Laboratory and Lawrence Berkeley National Laboratory, respectively. Other promising developments include an autonomous SAXS experiment driven by a ML algorithm recently demonstrated at an NSLS-II beamline⁴³ and a convolutional neural network used to extract microstructural features automatically in tomographic image analysis.⁴⁴ AI/ML also can enable the analysis of large amounts of data quickly, beyond the scope of current post-experimental data analysis software. With increasing instrumental capabilities, thousands of measurements could be carried out in a few minutes or hours, in both in situ/operando and high-throughput experiments. High-throughput alloy design has been shown to be amenable to data analytics, whether the target is thermoelectric materials or high-temperature alloys for power generation.⁴⁵ ML is also being applied to image reconstruction to both improve accuracy and streamline the process.⁴⁶

Most data analysis algorithms and codes in use today were developed by scientists in their own fields in the pre-AI era. Examples include TomoPy^{*} for tomography; GSAS-II[†] for crystallography; HEXRD,[‡] IceNine,[§] HEXOMAP,^{**} and MIDAS^{††} for diffraction microscopy; Irena^{‡‡} for small-angle scattering; and Athena^{§§} for x-ray absorption spectroscopy. To develop next-generation AI/ML-based software, it will be essential to use domain-specific knowledge to apply AI/ML in a meaningful and productive manner, bringing together beamline scientists, users, and computer scientists. As Steve Jobs said, “Ideas are worth nothing unless executed.”

AI/ML-based algorithms also may be needed to go beyond interpreting measurement results. An oft-repeated dictum is that we should “measure structures as they form, with theory and modeling to guide synthetic processes on the fly.” Because simulations performed at the same resolution can take vastly longer than the experiment itself, the reality is that the idea of on-the-fly modeling has not yet materialized for most experiments. For example, crystal plasticity models have been shown to benefit from HEDM experiments,⁴⁷ and residual stress measurement can validate finite element models. However, at present, those models would take too much time to run to be part of operando experiments. This is also evident in the case of high-speed visualization of laser melting, for which a single high-fidelity simulation that includes liquid and vapor flow occupies hours on a supercomputer, compared with milliseconds per experiment. Therefore, the connection between modeling and experiments is vital to both, and we need to evaluate the feasibilities of what can be implemented now and what needs to wait till more powerful computing infrastructure becomes available.

This thrust is allied with the increasing demand in virtual or remote experiments that has been accelerated by the pandemic of 2020. Such remote experiments can benefit research at university laboratories and nanoscience centers, as well as at user facilities. For many years, pharmaceutical companies have used

^{*} TomoPy (Tomographic reconstruction in Python). <https://tomopy.readthedocs.io/en/latest/>

[†] <https://subversion.xray.aps.anl.gov/trac/pyGSAS>

[‡] <https://github.com/HEXRD/hexrdgui/releases>

[§] <https://github.com/FrankieLi/IceNine>

^{**} <https://github.com/HeLiuCMU/HEXOMAP>

^{††} <https://github.com/marinerhemant/MIDAS>

^{‡‡} <https://usaxs.xray.aps.anl.gov/software/irena>

^{§§} <http://bruceravel.github.io/demeter/documents/Athena/index.html>

remote access for protein crystallography research. As demonstrated as an unanticipated benefit from Covid-19, online meetings and carefully defined remote experiments can be as effective as face-to-face meetings and on-site experiments. This development can be particularly important for industrial researchers because it can eliminate travel and time away from their quotidian responsibilities, reducing R&D costs and expanding the participation of experimentalists to form broader multidisciplinary teams.

Science and Technology Impact

This PRD seeks to advance in situ and operando characterization for a lasting impact on manufacturing by creating the necessary infrastructure, AI/ML-driven analytics, and solution-driven collaborative research. This PRD aims to better align basic research with manufacturing needs so as to unleash the full potential of these powerful capabilities for long-term economic benefits. By presenting challenging applied, real-world problems, this PRD also will impact the education of the next generation of scientists and engineers.

This PRD will impact manufacturing in the following three ways:

1. Establish a holistic research infrastructure around in situ and operando characterization, from sophisticated sample environments to AI/ML-driven analytics, for understanding materials processing-structure-property-performance relationships under real-world operating conditions.
2. Promote a paradigm shift in materials discoveries and manufacturing innovation based on knowledge and understanding, instead of empirical or Edisonian schemes, combining strengths and expertise from academia, national laboratories and industrial researchers toward long-term, transformative objectives.
3. Enhance the global competitiveness of US industry in energy-related technologies, especially in areas such as quantum materials, renewable energy, environment, and critical materials.

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PRD 4: Direct Atom and Energy Flow to Realize Sustainable Manufacturing

Summary

Manufacturing processes for many industrially important chemicals and materials have approached their thermodynamic limits in efficiency. Even if further gains were possible, the growing demand for energy and materials services using chemical feedstocks cannot be met sustainably in the decades ahead, a fact that jeopardizes the energy and resource security of the United States. Transformative change in manufacturing toward sustainability requires the development of revolutionary methodologies that more effectively localize and direct chemical and materials transformations at the primary sites of energy conversion from a variety of energy inputs (e.g., electrical, thermal, radiative, and mechanical), as well as integrated processes that couple kinetics and transport across length scales to dramatically reduce the water and energy use and increase efficiency of manufacturing, demanufacturing, and remanufacturing. The promise of renewable electricity and abundance of high-quality recycled feedstocks, when coupled to these methodologies and processes, has the potential to improve manufacturing by reducing waste and pollution while also decarbonizing the manufacturing of existing and future chemicals and materials.

This PRD seeks to address key questions that include the following: *What are the methodologies to achieve atom and energy efficiency for sustainable manufacturing? How can science enable adaptive and resilient manufacturing across scales to exploit renewable or recycled feedstocks?*

Scientific Challenges

Approximately 50% of global annual industrial greenhouse gas (GHG) emissions are released during the production of five major materials: steel, cement, paper, plastics, and aluminum. A significant fraction of the remaining 50% is attributed to the production of other metals, glass, and chemical feedstocks, as well as the manufacturing of materials into products.¹ Many of these industrially important chemicals and materials have been produced for decades, if not centuries. Iterative improvements in the efficiency of their production have approached either practical or thermodynamic limits.²

As demand grows for energy and materials services, both of which are overwhelmingly reliant on chemical feedstocks, so will stresses on the ways and means of production (Figure 23).^{3,4} It remains unclear whether adequate resources can be secured and managed for primary production in higher volume using today's best-practiced manufacturing processes without also incurring significant economic and environmental costs. On the current path, manufacturing supply chains appear increasingly at risk.

Basic science needs, described in the three challenges that follow, can be generalized in four areas:

1. Convergent theoretical and experimental frameworks are needed to understand and control multiphase reactions under the influence of chemical and electrochemical potential with respect to adsorption, bond activation, transfiguration of bonds, and diffusion of reactive species in complex reactive environments. Such frameworks also are needed to understand and control processes where transformations are driven by several different energetic inputs.
2. New research paradigms are needed for designing and integrating conversion and separations; coupling highly active catalysts with membranes and solvation; and replacing thermally controlled separations used to produce, refine, recover, or recycle important chemical and materials feedstocks.
3. Design principles are needed to develop next-generation materials and manufacturing processes that are intrinsically more circular and that enable remanufacturing to be less wasteful and require less energy.

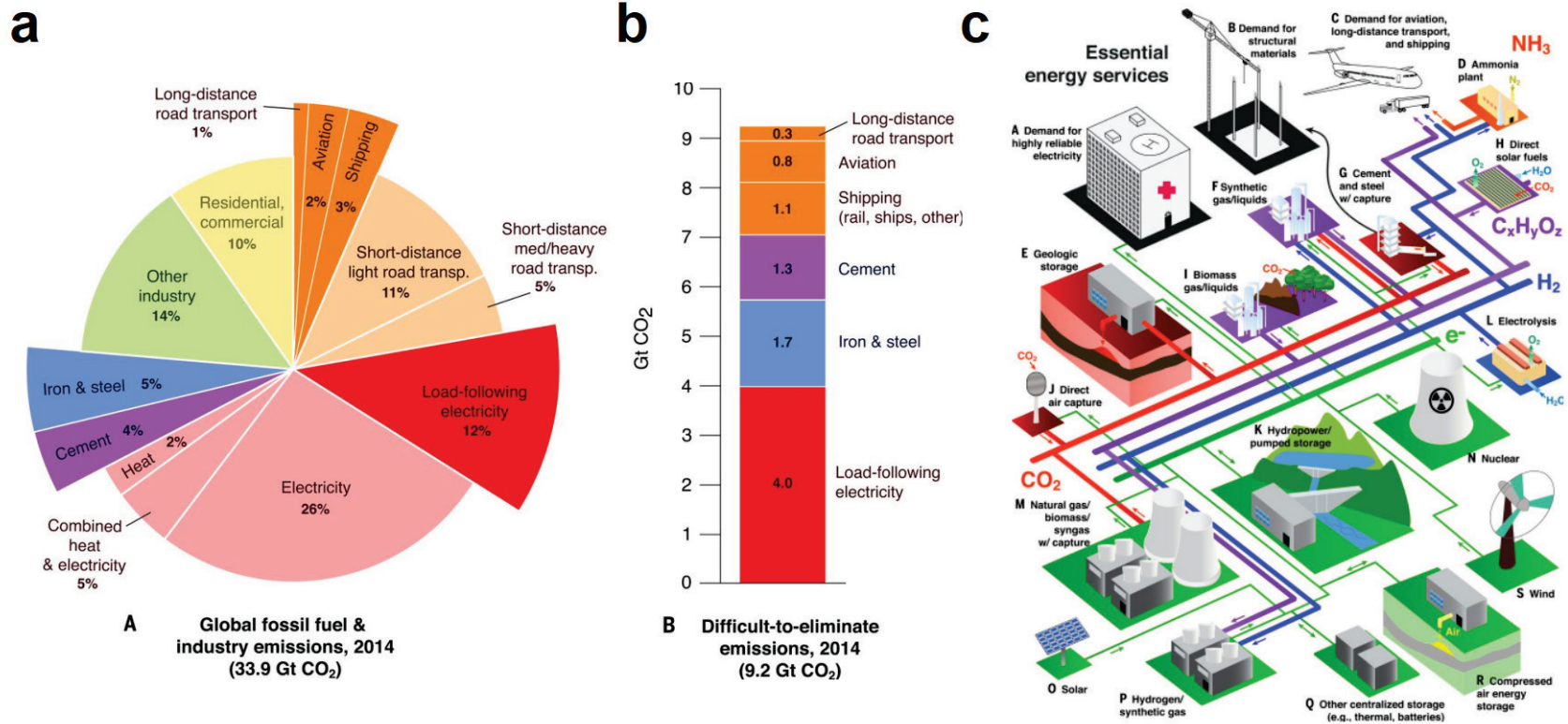


Figure 23. (a–b) Estimates of CO₂ emissions related to different energy services, including manufacturing, highlighting (for example, by longer pie pieces in A) those services that will be the most difficult to decarbonize, and the magnitude of 2014 emissions from those difficult-to-eliminate emissions. The shares and emissions shown here reflect a global energy system that relies primarily on fossil fuels. Both (a) the shares and (b) the level of emissions related to these difficult-to-decarbonize services are likely to increase in the future. Totals and sectoral breakdowns shown are based primarily on data from the International Energy Agency and EDGAR 4.3 databases. Residential and commercial emissions are those produced directly by businesses and households; and “Electricity,” “Combined heat & electricity,” and “Heat” represent emissions from the energy sector. **(c)** Schematic of an example integrated system that can provide essential energy services integrated with manufacturing without adding any CO₂ to the atmosphere. Colors indicate the dominant role of specific technologies and processes: green, electricity generation and transmission; blue, hydrogen production and transport; purple, hydrocarbon production and transport; orange, ammonia production and transport; red, carbon management; and black, end uses of energy and materials. Source: Reprinted from [S.J. Davis et al. Science 360, eaas9793, 2018](#).

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4. New multiphysics models, in situ and operando characterization capabilities, and data analytics are needed to reveal insights into how to couple kinetics and transport across scales and respond to changing conditions and environments in adaptive manufacturing processes.

Challenge 1. Pathways to reduce energy and enhance sustainability for primary production of commodity chemicals and materials.

The manufacturing of chemicals in the United States consumed more than 8 EJ of energy in 2018, accounting for ~25% of industrial energy consumption.⁵ By 2030, chemical manufacturing could account for almost 10% of all US energy consumption.⁶ Of the ~140,000 chemicals produced, the top 18 account for 80% of the energy consumption in the chemical sector.⁷ Although advances in catalysts and process intensification are expected to reduce the demand for energy by as much as 20–40%, a paradigm shift toward electrification of the chemical industry offers the promise of sustainability long term. Specifically, chemical transformations that today use heat and pressure may in the future be facilitated by light- or voltage-driven processes; coupling new classes of reactions at the anode and cathode offers compelling means for lowering GHG footprints for a broad spectrum of primary production processes.

To that end, while abundant renewable electricity has created an unprecedented opportunity to electrify and enhance sustainability of the primary production of a broad class of chemicals and materials—provided the electricity comes from zero-emission and renewable sources^{8,9}—more renewable energy is needed. The gap is along the lines of terawatts of capacity, in addition to longer-term energy storage (10–20 days) not feasible with conventional batteries. Thus, advances in energy storage in chemical bonds are opportunities that low-cost renewable electricity can facilitate. Historical examples of electrochemistry at scale in the manufacturing environment include the chlor-alkali process for chlorine production, the Hall process for aluminum, and electrolytic manganese dioxide for alkaline batteries. Electrochemical refining and processing of certain metals is already widely practiced and generates high-purity metals such as aluminum, zinc, copper, lead, and silver at low cost and in high volume. Advancing the science of electrorefining may make possible more sustainable production of other important metals, such as iron for steel production and rare earths for magnets for electric vehicles and wind turbines, as well as refined feedstocks for cement production and for manufacturing battery components (Figure 24).^{10–12} Gains on these fronts would open the possibility of more cost-competitive midstream management of resources in domestic manufacturing systems, obviating the need to export raw materials or concentrates extracted from ores for processing only to re-import them as higher-value products.

The electrification of the chemical industry could lay important groundwork for more sustainable production of high-volume chemicals, including ammonia,¹³ olefins (e.g., ethylene and propylene),^{14,15} alcohols (e.g., methanol), epoxides¹⁶ (e.g., ethylene oxide or propylene oxide), as well as monomers for chemically recyclable commodity polymers (e.g., caprolactam, glycols, and terephthalic acid). Along similar lines, low-temperature electrochemical activation of C–H bonds would also be revolutionary; in the case of methane, it points to compelling opportunities for a broad range of chemical feedstocks ranging from syngas to liquid fuels, oxygenates, and light olefins.^{17–23}

To prepare for future electrification of the chemical industry, basic energy research into the electrification of primary production and the refining of raw materials is needed. Processes undergoing electrification will be carried out using transformations at interfaces or in multiphase media, where the primary sites of energy conversion may involve electrons, ions, photons, phonons, and mass at complex phase boundaries. Sometimes these transformations may occur at extremes in temperature, pressure, or shear forces. Under such conditions, we do not know how to synergistically couple electrical, thermal, radiative, and mechanical energy at primary conversion sites to target conversions and direct the process selectively at

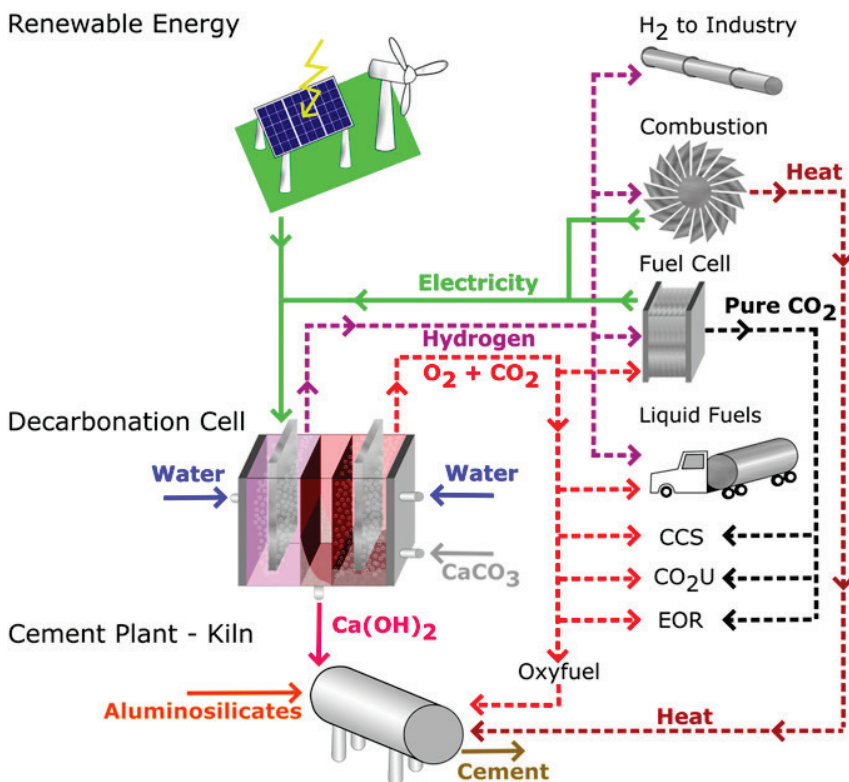


Figure 24. Scheme for a low-emission, electrochemically based cement plant. An electrochemical reactor powered by renewable electricity converts CaCO_3 to Ca(OH)_2 for use in cement synthesis. The decarbonation cell uses the pH gradient produced by neutral-water electrolysis to dissolve CaCO_3 at the acidic anode and precipitate Ca(OH)_2 where $\text{pH} \geq 12.5$. Simultaneously, H_2 is generated at the cathode and O_2/CO_2 are generated at the anode. These gas streams can serve several alternative roles in a sustainable production system. CO_2 can be directly captured from the inherently concentrated stream. Electricity or heat can be generated from the H_2 and O_2 via fuel cells or combustors. The O_2/CO_2 oxy-fuel can be recirculated to the kiln for cleaner combustion in the cement sintering cycle. CO_2 reuse and utilization concepts can be employed, such as use in enhanced oil recovery or production of liquid fuels. Source: [L.D. Ellis et al. *Proc. Nat. Acad. Sci.* 117\(23\), 12584–12591, 2020.](#) Used by permission.

low intensity. To bridge this gap, new theoretical and experimental frameworks are needed to understand adsorption; localized bond activation; surface diffusion of reactive species; and the transfiguration of bonds under realistic conditions when the transfiguration is driven by several energetic inputs and gradients in chemical potential local to the conversion site. We further lack molecularly informed knowledge for tying macroscopic system performance factors such as rate, Faradaic efficiency, catalyst selectivity, and lifetime to the fundamental energy conversion processes involved. Doing so would require new multiphysics models, in situ and operando characterization capabilities, and data analytics that reveal new insights into how to couple kinetics and transport across scales to realize sustainable primary production and refining.

Challenge 2. Scientific advances to realize lossless and waste-free circularity in manufacturing systems.

Manufacturing systems are linear by design: raw materials are extracted and processed, followed by production and assembly, use, and then disposal of the product at the end-of-life or end-of-use. Manufacturing for circularity considers how to design for reuse, recycling, or reconfiguration at the end of use rather than disposing of the product and how to reduce the need for raw materials. While there is considerable industrial interest in circularizing manufacturing systems to improve their sustainability,^{24–29} it is not yet clear how to design and deconstruct products efficiently and recover inherently valuable chemicals and materials at high purity for recirculation in closed-loop life cycles. This is particularly true for emerging technologies such as additive manufacturing (AM).³⁰

To underscore the problem, an estimated ~5 billion metric tons of plastics and their embodied energy (~165 EJ) and value as chemical feedstocks have been irrevocably lost to the global economy because of the impracticality of retrieving them.³¹ Furthermore, there is growing demand for high-quality recycled feedstocks for secondary manufacturing of polymer resins, yet these are scarce because of scientific challenges in polymer deconstruction and additive removal (i.e., dissociation).^{32–36} To meet the demand for resource circularity will require transformative advances for managing and preventing feedstock losses, or for downgrading these valuable resources at the level of individual bonds along polymer chains to permit their reuse in fully closed-loop recycling processes (Figure 25). Closed-loop chemical circularity has been demonstrated at scale for poly(ethylene terephthalate), high-density polyethylene, polystyrene, and nylon-6. However, to date, these processes remain energy intensive, inefficient, and intolerant to common polymer additives, impurities, and mixtures. Nonetheless, these early demonstrations point to a future in which chemical recycling of commodity polymers could be more widely implemented, if further innovation could be realized via breakthroughs in polymers, catalysts, processes, and chemical separations for efficient monomer recovery and additive removal. The sustainability of future digital manufacturing practices for polymers and composites may likewise hinge on the design and development of the next generation of circular polymer resins for AM.

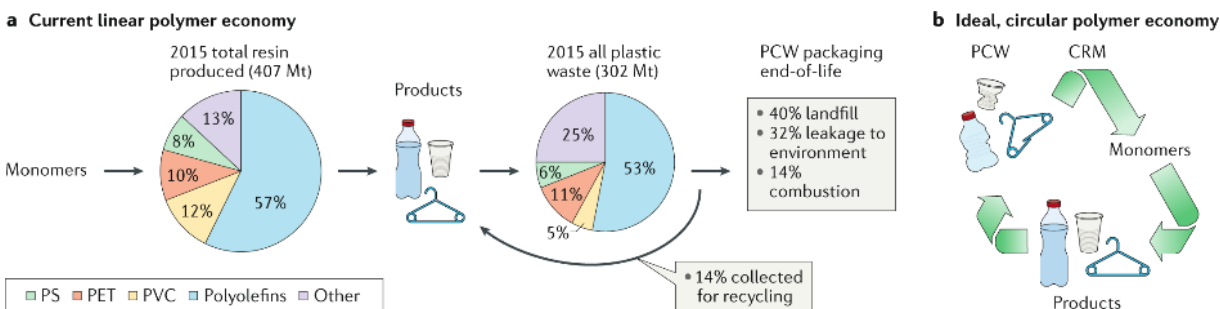


Figure 25. (a) In 2015, the mass of plastic waste was 74% of the mass of resin produced. The graphs show the contributions of different polymers to total plastic resin production (values averaged over 2002–2014) and waste (2015 values). A small percentage of plastic waste is collected for recycling; the remainder goes to landfills, leaks to the environment, or is combusted (the values shown are for plastic packaging only). (b) Cycle showing the idealized polymer economy, in which post-consumer waste (PCW) plastics undergo chemical recycling to monomer (CRM). Mt=megatonnes; PET=poly(ethylene terephthalate); PS= polystyrene; PVC=poly(vinyl chloride). Source: Reprinted by permission from Nature Publishing Group: *Nat. Rev. Mater.*, [Chemical recycling to monomer for an ideal, circular polymer economy](#), G.W. Coates and Y.D.Y.L. Getzler, 5, 501–516, 2020.

The recycling of metals, compared with recycling of polymers and composites, is conducted at markedly higher rates of ~50% for steel, aluminum, copper, zinc, lead, and nickel.^{37–41} However, if high purity is the primary criterion for assessing the landscape of reuse markets, thermodynamics may be the ultimate arbiter, given how few products incorporate metals in pure form. The recycling of complex alloys,

ceramics, and other inorganic materials remains an obstacle. For alloys in particular, including those being developed for AM,⁴²⁻⁴⁴ the foundational thermodynamic behavior of alloying elements in those materials makes their separation in a recycling process either very energy intensive or practically impossible with technology currently available. Electrochemical extractions, separations, and other refining processes may further open doors to new and efficient recycling processes for recovering important metals for reuse from complex alloys, ceramics, oxides, and other materials used in AM, magnets, fuel cells, and batteries for electric vehicles and the grid at end-of-life. The materials complexity inherent in these products currently presents formidable but not insurmountable challenges in separations to recover resources in high purity for secondary production from recycled materials.

Realizing circularity in manufacturing systems for specialty and commodity chemicals, polymers, metals, and other materials requires scientific breakthroughs that address outstanding challenges in understanding and controlling thermal and material entropic losses associated with circularity. These include the dilution or mixing of components after the deconstruction of materials in use today. Furthermore, it may be necessary to design the next generation of materials for new manufacturing processes that are intrinsically more circular, to purposefully design out waste and yet enable emerging technologies. Such designs would need to address a critical yet largely unmet need to deliver performance concomitantly with reduced complexity to minimize entropic losses in circular manufacturing systems. Innovative strategies are likewise needed to lower the consumption of resources and energy in the chemical recycling processes necessary to isolate refined feedstocks for remanufacturing. Although separations and feedstock refining processes are currently conducted thermally or under thermodynamic control, electrification of these processes may be possible to enable them to be sustainable. Exploiting kinetic control may likewise present compelling new opportunities to overcome thermodynamics limits for energy and atom efficiency in the separation of material components after deconstruction.

Challenge 3. Enable robust and resilient manufacturing when using unconventional energy inputs and feedstocks that exhibit heterogeneity.

Prospects for scaling production to meet demand using renewable or recycled feedstocks, and even waste, are enticing for lowering GHG and environmental impacts. While mineral ores, petroleum hydrocarbon gas liquids, and natural gas are raw materials used today in metals, petrochemicals, plastics, and other carbon-containing materials, the feedstocks of tomorrow will likely be alternative and more sustainable sources such as H₂O, CO₂, and N₂. Electronic waste, waste plastics, manures, sludges, municipal solid waste, and flue gases are also emerging feedstocks that are plentiful but geographically dispersed, fluctuating in availability, heterogeneous in makeup, often refractory, and expensive to aggregate.⁴⁵ Collectively, these materials inputs are rich sources of carbon, hydrogen, nitrogen, and oxygen atoms. However, they will need to be reconfigured selectively and efficiently to new carbon-carbon, carbon-hydrogen, carbon-nitrogen, and carbon-oxygen bonds during chemical production. Understanding and controlling such chemical transformations with these unconventional feedstocks requires an inversion of precedents in reaction discovery, design, and engineering: compounds that have historically been regarded as non-reactive combustion and waste products could become plausible reactants if they could be reduced to usable chemicals by thermochemical, electrochemical, photochemical, and biological routes (Figure 26). Transforming highly oxidized feedstocks also may require new sources for the energy that today typically comes from H₂-derived from fossil fuels but in the future may be supplied by lower-carbon energy sources, such as renewable electricity that will fluctuate in availability.

In situations where materials and energy inputs are expensive to aggregate, or where the product has dispersion benefits, smaller-scale modular systems provide advantages over large centralized manufacturing facilities in use today, including resiliency, individual capital, reduced risk, and reduced time to market.⁴⁶ To realize atom and energy efficiency, distributed manufacturing at smaller scale requires new methodologies, catalysts, and reactor designs that can accommodate or adapt to feedstock

heterogeneity and production scale, rather than miniature versions of larger-scale facilities. The need for smaller-scale processes does not preclude the large-scale facilities used now; indeed, refined intermediates from small-scale plants are more easily transported than the raw material, and they may feed larger-scale facilities where downstream manufacturing processes may be conducted more efficiently.^{46,47}

Basic scientific efforts to realize resilient manufacturing with unconventional yet sustainable energy sources and feedstocks must address catalyst resiliency under variable operating conditions, often in the condensed phase. Scientific advancements in high-throughput characterization of variable feedstock compositions and energy inputs to enable rapid adjustments of processing conditions in operando are needed. Designing catalysts that can handle feedstock and energy input variability would also be impactful. Understanding how to integrate conversion and separations to drive key processes under nonequilibrium control also may provide opportunities to improve efficiency and lower the intensity of unit processes: examples are coupling highly active catalysts with membranes and using multiple solvents.

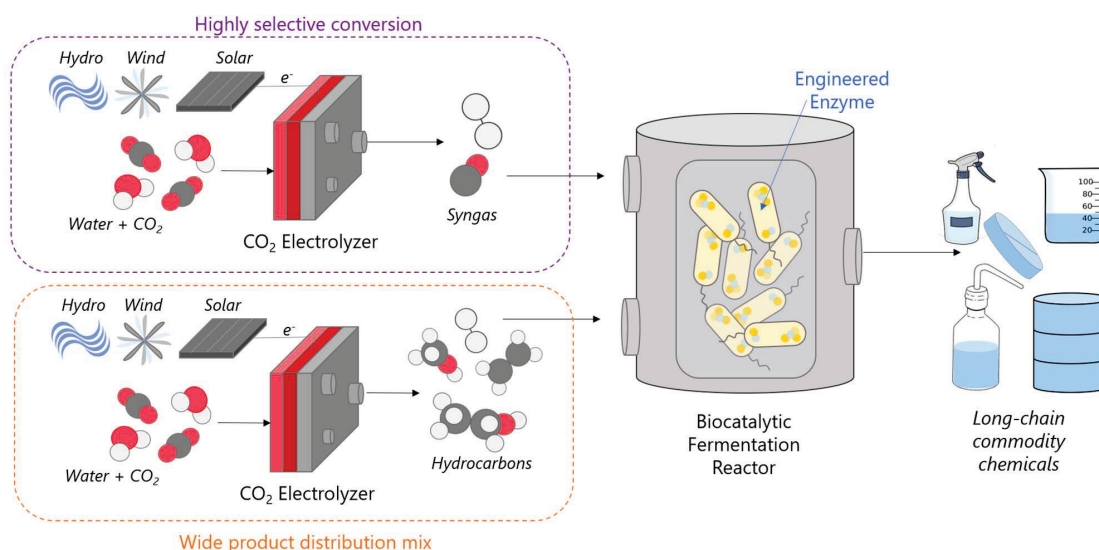


Figure 26. Coupled catalytic pathways toward long-chain commodity chemicals. Today, CO₂ may be converted to syngas at very high selectivity using silver- or gold-based catalysts (top left). Alternatively, CO₂ can be converted into a wide range of hydrocarbon and oxygenate products using copper-, tin-, or palladium-based catalysts (bottom left). These products can then be used as inputs for genetically engineered enzymes and bacteria to convert to more complex commodity chemicals. Source: From [P. De Luna et al., *Science* 364, eaav3506, 2019](#). Reprinted with permission from AAAS.

Researchers lack a complete understanding of solvation effects in complex reaction mixtures having multiple liquid phases under non-ideal conditions. In multiphase reaction sorption, catalyst distribution and catalyst lifetime are exceptionally challenging to control and sustain in manufacturing processes. Further, the phases may be used to remove and capture inorganics, acids, bases, and various catalyst poisons as well as remove products from reactants. Electrocatalysis enabling the use of renewable electrons to provide the driving force for reactions is likely to have a more important role. An understanding of the influences that control Faradaic efficiency is needed so that energy inputs can be used more efficiently. Theory will need to be developed that can accurately describe multiphase reactions under the influence of external electrical potential on rate and selectivity.

A summary of research directions focused on advancing the basic science to enable sustainable manufacturing is detailed below.

Research Thrusts

Thrust 1. Harness diverse forms of energy for atom- and energy-efficient manufacturing

Revolutionary discoveries in methodologies and processes that achieve higher atom and energy efficiency in producing commodity chemicals and materials are needed to change course toward more sustainable manufacturing. Basic research to enable such a manufacturing revolution could range from the discovery of new reagents, reaction pathways and intermediates to electrocatalyst design and entirely new reactor concepts for high efficiency and throughput. If realized, these discoveries could lead to disruptive innovations for lowering the resource, carbon, and energy intensity of primary production across industries, ranging from extractive metallurgy to the production of commodity and specialty chemicals.

Addressing this challenge will require the advancement of scientific methods that harness diverse forms of energy more productively than is possible today to achieve a desired transformation under mild conditions with high Faradaic efficiency and with minimal or no waste generated. Efforts building on such discoveries will make further inroads by developing new research frameworks to understand how to implement such methods in manufacturing processes so that they synergize with kinetics and transport across scales to direct transformations selectively and efficiently.

Increasing the efficiency of chemical transformations requires reducing the difference between the standard chemical state of molecules before the reaction and at their activated state, compared with current best practice. Although researchers are beginning to understand how the excess potentials of ground and activated states are determined by the reaction environment, it is unclear to what extent the two are linked. Uncoupling manipulations of ground and transition states will enable new design parameters for chemical transformations. One way of achieving such manipulations is through catalysts that maintain reacting molecules in a high-energy state while stabilizing transition states. An example of such approaches is manipulating entatic states of reactants under confinement, e.g., in porous media or in the active sites in biocatalysts. Using either light or electrical energy for the (re)generation of reactive intermediates and co-factors along the reaction coordinate could provide valuable chemicals in more eco-friendly processes.⁴⁸ This could include chemical looping (mediated) reactor systems. Examples include electrochemical regeneration of active metals, such as lithium, which spontaneously reacts with nitrogen to form lithium nitride. Lithium nitride can then react with a proton source to generate ammonia⁴⁹ (Figure 27) and light or electrical energy for reducing iron oxide nanoparticles to generate reactive iron nanoparticles for hydrogen generation.⁵⁰

It will be increasingly important to be able to use electrochemical potential and electrochemically driven gradients in chemical potential, locally at interfaces and spatially in reactors, to manipulate the reaction coordinate or control the selectivity and efficiency in chemical transformations. The molecular basis for the overpotential in potential-driven chemical transformation remains highly obscure, but it is foundational to the sustainability of electrochemical production and refining of chemical feedstocks used in cement, steel, alloys, magnets, ceramics, polymers, and batteries. Likewise, it remains a challenge to differentiate the chemical basis for reactivity from deactivation in a catalytic process implementing several energetic inputs. This indicates an imminent need to develop more advanced in situ, operando, and multi-modal characterization capabilities to connect the scales (i.e., energy and space) and to more effectively characterize transient processes at catalytic sites in a time-resolved manner with chemical specificity. There is also a critical need for new theoretical frameworks and atomistic simulations from first principles to map reaction coordinates under realistic reactive environments when mediated by catalysts, particularly for light- and voltage-driven processes, as well as in energetically coupled systems.

To accelerate the discovery and development of novel and more sustainable manufacturing methodologies, it will be necessary to seamlessly integrate data analytics across workflows. The aim will be to understand the basis by which catalyst systems realize their activity, selectivity, stability, or

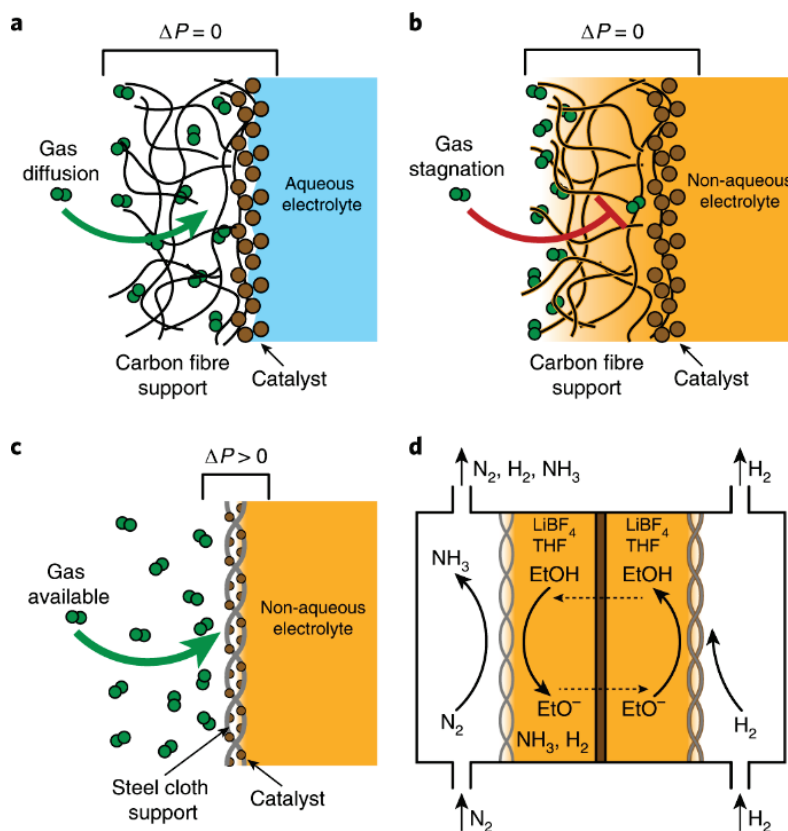


Figure 27. Overcoming transport limitations in electrochemical ammonia synthesis under non-aqueous conditions with near-unity Faradaic efficiency using tailored gas diffusion electrodes (GDEs). (a) A hydrophobic GDE with an aqueous electrolyte, where well-defined gas-liquid contacting exists. (b) A hydrophobic GDE with a non-aqueous electrolyte, where considerable wetting of the carbon fibers occurs, effectively flooding the catalyst. (c) A catalyst-coated steel cloth is shown. A lack of substantial capillary action and the presence of a non-zero pressure (P) gradient across the cloth prevent complete catalyst flooding. (d) Proton donor cycling is shown in a cell with a proton-producing anode. *Source:* Reprinted by permission from Nature Publishing Group: *Nat. Catal.*, [Non-aqueous gas diffusion electrodes for rapid ammonia synthesis from nitrogen and water-splitting-derived hydrogen](#), N. Lazouski et al. 3, 463–469, 2020.

precise and energy-efficient reconfigurations of chemical bonds, as well as reversible stimulated deconstructive transformations, are essential for realizing circularity in manufacturing systems. In the absence of such innovations, recycling invariably will lead to significant quantities of waste and will possibly exacerbate the consumption of resources and energy to cope with entropic losses in the process. Achieving circularity by closing the loop in manufacturing, demanufacturing, and remanufacturing can prevent raw materials from reaching criticality.

Basic energy research in resource circularity will advance by managing complexity across scales and in recycling processes in endeavoring to recover constituent resources in sufficient purity for recirculation. For polymers, this will include the development of innovative polymer chemistries for quantitative scrap recovery, as well as new chemical or catalytic depolymerization methodologies to transform synthetic or natural polymers back to re-polymerizable monomers with low intensity. Reaction coordinates for polymer

durability in a process with different feedstocks or energy inputs, and how such characteristics in turn influence the energy, carbon, and water intensity of the process. This will require the development of new ML algorithms that are physics-, chemistry-, materials-, and process-aware so that comprehensive assessments of both successful and failed experiments quickly lead to new discoveries and new knowledge. The parallel development of custom-purposed instrumentation, capabilities, and data management for automated experimentation will reduce the time, and in some cases cost, necessary to elucidate the design rules for new catalytic processes and manufacturing methodologies. If paired successfully, combined approaches could lead to fundamental knowledge for connecting the scales in sustainable manufacturing and to highly adept artificial intelligence regarding how to direct electrified manufacturing processes in response to specific inputs.

Thrust 2. Minimize entropic losses to enable circular manufacturing systems

Next-generation materials and methodologies that allow for

deconstruction may be driven thermally, mechanically, photochemically, or electrochemically, or by using a combination of inputs and catalysts that are tolerant to common additives and acquired impurities. For metals, it will be important to lay new foundations by which alloys and other inorganic materials may be deconstructed to remanufacturable building blocks, e.g., using new extractive or electrorefining methods.⁵¹ The science of combining reactions and separations for efficient deconstruction and resource recovery in circular manufacturing systems is presently underdeveloped; it could provide new opportunities for managing complexity by controlling bond reconfiguration and molecular separation events. Further inroads to resource circularity may be made through the development of new materials with useful properties, and through reduced complexity to minimize entropy losses in resource recovery from additives or other components added to products. Advances in data analytics and machine learning will be critical to accelerating the co-development of materials and processes with molecular precision by fully elucidating the interdependence of variables as they relate to entropy losses within the closed-loop product life cycle. By analogy to the Carnot cycle, this fundamental yet molecular-level understanding of entropy in circular manufacturing systems is foundational to future successes in the sustainability of resource recovery for reuse.

Advances in circularity are likely to be in step with future manufacturing practices, which are making the shift to digital processes.^{52,53} Catalyzing this shift has been AM and 3D printing, which enables rapid production on any scale of nearly finished parts entirely from digital information. For parts printed from polymers or metals, a burgeoning group of layer-by-layer and extrusion processes are being developed alongside printable material formulations that are overwhelmingly not recyclable. For polymers and composites, it is also possible to print parts using digital light manufacturing (Figure 28).⁵⁴⁻⁵⁷ To ensure the future

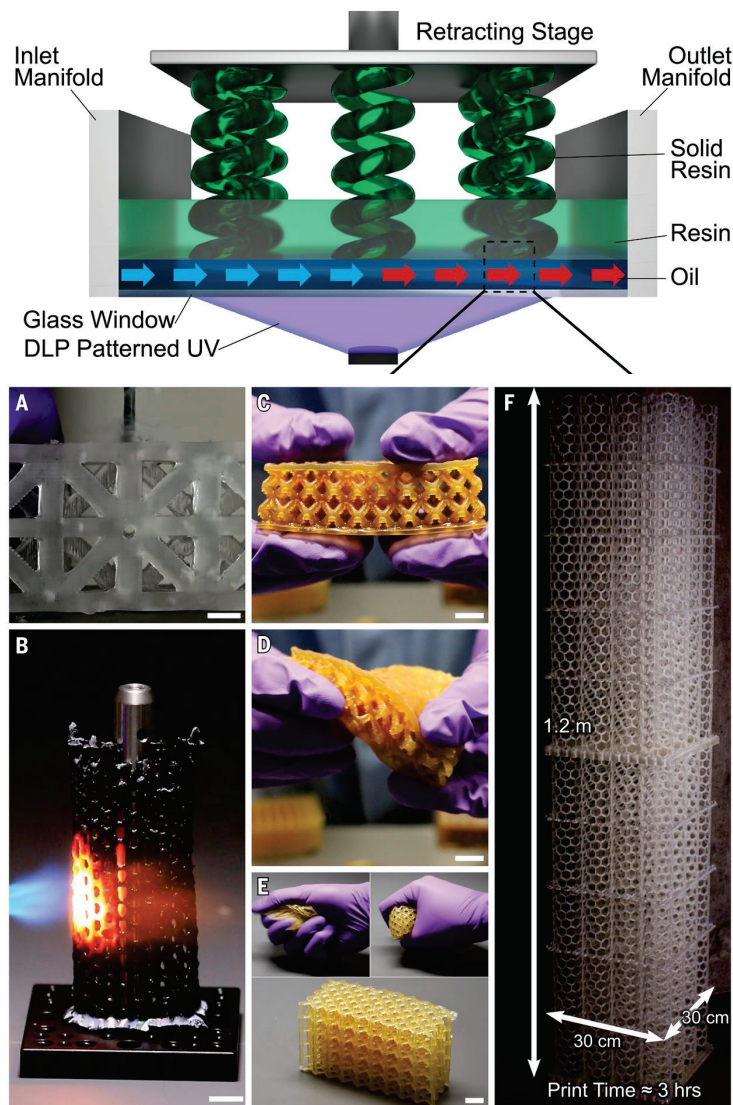


Figure 28. Scheme of a 3D printed part emerging from the HARP 3D printer. (a) A hard, machinable polyurethane acrylate part with a hole drilled against the print direction. Traditional noncontinuous layer-by-layer printing techniques typically delaminate and fracture when drilled in this orientation. (b) A post-treated silicon carbide ceramic printed lattice stands up to a propane torch (~2000 °C). (c–d) A printed butadiene rubber structure in a relaxed state (c) and under tension (d). (e) Polybutadiene rubber returns to expanded lattice after compression. (f) A ~1.2-m hard polyurethane acrylate lattice printed in less than 3 h. Scale bars, 1 cm. Source: From [D.A. Walker et al. Science 366, 6463, 360–364, 2019](#). Reprinted with permission from AAAS.

sustainability of AM,⁵⁸ the molecular design of printable materials for AM must be revisited to unlock circularity. Lossless recovery of the embodied resources will require the development of new materials and chemistries for precise control over transformation, transfiguration, and reversibility of chemical bonds from printing to use to recycling. Innovative technologies also could emerge from the design of new chemistries for both metal alloys and polymers that are resilient to contamination/material mixing during end-of-life processing; development of processes that are resilient to suboptimal chemistries in heterogeneous waste streams; and novel low-energy, high-throughput multiphase separation techniques that can, for example, prevent liquid copper from wetting steel scrap,⁵⁹ remove trace iron from aluminum melts,⁶⁰ or selectively and sequentially recycle polymers from mixed-polymer waste streams.^{61–64}

Thrust 3. Design adaptive methodologies to exploit unconventional, renewable, and recycled feedstocks in manufacturing

Transformative breakthroughs in sustainable manufacturing of chemicals and materials could arise from disruptive technologies that make use of unconventional feedstocks that are either recycled from products at end-of-life, bio-sourced, considered waste, or even the byproducts of combustion. Understanding how to utilize such feedstocks effectively and efficiently in manufacturing is particularly challenging because of their unpredictable heterogeneity, which is typically not accommodated using current manufacturing methodologies. Developing future manufacturing processes that are tolerant of and even adaptive to feedstock heterogeneity during production is therefore critical to sustainable and resilient production, and it may lower water use as well as the carbon and energy footprint of the process by obviating the need for extensive feedstock refinement (Figure 29).

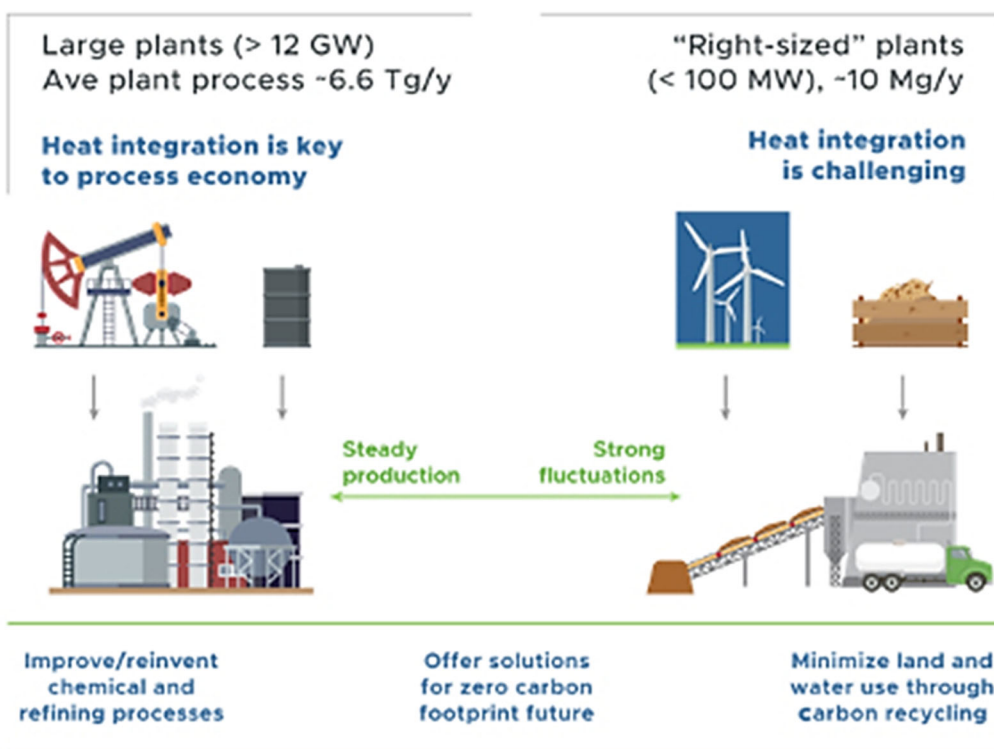


Figure 29. Adaptive manufacturing that incorporates inputs with strong fluctuations and heterogeneity will require innovative chemical processing. Source: Image provided by Pacific Northwest National Laboratory.

The design of new manufacturing methodologies that are adaptive to feedstock source and heterogeneity may take advantage of a variety of energetic inputs or catalysts to direct chemical transformations efficiently, either selectively or specifically. For processes employing a catalyst, tailoring active sites alongside the reactive environment using site-isolation or catalyst confinement in a selectively permeable matrix may unlock new mechanisms for addressing feedstock heterogeneity or variability between production runs. Selectivity may be further influenced by chemical or electrochemical potential on (photo)electrodes, which can be modulated in real time to redirect reaction coordinates to a desired outcome.^{65,66} To understand how such catalyst systems respond to changing operating conditions, energy inputs, and feedstock heterogeneity, it is necessary to advance computation and modeling to unravel the complexity of variables dictating selectivity, yield, stability, and turnover rate. It also may be beneficial to combine cooperative catalysts in responsive and adaptive networks to actively manage feedstock heterogeneity in situ, e.g., by transforming reactive contaminants in an input stream to chemically inert species. Catalysts and networks also could be put to work for circular manufacturing systems, in which new methodologies are needed to create purified metal and polymer/monomer streams that can be processed similarly to virgin resources.

Both the heterogeneity of the feedstock and the likelihood (if not necessity) of smaller-scale distributed processing plants require the development of new fundamental knowledge in adaptive manufacturing that is not currently available. Specifically, the molecular basis for achieving high thermodynamic efficiency and high selectivity at ambient conditions is poorly understood but is of critical importance at small scale. Reactor designs that afford process intensification through enhanced transport or reactive separations may be needed for atom and energy efficiency. Directing primary energy and chemical conversion processes at interfaces to exploit areal kinetics, rather than volumetric kinetics, also may be needed, e.g., in membrane reactors, electrocatalytic reactor systems, and micro- and meso-channel reactor designs in which rates of mass transfer and heat transfer can greatly exceed those typically exhibited by the volumetric reaction systems in use today. Processes using unconventional feedstocks, particular waste or complex recyclates, may be run in condensed multiphase systems in which a greater understanding of the underlying thermodynamics, solvent effects, transport, and kinetics for condensed phase transformations is needed; the theory of condensed phase reactions and processes lags theory for gas–solid systems. Catalyst deactivation mechanisms also differ considerably in the condensed phase. For processes that will be run in gas–solid phases—including processes using H₂, H₂O, O₂, CH₄, CO, CO₂, N₂, NH₃—fluctuations in input remain a challenge that can be addressed in adaptive manufacturing systems. To enable robust manufacturing systems that exploit unconventional, renewable, and recycled feedstocks alongside responsive catalysts and networks in tailored reactor designs, novel sensors and data science will be needed that enable rapid adaptive control, both feed-forward and feed-back, to contend with the fluctuating feeds.

Science And Technology Impacts

Globally, GHG emissions attributed to chemical and petrochemical processes currently amount to ~1.24 Gt of CO₂ equivalents annually.⁷ Discoveries aimed at lowering the intensity of their production through electrification and other means would substantially improve the sustainability of manufacturing systems on a global scale. To cope with fluctuating resources for energy, raw materials, or other resources, future manufacturing facilities that are distributed and smaller in scale may be advantageous and more sustainable. The transition to distributed production could lower both operating and capital costs, as well as promote resilience to changes in feedstock composition or grade, changes or seasonality in market demands, and unavoidable shutdowns due to natural disasters or pandemics. Exploiting the potential benefits of forced variable or periodic operation in distributed chemical and materials production requires breakthroughs that can be achieved only through basic science, as the impacts of the various driving forces on reaction pathways and the kinetics of the transformations remain poorly understood.

A convergence of basic energy research needs in both materials and recycling processes is needed to address outstanding challenges in manufacturing circularity, given the complexity of manufactured products, the unpredictability of waste streams undergoing reprocessing for reuse, and the desire to increase the recycled content of manufactured products to lower GHG emissions. Circularity in manufacturing systems also improves their sustainability by reducing supply chain interdependencies and prioritizing recycling strategies for recovering embodied resources in specific forms. If entropic losses are minimized in such processes, so in turn are the energy and environmental costs, increasing the likelihood that end-of-life products will be collected and processed for recirculation. Compared with business-as-usual, a circular economy of polymers has the potential to reduce the annual volume of polymer waste entering our oceans, reduce GHG emissions and overall energy requirements, and reduce virgin plastic production.⁶⁷ Similar gains have been demonstrated or are anticipated for circularity in metals for steel, aluminum, lead, zinc, noble metals, alloys, rare earths, and feedstocks for building materials, batteries, photovoltaics, fuel cells, electrolyzers, and other energy-converting and storage devices.^{27–29,37–41}

Realizing the technology impact of reinventing sustainable manufacturing will enhance computation, materials, and chemistry understanding. Making strides in the new multiphysics models coupled to in situ characterization capabilities, data analytics, and theory that accurately describe multiphase reactions under potential will provide powerful cross-cutting science yielding insights into kinetics and transport phenomena across scales. New materials for catalyst supports, electrodes, membranes, and materials enabling reactive separations will be realized for production, including refining feedstocks from recyclates for reuse. Additional synergies are expected as new design principles are developed for next-generation hard and soft materials that are intrinsically more circular. As tools and methods emerge for predicting how operating conditions and catalysts must adapt to varying inputs, for understanding how reactive steps couple to form cascades of reactions, for deconstructing complex materials, and for coupling reactions and manufacturing processes to produce new materials, new science will also emerge that will be transformative.

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PRD 5: Co-Design Materials, Processes, and Products to Revolutionize Manufacturing

Summary

Early-stage experimental and computational research often begins with a homogenized material or chemical property that is important for a given application, such as high stiffness, high conductivity, or high reaction selectivity. This property is then explored using a variety of constituents or configurations, usually at a single length scale, to establish processing protocols that achieve the desired values of the target property. The resulting material or process is then considered for use in a specific component in a system that collectively has the functionality needed for a targeted application. As a result, fundamental chemical and materials research is usually a sequential process viewed through a lens of one or two objectives at each scale, with little attention paid to potential coupling between scales, objectives, or components. A powerful alternative to this sequential approach is co-design, in which information sets from multiple phases of research and development are used in an integrated way. The core premise of this PRD is that a rich set of basic science challenges exist that, if addressed, will allow co-design to powerfully improve the materials, processes, and products in many manufacturing settings.

In end-use environments, however, a system is almost always a complex set of interacting components that are combined to create a product or device. In this setting, the behavior of one component often has many impacts on other components and ultimately on the entire system. The overall behavior of a system developed in a sequential manner is frequently impacted by unanticipated consequences arising from a lack of understanding at the interfaces or interconnections between components. Because of the complexity of multiscale and multidisciplinary systems, practical manufacturing processes are often empirically optimized without a fundamental science basis. This situation strongly limits capabilities to adapt processes or materials to situations outside the immediate envelope in which they have been developed.

The descriptions above highlight the critical challenges of finding science-based approaches to integrate multiscale/multiphysics knowledge, including emergent behaviors among interacting system components, to enable informed decisions regarding materials selection and creation. Figure 30 depicts an example situation in which insights are beginning to be obtained into the role of phase change and defects in complex nano- and micro-scale material structures, and their impacts on properties and performance for additive manufacturing (AM) with metals. Similar descriptions also can be given in many chemical and physical settings. Tools are needed to overcome the nonintuitive nature of manufacturing design that involves nonlinear interactions and discontinuities. There is also increasing emphasis on the system's life-cycle behavior through concepts such as maintainability, durability, and resilience. Approaches are needed to account for these long-term behaviors and to connect them with fundamental insights into synthesis and processing strategies. Strengthening the connections between science-based development of system components and multi-objective performance characteristics, such as resilience, will create significant opportunities for agile design and manufacturing.

There is a need to develop fundamental science approaches that enable co-design of molecules and material(s) and/or system(s) involving multiple and competing objectives and trade-offs between many aspects of a system's full lifetime performance goals. Several recent mathematical and scientific developments provide a foundation for tackling this co-design challenge. Mathematical optimization and artificial intelligence offer a plethora of systematic methods to explore high-dimensional nonlinear design spaces with multiple objectives. The introduction of exascale computing can provide the necessary computing power to explore design and processing options in ways that previously were unattainable. Machine learning and uncertainty quantification also will be vital tools in efficiently exploring high-dimensional multiphysics models. These developments, however, will require concomitant advances in

knowledge of the fundamental mechanisms that ultimately control the performance of individual materials and assemblies of components, and advanced tools and methodologies to reliably measure markers of these mechanisms on a wide range of length and time scales.

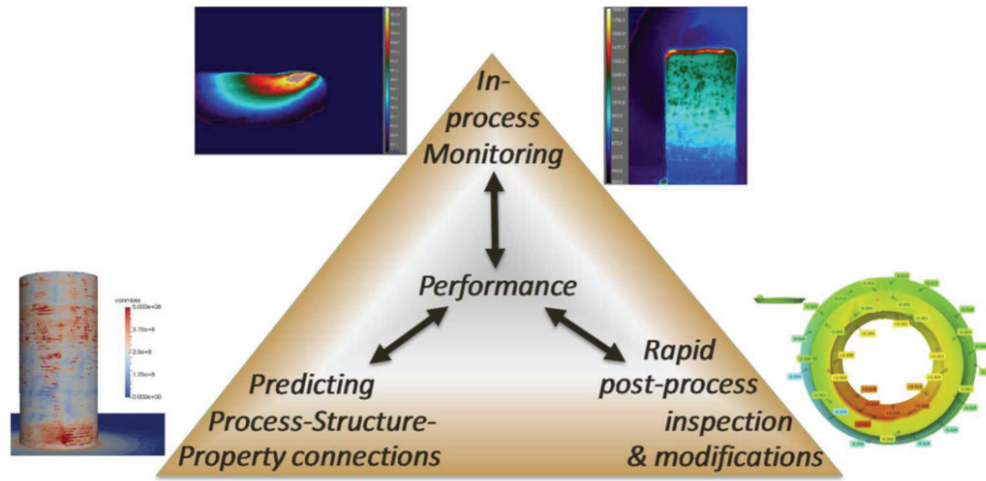


Figure 30. Tight relationship among process-structure-property-performance of a metal-based system for additive manufacturing. Source: Reprinted with permission from R.A. Roach et al., [Using additive manufacturing as a pathway to change the qualification paradigm](#), presented at International Solid Freeform Fabrication Symposium, Austin, TX, August 13–15, 2018.

This PRD seeks to address key questions that include these: *How can bottom-up scientific discovery be combined with top-down system-focused design to identify new and efficient manufacturing modalities? What new approaches will allow the control of matter in the presence of impurities and/or nonequilibrium states? How can science enable multiple performance objectives to be achieved simultaneously for complex, multicomponent processes?*

Scientific Challenges

To address these key questions, a number of scientific challenges must be overcome.

The first challenge includes the need for new, fundamental insights into phase changes, defects, impurities, complex compositions, and nonequilibrium character in processing and their impacts on the performance of the top-level system needs. As materials, molecules, or processes move from discovery to application and manufacturing, multiple complicating factors tend to arise that risk becoming “showstoppers.”¹ Examples of these factors include impacts of defects or trace impurities, particularly in multi-component or nonequilibrium materials, and long-time scale processes such as aging. A key aspect of basic science approaches for co-design is to both understand and predict these influential factors at the systems level. Doing so could lead to new strategies for control over the behavior of materials and systems, e.g., creating metastable states with unique properties. This challenge is especially important in efforts to develop products that are characterized not by individual physical properties (e.g., high thermal conductivity) but by broader goals of manufacturability, systems-level performance needs, and life cycle requirements. Fundamental insights into these complex characteristics are likely to lead to unexpected discoveries of excellent performance through simultaneous manipulation of multiple structure-composition-processing parameters.

Additionally, approaches are needed to integrate uncertainties associated with interacting components across multiple length scales, “from beaker to barrel” and “from atoms to attributes,” to quantify robust co-design. Although multiscale approaches have been a mainstay of basic research for many years, the

enormous diversity of scales that are relevant in manufacturing settings is daunting. The concepts of uncertainty and robustness in developing new systems are closely related. It may often be preferable to develop a system that is more robust against variations in manufacturing conditions or operating environments, rather than one that nominally has higher performance but requires far tighter bounds. There is a great need for systematic approaches that can translate information about uncertainties and robustness in a predictive manner, from short time and length scales that are accessible with high fidelity across orders of magnitude, to the longer and larger scales relevant in manufacturing. This task goes far beyond simply propagating experimental uncertainties toward quantitative, scientific measures of systems-level behavior and the relevant contribution from each interacting component or process choice. Enormous opportunities exist not only in the development of new products but also in new manufacturing processes. Fundamental developments in processes can lead to dramatic shifts in manufacturing.²

It is important to acknowledge that new manufacturing technologies require research and development to control and manage the uncertainties in their end products. Research on this topic involves experimental measurements and large simulations, which result in huge quantities of data to store and analyze. This challenge needs to be addressed by alleviating data storage requirements via continuously developing accessible data compression methodologies and efficient data sharing protocols and platforms.³ Furthermore, there is a need for real-time control and error-correction methods over multiple length and time scales through analysis of these data at the speed of manufacturing processes, as discussed in PRD 2.

A third scientific challenge is achieving bottom-up design across many time and length scales or systems-focused co-design from system scales to achieve inherently resilient systems. The aspects of a system that make it resilient differ among manufacturing sectors; but in every sector, resilience is an overarching factor in successful systems. In today's world, resilience is often sought or added at later stages in a research path. There are opportunities for creative research, however, to develop broad approaches that make chemicals or materials inherently resilient. These approaches could include bottom-up design in which resilient properties cascade from small to larger scales or emerge naturally at larger scales. Finding active techniques to express the needs on small scales, via descriptions of system-scale properties by system-focused design, also would reap huge benefits. Resilience can be a property of manufactured products, but it is also an important goal in manufacturing processes. The notion of resilient processes is very important, for example, in chemicals processing, where feedstocks can include enormous numbers of components with composition that varies over short and long periods of time.⁴

Another challenge includes exploiting strong feedback between experimental data and modeling in high-dimensional domains to achieve co-design of multifunctional systems, including information from both successful and "failed" experiments. Iterative interactions between modeling and experiment are already commonplace. The prevalence of "big data" tools has great potential in co-design of systems, but an Edisonian application of these methods is unlikely to lead to significant breakthroughs. When considered on the full spectrum of relevant length and time scales, multifunctional systems are characterized by high-dimensional domains in which data are sparse and often highly uncertain. Important challenges exist in developing approaches to this class of problems that make appropriate use of (multi)physics-based models or constraints and that recognize the resource costs associated with various kinds of experiments. At the same time, techniques allowing strong coupling and feedback among experimental data—including data obtained from diverse sources—and computationally driven search methods are greatly needed to explore the co-design space and to identify high-value experiments or data gaps. Examples of an approach like this for materials and molecular discovery are shown in Figure 31 and Figure 32, respectively.

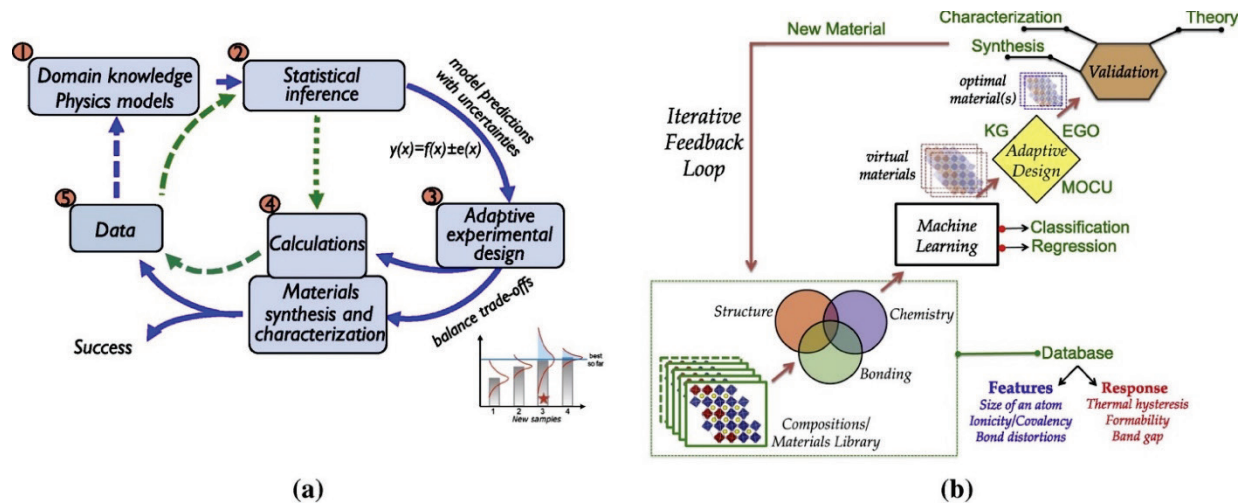


Figure 31. (a) The adaptive design loop where the key challenge is to minimize the number of iterations it takes to discover a new material with desired properties by finding a reliable surrogate to the true unknown function $f(x)$. Existing work on materials design is largely based on following one or two of the green arrows. The approach introduces the adaptive experimental design step that uses uncertainties, $e(x)$, to balance the trade-off between exploration and exploitation in suggesting the next experiment or calculation to be performed. (b) The loop in practice, where EGO (efficient global optimization), KG (knowledge gradient), and MOCU (mean objective cost of uncertainty) are acquisition functions or selectors that choose the next optimal experiment or calculation. Source: [T. Lookman et al., Curr. Opin. Solid State Mater. Sci. 21\(3\), 121–128, 2017.](#)

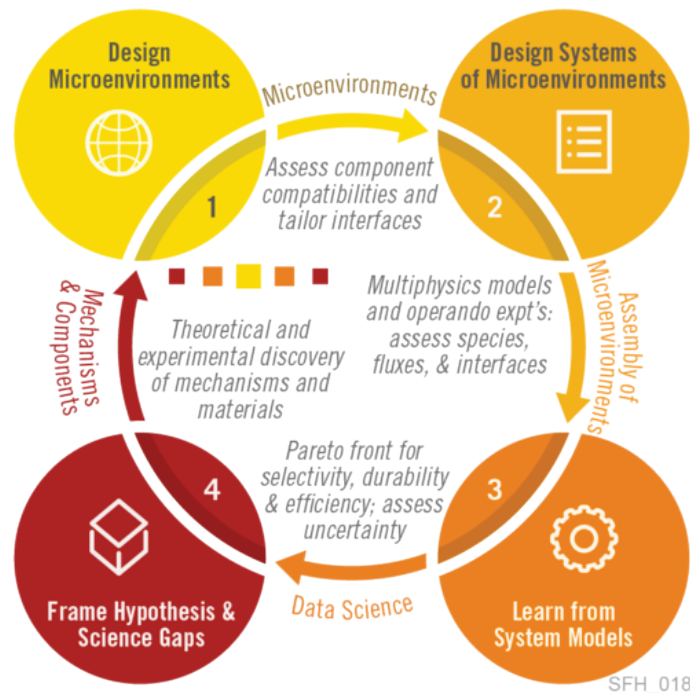


Figure 32. Components-to-systems loop is closed by data-driven hypothesis generation in DOE's Liquid Sunlight Alliance Hub. Source: Used by permission from the Liquid Sunlight Alliance (LiSA) and LiSA/Caltech, liquidsunlightalliance.org/co-design.

Finally, this PRD challenges the scientific community to use co-design to generate materials and systems with superlative performance properties. Innovative combinations of knowledge from multiple stages in the development of manufacturing processes could unlock examples of outstanding performance that

cannot be anticipated by more sequential approaches. Broadening even further, a co-design approach brings with it the grand challenge to simultaneously consider manufacturability, performance, reliability, resiliency, and sustainability, which are at the heart of all the PRDs combined.

Research Thrusts

Thrust 1. Achieve knowledge discovery of structure-performance-process relationships across many length and time scales to expand options for co-design.

By their very nature, systems are assembled from multiple components, which may in turn contain individual chemicals and materials or, more commonly, complex mixtures of constituents. To allow the broadest range of possibilities for co-design, it is vital that knowledge of performance relationships for each component overlies the conditions used to manufacture or operate other components. Many examples exist in which simplified metrics used to screen materials for particular uses are found to give at best partial information when insights from more detailed process-level descriptions are used.⁵ Structure-performance-process relationships have long been a goal of chemical and materials science, but the scope at which these relationships are examined needs to expand dramatically to fully realize systems co-design. Research progress is needed to reliably extend the structure-process relationships available with current methods to far broader ranges of operating environments, and to allow active interactions with other materials classes and components without simply sampling the vast search space defined by these conditions.

Thrust 2. Realize multiscale co-design approaches for desired behaviors and decision-making at systems scales.

Co-design is often thought of as a staged process in which information flows from “smaller” to “larger” scales. In this environment, it is not necessary to achieve a perfect outcome at each scale before proceeding. Instead, a key goal is to make more reliable decisions about options at the next scale. Developing science-driven ways to allow system-focused design, in which desired properties from the system scale drive exploratory work at smaller scales, has immense potential. Success in this domain cannot arise simply from formal methods. Rather, creative efforts to incorporate learnings from broad classes of materials or examples of interacting components must inform co-design strategies. It is likely that success in these endeavors will emerge initially from focused case studies that combine investigators with diverse expertise.

Although the concept of system-focused co-design has many facets, a particularly challenging area is nonequilibrium processes. If fundamental principles can be used to better understand these processes from the viewpoint of chemical and materials synthesis, it will be possible to exploit the emergent phenomena from these studies in a manufacturing context. Advances in this area will create the integrated modeling and prediction necessary for advanced materials synthesis and device-level operation in many kinds of materials, including but not limited to metals and ceramics.⁶

Thrust 3. Elucidate formation pathways of matter in equilibrium and nonequilibrium states, including the impacts of defects, impurities, multiple length scales, complex processes, and rates to identify influences on systems performance.

Complex assemblies of molecules and materials in multifunctional systems can vary over the multiple time scales associated with manufacturing steps, and over the much longer time scales associated with their life cycles. In both cases, many opportunities exist to describe the pathways associated with the formation and evolution of matter at fundamental levels. In many instances, crucial system-scale impacts arise from highly localized events, such as the deliberate or unavoidable incorporation of defects or interactions with trace contaminants. Preparing matter in nonequilibrium states is a powerful tool for creating design options, but these kinds of materials are subject to evolution on multiple time and length

scales. Mechanistic insights that go beyond observational records for nonequilibrium states would have great value. It is perhaps tempting to view aging and degradation as engineering challenges to work around; but if phenomena of this type are understood fundamentally, then it will be possible to mitigate these challenges in powerful ways during co-design. Mechanistic studies of these processes can also open up previously unanticipated avenues for producing new properties.⁷

Thrust 4. Impart tunability, durability, and resilience by understanding uncertainty associated with interacting components in multi-material, multifunctional assemblies and processes.

Tunability, durability, and resilience are indirect properties that are far harder to quantify than directly observable characteristics such as electrical conductivity. Many open questions exist in quantifying how properties directly measurable at the small scales typically accessed in basic research can be used to predict and ultimately control these indirect properties at the systems level. A core challenge in this area is to understand what kinds of uncertainties can arise at each scale and how these can be manifested at large scales in more complex assemblies. In this context, it is important to note that, at a manufacturing level, it can be just as critical to control the distribution of properties in a system as it is to maximize the performance of the “best” possible individual outcome.^{8,9} Research opportunities exist in using diverse sources of experimental and modeling data to meaningfully predict the distribution of performance expected in systems operated under conditions outside those for which direct tests are possible. One avenue for this research will be to use fundamental insights to place empirical ideas in accelerated testing on a firmer footing that can reduce the overall time required to test complex systems.

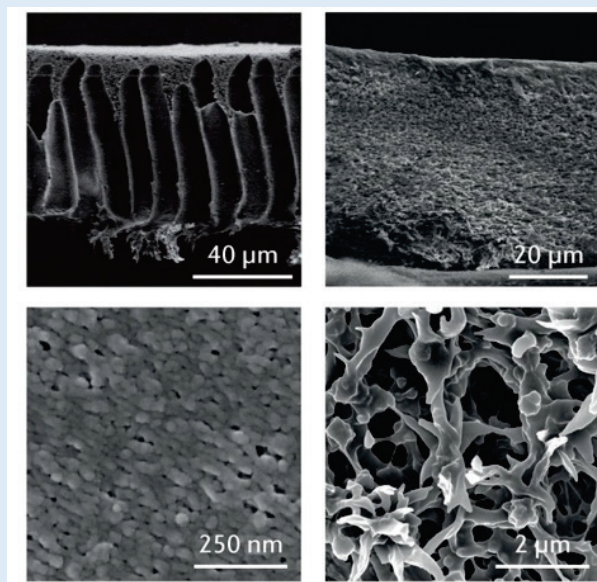
Science and Technology Impacts

The primary outcome of this PRD will be science-based tools for design, scalability, and manufacturing of complex systems where the level of system complexity is far beyond human intuition. Current empirical and experience-based design and optimization for one or two objectives or functionalities cannot account for nonlinear coupling among multiple components, emergent behavior across the scales, or the significance of uncertainties in the overall system behavior. Historically, the design evolution of complex systems typically required decades to achieve energy-efficient and widely deployable systems. Fundamental principles and science-based tools for co-design will have an immediate effect on the development of energy-efficient systems that are inherently resilient and durable. An example is shown in the sidebar “Improvement in Water Desalination.” Top-down co-design approaches will reveal new fundamental questions about interactions among components in complex systems; and mechanistically grounded tools will provide agility, allowing rapid response to changing needs in manufacturing. These approaches will also provide opportunities to lean in to complexity, thus achieving energy and time efficiencies in the development of new chemical- and materials-based technologies. An example of what is possible is shown in the sidebar “Designing Complex Materials.”

Advanced modeling tools will be vital to harnessing the complexity of transformative manufacturing. Modeling across the many relevant scales will enable a fundamental understanding of materials design and formation, allowing successful design of reliable materials for transformative manufacturing. First principles modeling will be enabled by extension to broader scales (via, for example, ab initio molecular dynamics) to model real-world systems, including contaminants, co-solvents, and so on.¹⁰ Information from modeling, especially as made possible using exascale computing resources, will enable system-wide quantification of manufacturing uncertainty. Advanced computational methods development will be needed for this focus, and BES computational user facilities will play a critical role in adapting these methods to more complex, real-world condition predictive modeling.

Sidebar: Improvements in Water Desalination through a Co-Design Approach

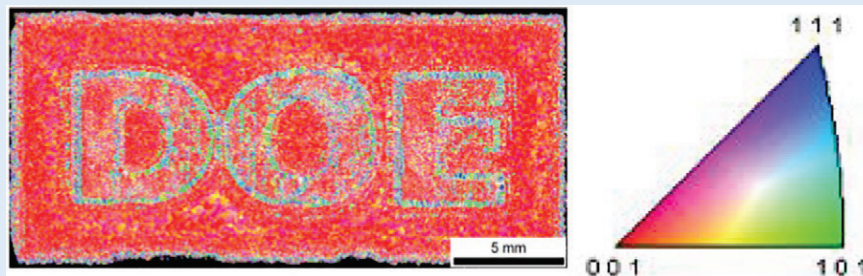
The energy efficiency of membranes for water desalination, relative to alternative strategies such as distillation, have led to membranes dominating worldwide desalination installations. One simple metric that quantifies the performance of a membrane is its permeability. Higher permeability means higher throughput, and this observation underlies the fact that increasing membrane permeability is often a central research goal in this field. Process-level models, however, have shown that increasing permeability above levels that are already commercially feasible has at best a small impact on the energy use or capital cost for water desalination.¹¹ These models indicate that more impact can be gained by focusing on developing more selective membranes, even though existing membranes already reject well over 99% of salt under typical conditions. This is one of many examples of how careful consideration of process modeling can allow fundamental research to focus on performance metrics that will have the greatest long-term impact, especially when those metrics are not intuitively obvious.



Scanning electron microscopy (SEM) images of membranes formed from phase inversion. Upper left: a cross-section of an asymmetric ultrafiltration (UF) membrane with finger-like macrovoids cast from 9% polysulfone (PSf) in dimethylformamide (DMF). Upper right: a cross-section of an asymmetric UF membrane with a sponge-like structure cast from 12% PSf in DMF. Lower left: a top view of a hand-cast PSf UF membrane. Lower right: a top view of a commercial 0.22 μm nominal pore size polyvinylidene fluoride microfiltration membrane. Source: Reprinted with permission by Nature Publishing Group: *Nat. Rev. Mater.*, J.R. Werber et al. [Materials for next-generation desalination and water purification membranes](#), 1, 16018, 2016.

Sidebar: Designing Complex Materials by Combining Understanding of Material Properties, Processing, and Assembly

Transformative manufacturing will require a holistic approach to materials, processes, and production designs. Cutting edge integration of optimization tools and additive



Control of grain texture in AM Inconel 718 via electron-beam melting.

manufacturing processes with holistic system-focused design paradigms will enable the concurrent optimization of design topologies, material constructs, and assembly methods. To this end, optimization design tools are experiencing a recent revival.¹² Additive manufacturing offers unprecedented opportunities to design complex structures optimized for performance envelopes inaccessible under conventional manufacturing constraints. It can also promote the realization of engineered materials with microstructures and properties that are impossible via traditional synthesis techniques.

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4. Panel Reports

The Basic Research Needs Workshop for Transformative Manufacturing was structured around six panels, including a panel focused on crosscutting themes.

Panel 1: Precision Synthesis Science

Panel 2: Processing and Scale-up Science

Panel 3: Systems Integration Science

Panel 4: Sustainable Manufacturing

Panel 5: Digital Manufacturing

Panel 6: Crosscutting Topics

The panel reports formed the basis for identifying the five PRDs described in Chapter 2.

Panel 1: Precision Synthesis Science

Introduction

Precision synthesis of materials through deposition, assembly, or processing leading to deterministic organization from the atomic to the macroscopic scale—particularly when driven by information content built into the basic constituents—is the *ne plus ultra* of materials science. Because phenomena like electron conduction, spin coherence, photon adsorption, charge separation, and phonon transport are intimately connected to material architecture from the atomic to the mesoscale—including the arrangement of defects and interfaces—a science of precision synthesis will deliver unprecedented chemical, thermal, optical, electrical, magnetic, and mechanical functions. Recent advances in chemical methods are enabling the atomic precision so prized in molecules to be translated to the synthesis and assembly of larger, more complex organic, inorganic and hybrid materials. When combined with recent advances in high-performance computing—both for simulating synthetic processes through physics-based models and for applying machine learning (ML) and artificial intelligence (AI) to predicting structure-function and synthesis-structure relationships—the possibility of precision synthesis across scales lies within our grasp, provided fundamental challenges in controlling and scaling synthetic processes can be overcome. This panel explored the current status of and challenges and opportunities associated with experimental, computational, and combined experimental-computational approaches in precision synthesis science. The findings are broadly grouped into two themes:

- *Advancing Autonomous Synthesis and Processing for Materials Manufacturing*
- *Achieving Materials with Designed Functions and Structures Via Atomically and Molecularly Precise Synthesis and Processing to Transform Manufacturing*

In both cases, the panel’s focus was guided by the recognition that the intersection of precision synthesis science with transformative manufacturing places an emphasis on concepts of scale-up, high-throughput processes, feedback through real time in situ diagnostics and analyses, and materials systems and integration across scales.

Current Status and Recent Advances

Theme 1. Advance autonomous synthesis and processing for materials manufacturing

1. Exploit advances in physics-based models, theory, and computation for ML/AI-constrained synthesis and processing

A grand challenge for synthesis and processing would be to predict and make, with atomic-scale precision, a given material or molecule with a prespecified function. Recent advances in physics-based models, theory, and computation through ML/AI-constrained synthesis and processing have brought us closer to this goal and created several future research opportunities. Automated high-throughput ab initio calculations have rapidly facilitated rapid materials synthesis and discovery of a number of new functional materials.¹⁻⁵ A predictive “grand unified theory of materials and chemical synthesis” for synthesis and processing does not exist; but a combination of automation and interpretable ML techniques to create AI-driven synthesis-driven platforms has the potential to considerably advance existing brute-force serial and combinatorial screening approaches, provided enough data are available for the models.

2. Develop high-throughput synthesis, processing, and characterization techniques

In the past 20 years, high-throughput experimentation (HTE) has been used by the chemical industry to speed up the research and development process. Examples include the accelerated development of new pharmaceuticals,⁶ coatings⁷ and thermoplastics.^{8,9} Large industrial HTE platforms typically include rapid synthetic and analytical workflows enabled by fully integrated automation and database software. Methods such as design of experiments are often implemented to cover a broader swath of variable space. More recently, ultraminiaturization has been applied to increase throughput and decrease costs.¹⁰ Advances in flow chemistry have been paired with HTE for use in ultraminiaturization and to permit real-time adjustment of reaction parameters.¹¹ Moving forward, as shown in Figure 33, the integration of computational chemistry and AI with HTE and automation offers the promise of truly integrated and autonomous research platform.^{12–14} These advances are very recent and have only been applied at the proof-of-concept level; but, when combined with large integrated HTE workflows, they may ultimately provide a very powerful toolkit for industrial research and development.

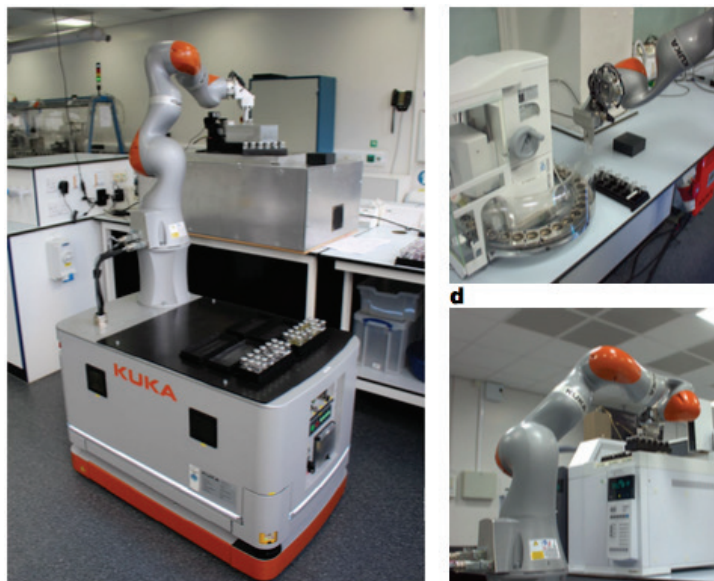


Figure 33. Autonomous mobile robot and experimental stations. Source: Reprinted with permission by Nature Publishing Group: *Nature*, [A mobile robotic chemist](#), B. Burger et al. 583, 237–241, 2020.

3. Identify structure and function signatures for real-time, autonomous manufacturing

Automated synthesis with real-time feedback control was identified as a stretch goal. Arguably, the ability to apply that level of “smart” automation to synthesis and processing in manufacturing is an even more ambitious target. Requirements would include a deep understanding of correlations between each step of the synthesis or processing step and a resulting outcome for the product in terms of manufacturing-relevant elements of structure or function. Establishing that knowledge could be aided by developing physics (chemistry)-based theories and models, as discussed in Theme 1, item 1, which could form the basis for new ML/AI tools for that synthesis or processing step. Alternatively, experimental training sets could be used to develop the desired algorithms for synthesis control. In either case, however, real-time algorithm-directed control will need real-time feedback from the system to inform its next step.

Theme 2. Achieve materials with designed functions and structures via atomically and molecularly precise synthesis and processing to transform manufacturing

1. Develop new scalable synthetic routes to organic, inorganic, and hybrid materials

Chemists have long sought to develop methods and strategies to synthesize materials with precise structures. Much progress has been made in the precision syntheses of, for example, polymers,^{15–17} nanomaterials,¹⁸ and composites,¹⁹ including extending control over composition and phase to achieve exquisite manipulation of dimensionality (0D, 1D, 2D), surface and interface properties, and other characteristics.^{20,21} However, challenges remain in maintaining the same level of precision when these

reactions are carried out at large scale. Furthermore, “scalable” can have very different meanings depending on the material. For many new quantum technologies, “scalable” means deterministic placement, organization, or patterning at the level of atoms or molecules and transitioning from exotic systems to solid state materials. Of the scalable methods for synthesis of classical materials, there are a few stand-out techniques, such as continuous flow synthesis with or without droplet microreactors,^{22–24} spray pyrolysis,²⁵ chemical vapor deposition,^{26,27} scanning tunneling techniques,²⁸ and 3D printing,²⁹ that show potential. Opportunities exist to enable the large-scale synthesis of greater compositional and architectural complexity and precision. Quantum materials³⁰ present a particular challenge, and new routes are needed for their scalable synthesis.

2. Understand the chemical and physical forces that direct the synthesis and processing of molecules and nanomaterials to realize targeted functions

Ideally, manufacturing of atomically and molecularly precise materials would be based on a deterministic understanding of the final structure and function obtained from a defined set of constituent building blocks, whether they consist of atoms, molecules, polymers, particles, or some combination of these disparate units. Moreover, the desired end state may well be nonequilibrium and require control over the formation pathway to end in a kinetically trapped state. Compounding the challenge in deterministic manufacturing of atomically and molecularly precise materials is the need to understand how co-varying factors associated with the manufacturing process itself, such as temperature, surface chemistry, and rate of reaction, impact the pathways and outcomes of synthesis and processing. Significant strides have been made in understanding—and thus predicting—phase diagrams for many systems of interest, such as colloidal arrays, block copolymer films, and epitaxial heterostructures. However, formation pathways, kinetically accessible nonequilibrium states, systems of disparate building blocks, and the impact of manufacturing processes lie beyond our current ability to predict either outcomes, pathways, or rates. A major challenge also lies in quantitatively accounting for the effects of surfaces and interfaces on both the materials processing and the resulting function.

3. Create and translate the scientific understanding of spatial and compositional control down to atomic scales in 0D, 1D, 2D, and 3D (and with arbitrary design) at manufacturing-relevant scales

With the push to realize atomically and molecularly precise, 0D, 1D, and 2D building blocks; understand their structure and function; and scale their synthesis to manufacturing scales, mastery of precise spatial and compositional control and subsequent organization and integration is required for a broad range of energy technologies. Bottom-up assembly enables the organization of nanomaterials at subnanometer-scale resolution, often by (weak) van der Waals interactions. However, assembly techniques often yield architectures that are close-packed and lack the flexibility to form sparse and open architectures with the exquisite geometrical definition possible at larger scales by top-down methods of lithography. Overcoming these challenges requires fundamental understanding of the chemical and physical forces that are at play and that can be introduced to direct the assembly of nanomaterials.

4. Determine structure-function relationships across multiple scales, in complex organizations, and at interfaces

New and superior function can be derived when materials are organized across multiple scales, with hierarchical ordering and with interfacial complexity. Current examples are inorganic solid electrolytes with superionic conductivity realized using superatoms as building blocks for artificial crystals,³¹ catalysts with superior transport properties and efficiency achieved via metal organic frameworks with ordered macro- and micro-pores,³² superconductivity obtained in 2D layered heterostructures with precisely controlled inter-layer rotation,³³ energy-efficient separations realized using atomically thin crystals with atomically precise pores (see Figure 34),³⁴ and quantum technologies achieved by positioning multiple

nanomaterials in junctions and cavities.^{35,36} The complexities that emerge over multiple scales and at interfaces are often beyond human intuition, and the phase-space is too large to be explored by trial-and-error. Accelerating discovery will require inversion of the make-then-measure mode of research. Knowing a priori the hierarchical structure, organization, and interfaces needed to obtain desired properties will be critical to increasing the rate and efficiency by which new materials can be manufactured and phenomena realized.

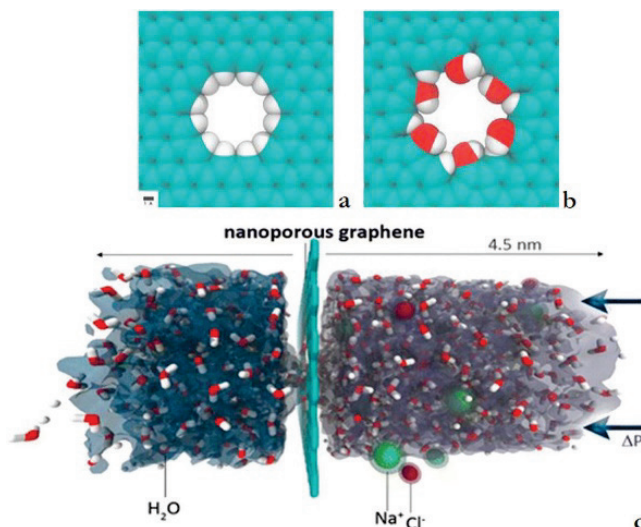


Figure 34. The images provide an example of how new and superior function can be achieved when materials are organized across multiple scales, with hierarchical order, and with interfacial complexity. Materials with remarkable fluid and chemical transport properties have been designed and created from 2D materials building blocks only a single atom thick, with atomically precise pores, and from 2D materials laminates with precisely controlled interstitial spaces. Source: Reprinted from [A. Boretti et al. *npj Clean Water* 1, 1–11, 2018](#). Licensed under a [Creative Commons Attribution 4.0 International License](#).

Basic Scientific Challenges and Opportunities

Challenges

Theme 1. Advance autonomous synthesis and processing for materials manufacturing

1. Access nonequilibrium, kinetically trapped metastable materials structures and compositions

The discovery and manufacture of complex materials and materials structures—with multiple, and often competing, functionalities—can require as-yet unidentified synthetic routes and processing chemistry. Targeted synthesis of multi-component, hierarchical, and nonequilibrium materials requires exploring and mastering enormous phase spaces defined by material composition and processing history; efficient strategies are needed to predict, explore, and navigate complex materials and processing spaces. Because of recent developments in automated synthesis and data collection approaches,^{37,38} advances in data repositories and platforms,^{39–41} and new theory and simulation approaches,^{42,43} physics-informed autonomous experimentation now has strong potential to revolutionize materials discovery, facilitating the study and ultimately the manufacture of ambitious classes of materials. Targeted design principles of system engineering will allow not only design of required building blocks but also, critically, fabrication pathways, as synthesis routes for many desired materials are unknown. There are now opportunities to create steered synthesis approaches reliant on AI/ML that provide routes to nonequilibrium materials. Further research in this area can deliver fundamental knowledge for future digital manufacturing technologies that will similarly rely on active computation and control of processing.

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2. Construct large data sets based on experiment and prediction that adequately capture kinetic and thermodynamic parameters

Critical to autonomous synthesis approaches is the accessibility of relevant data—via rapid, on-the-fly acquisition or via real-time connectivity with curated databases—of sufficient quality and quantity to guide robotics-coupled AI approaches based on ML algorithms. Current robotic automation offers precise control over reaction conditions for certain classes of materials, as well as the ability to rapidly and systematically vary reaction parameters. Thus it can generate reliable and comprehensive data sets from which to discover and design atomically defined functional materials.⁴⁴ Prior examples include core-shell nanocrystals for LED lighting,⁴⁵ upconversion of nanoparticles with atomically precise placed lanthanide dopants for bioimaging,⁴⁶ and metal-organic framework photocatalysts for chemical transformation and water splitting.⁴⁷ Future work allowing for the statistical quantification of the total experimental space and for the likelihood of crystal formation would drive the synthesis of ever-more-complex atomically defined materials and materials structures. New automated capabilities for synthesizing multicomponent structures could dramatically accelerate time-consuming, multi-step synthesis workflows; and AI-guided synthesis could generate meaningful data for increasingly complex materials under different conditions by intelligent sample screening and optimization.

3. Obtain actionable signatures for real-time analytics to support autonomous synthesis and processing

A key challenge for applying autonomous synthesis and processing in manufacturing is to identify a real-time assessable “signature” or measurement observables associated with the desired product outcome. Complexity arises when the outcome is, for example, not an easily characterized structural feature, such as a single atomic defect, or is a multi-component and/or multi-functional product. In these cases, designing diagnostic-model feedback loops will require uncovering the key, detectable signature of the targeted outcome. Using AI-enabled autonomous synthesis to develop new syntheses or discover new materials is a clear science-of-synthesis challenge; but in manufacturing, the challenges lie instead in the areas of process control and improved efficiency minimization through reduced reagent consumption, waste, and/or the ability to make use of less-pure feedstocks. With the advancing robotic synthesis approaches, and advancing data acquisition and curation platforms, there is now the opportunity to develop protocols to acquire or mine relevant data to enable AI control of complex molecular or materials synthesis/processing relevant to manufacturing. These studies also will be in a position to map out relevant structure/function signatures for real-time feedback process control. Addressing critical questions—such as whether ML/AI-driven synthesis also could be used for faster discovery of pathways-to-product (including precursor development) with improved efficiency and optimal yield—will help validate the usefulness of the autonomous synthesis and processing approaches to transform manufacturing.

Theme 2. Achieve materials with designed functions and structures via atomically and molecularly precise synthesis and processing to transform manufacturing

1. Design materials and products and those used in manufacturing processes for two or more materials properties

Basic research needs in precision synthesis and in the context of manufacturing often incorporate goals and objectives related to fundamental understanding of how to impart two or more often orthogonal properties into a single material. Remarkable materials with two or more application-specific manufacturing-oriented properties have been developed by iterating around the chemistry of a known material possessing one critical property. However, this approach tends to explore and develop understanding of only local solutions to design problems, whereas those problems present substantially more complicated landscapes for which global exploration and understanding are often required.

Materials genome and ML approaches and related research needs highlighted elsewhere in this report will undoubtedly play a major role in materials design for two properties: identifying parameter spaces over which to effectively search, and efficient experimental design. However, knowledge gaps continue to exist in developing methods of high-throughput precision synthesis and characterization to use in conjunction with computational models and tools.

2. Create interfaces between dissimilar materials

The combination of dissimilar materials at the atomic scale (for example, see Figure 35) offers opportunities for structures and devices with new function. In many applications, such as electronic,

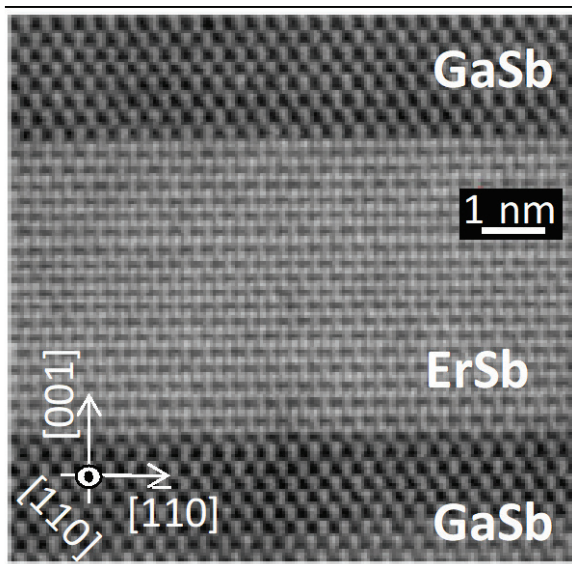


Figure 35. Transmission electron microscopy image of an epitaxial semiconductor/semi-metal/semiconductor heterostructure grown by molecular beam epitaxy. This is a heterostructure of two dissimilar materials: a zinc blend, covalently bonded semiconductor and an embedded rock salt, an ionically bonded semi-metal creating an embedded metallic layer. Source: Used by permission of Chris Palmstrom, UC–Santa Barbara

optoelectronic, magnetic and spintronic devices, the interface properties are the essential ingredients. Future applications may involve creating and controlling topological surface and edge states at interfaces with potential applications in topological quantum computation and quantum information systems.⁴⁸ Examples include Majorana fermions⁴⁹ and other more exotic states such as parafermions.⁵⁰ The challenge in creating the interfaces with desired properties is controlling the interfacial bonding, interdiffusion, reactions, and defects. It is particularly difficult to predict the interfacial structure between materials with different bonding and crystal structure. For example, what is the nature of the association between covalently bonded and ionically bonded materials? Can interfacial bonding layers be predicted and realized? Can general rules for creating interfacial bonding layers and surfactants at the atomic scale be generated that would allow disparate inorganic materials to be combined into artificial structures with novel properties? Can the properties be predicted? These are both experimental and theoretical challenges requiring an atomic-level approach. Current growth processes such as molecular beam epitaxy, pulsed laser deposition, metal organic vapor phase epitaxy, and atomic layer deposition processes—combined

with in situ scanning and other surface science probes, such as angle resolved photoemission spectroscopy—offer the precise control and feedback needed to address these challenges.

3. Enable control of surface chemistry to direct synthesis and processing and tailor materials functions

Surface chemistry will play a key role in transformative manufacturing. The ability to direct chemical reactions on surfaces with atomic-level control over composition, position, and phase will enable atomically precise manufacturing of materials for energy. Surface chemistry underpins many technologies used in semiconductor manufacturing, including chemical vapor deposition, atomic layer deposition, and atomic layer etching. Selective growth of materials with the ability to tailor the composition and phase will allow for the synthesis of novel catalysts, energy harvesting devices, and materials for energy storage and energy conversion. Patterns created on surfaces using scanning probe or scanning beam methods can be amplified by selective surface chemistry on the patterned regions to grow complex materials and devices. Surface chemistry is a versatile strategy to functionalize structures created using other

techniques, such as additive manufacturing (AM), to impart the desired electrical, optical, or chemical properties. Templated growth is a method to create atomically precise pores and cavities through surface chemistry for applications such as shape-selective catalysts⁵¹ and selective sorbents for water treatment.⁵² Spatial control over surface chemistry, either in solution or through reactive chemical vapors, can be achieved using directed beams of photons or ions to provide the energy needed to overcome reaction barriers.⁵³ These methods are employed to create 2D patterns by lithography,⁵⁴ or 3D structures using techniques such as multi-photon polymerization for high-resolution AM.⁵⁵

4. Enable high-throughput, massively parallel, positioning of materials over multiple scales down to the atomic scale

Precision synthesis has demonstrated the ability to produce atomically precise structures in a wide range of materials via a wide variety of approaches, including chemistry, self-assembly, and top-down controlled assembly. However, transformative manufacturing must create complex systems comprising a hierarchy of components and subsystems. In the atoms-to-microns regime, there are promising developments in bottom-up self-assembly such as DNA origami⁵⁶ and other biomolecule-enabled assembly processes.⁵⁷ These approaches have the potential for massively parallel assembly, but they currently are limited by increasing error rates as the size and complexity increases. Top-down controlled assembly processes at the atomic scale have recently demonstrated that useful structures can be produced with an additive approach,⁵⁸ but serial processes operating at the atomic scale are simply too slow for all but the extreme end of the high-value spectrum of products. Going massively parallel with micro-electromechanical system nanopositioners is promising for top-down approaches. As the atomically precise top-down assembly approaches begin to create nano-electromechanical system nanopositioning devices, the level of parallelism can scale dramatically.

5. Manage defects

Defects in manufacturing associated with either materials or processing can take on many roles and must be managed accordingly. Atomic and molecular defects in crystals have long been prized for the colors they create in minerals, including precious stones such as ruby ($\text{Cr}^{3+}:\text{Al}_2\text{O}_3$) and sapphire ($\text{Fe}^{2+}, \text{Ti}^{4+}:\text{Al}_2\text{O}_3$), laser materials such as ruby and neodymium yttrium aluminum garnet ($\text{Nd}:\text{Y}_3\text{Al}_5\text{O}_{12}$, YAG), and spin qubits, such as the nitrogen-vacancy pairs in diamond quantum emitters capable of photon indistinguishability,⁵⁹ which address emerging needs in quantum information science. However, computational and experimental methods are needed to design and create defects for quantum information science, to identify compositions that create suitable electronic structures, and to position defects with accuracy in junctions and cavities to form charge and spin qubits or photonic nodes.⁶⁰ Geometric and topological defects also can be advantageous. While most materials and systems are designed to be periodic, introducing defects that break symmetry can yield materials with “exotic” properties.⁶¹ Geometric defects have been used to design optical, acoustic, and mechanical materials with unconventional or superior properties.^{62,63} For example, advances in targeted design methodologies used to design optical metamaterials are uncovering arrangements of building blocks with defects and/or aperiodicity that allow for the realization of new functions.⁶⁴ Expanding computational design and creating techniques that enable precise control of defects in 2D and 3D are necessary to realize computational design of materials and structures with unprecedented and targeted physical properties.

6. Enable targeted design of materials synthesis, structures, and functions

Inverse design aims to begin with a desired specific property, and then subsequently identify or develop a stable and synthesizable material possessing precisely that property. There have been several examples of attempts at inverse design in recent years with varying degrees of success. While accuracy of theory can be a limitation in certain cases, a major bottleneck nowadays is the lack of a reliable synthetic pathway to

an identified material or structure: although state-of-the-art theory can predict with good accuracy the (meta)stability of a given bulk crystalline compound, there are no general theoretical frameworks for predicting whether a given compound can be made. Although the space of crystalline compounds is vast, the possibilities become exponentially greater if one considers structural features such as defects, and if one includes interfaces, patterns, and other efficacious combinations of materials. While *ab initio* methods and intuition can help predict the stability of many crystalline materials and molecules, there remain significant challenges to theory in predicting the stability of structures beyond bulk crystals, such as complex interfaces and heterogeneous systems with defects. Additionally, although there have been significant advances in time-dependent *ab initio* methods and kinetic modeling, practical prediction of materials dynamics, including synthetic pathways and stabilities, in relevant environments continues to be a challenge. Standard synthetic efforts have been successful in uncovering promising new compounds based on intuition, and rules derived empirically using advanced theory that explain the resulting properties in detail; but advances in precision synthesis and ML offer an opportunity to further develop and more tightly integrate theory, data science, and experiment for materials design and synthesis. A tightly integrated iterative loop, starting with theory and proceeding with synthesis and characterization, can accelerate the discovery of new materials and enable the targeted design of precision materials with desired functionalities, while deepening our understanding of the theory of materials.

Opportunities

Theme 1. Advance autonomous synthesis and processing for materials manufacturing

1. Develop maps of reaction pathways and routes to controlling them and providing feedback to refine theory

The rapid and automated synthesis of materials and materials structures with atomic precision—combined with data acquisition techniques, with data storage and dissemination platforms, and with interpretable AI/ML models—can lead to rich data sets. This information can refine state-of-the-art *ab initio* theory and kinetic models, leading to predictive time-dependent behavior beyond ground states and steady states. Past work using ML algorithms trained on reaction data has been used to successfully predict reaction outcomes. High-throughput workflows for lanthanide-doped nanoparticles have allowed the validation of a rate equation modeling approach,⁶⁵ enabling the screening of any combination of dopants *in silico* to predict their spectra. Beyond simple rate equation-based models, ML has the potential to transform the nature, efficiency, and accuracy of interatomic potentials—leading to simulations with chemical and structural specificity—and do so orders of magnitude faster. ML can also potentially inform the approximate functionals used in *ab initio* methods themselves, improving the treatment of strong correlation effects to enable better prediction of reaction barriers and kinetics.

2. Enable faster discovery of efficient pathways to products (including precursor development) that are structurally phase-pure and have optimal functionality

The development of new materials, in particular polymeric materials, is a long process with many low-probability events. For example, taking a new feedstock to make a new monomer, then developing a new polymerization process to make a new-to-the world polymer with unknown properties and applications, is almost certainly destined for failure. To discover more efficient processes to create novel materials with optimal properties, one emerging strategy is to discover new ways to use existing monomers, from readily available sources, to make polymers that are different from yet similar, to existing polymeric materials. With this strategy, the only barrier to the development of a new material is the identification of a new polymerization pathway. One example of how this tactic has been recently employed is the isospecific polymerization of propylene oxide (iPPO). The atactic version of this polymer (aPPO) has been used commercially for decades as a midsegment in polyurethanes. Similar to aPPO, iPPO is a photodegradable polymer; however, in contrast to aPPO, iPPO is a polymer that strain-hardens to form a material with

ultimate tensile strength comparable to that of Nylon-6,6.⁶⁶ This material has promise as a replacement for non-degradable polyolefin and polyamide marine nets and lines, which contribute to more than half of ocean plastics.

3. Create materials with functions optimized for energy applications

Transformative manufacturing will impact the entire spectrum of energy applications, including energy harvesting (solar, thermoelectric, piezoelectric), energy conversion (fuel cells, LEDs, motors), energy storage (batteries, capacitors) and energy transmission (superconductors, wide bandgap semiconductors). In virtually all of these applications, efficiency and performance are dictated by the ability to create a pristine interface between dissimilar materials. Interfacial control can be extremely challenging because issues such as lattice mismatch, high interfacial surface energy, chemical incompatibility, interdiffusion, and contamination can lead to poor interfaces and degraded performance. Consequently, the design and synthesis of materials for energy applications must include both the particular function, such as light absorption or energy storage, and facile integration within the device to produce high-quality interfaces with few defects. Quite often, the performance of energy devices can be improved by including a third component, such as a buffer layer or additive, that stabilizes the interface between two functional materials. The design of high-quality interfaces and the discovery of effective additives and buffer layers can be accelerated by high-throughput experimentation, AI, and multiscale modeling.

Theme 2. Achieve materials with designed functions and structures via atomically and molecularly precise synthesis and processing to transform manufacturing

1. Realize application-specific hierarchical, multifunctional materials and structures

The complex functions of energy production, storage, and use are enabled by hierarchical materials design, often involving heterogeneous materials. Although remarkable structures have been created in research environments, major gaps in the ability to manufacture such materials exist at two levels. First, there is an inadequate understanding of the underlying controls on their formation, whether in the synthesis of the fundamental building blocks or the processing of those blocks to predictably produce the targeted hierarchy with a desired uniformity and distribution of defects. Second, the scalable synthesis and processing methods needed for manufacturing is lacking. In hierarchical materials, function is an emergent outcome of organization at multiple length scales, so the organization at larger length scale is dependent on that at shorter length scales. Consequently, the challenges associated with predicting pathways, equilibrium end states, and kinetic traps discussed above are greatly amplified. Changes in atomic and molecular structure or processing conditions at one scale cascade through larger length scales; they grow, at best, combinatorially and, at worst, exponentially as a result of nonlinearities in cross-scale coupling. Even for systems in which hierarchical organization has been mastered at the lab scale, the sensitivity to local conditions and the difficulty of controlling materials critical events, such as nucleation or phase separation, render scale-up equally challenging. Still, there are notable exceptions when such problems are largely conquered by instituting global controls, such as substrates with physical or chemical patterning that forces targeted outcomes.⁶⁷ Such examples offer hope that a general strategy to achieve scalability is possible.

2. Develop a fundamental understanding of materials with emergent properties over multiple scales

The capability to predict structure-property relationships in multiscale materials is challenged by the disparate phenomena that govern behaviors at vastly different scales. However, recent advances in multiscale modeling and ML provide promising opportunities to predict structure-property relationships in multiscale materials. Another complexity of multicomponent systems is the abundance of interfaces, which can advantageously or disadvantageously dominate a system's properties. Developing tools to

understand and predict the structure and properties of interfaces is therefore another critical component of advancing the manufacturing of complex materials with desired behaviors. Here, new opportunities are provided by recently developed microscopy and scattering capabilities sensitive to the interfacial atomic structure,⁶⁸ and computational methods that can powerfully predict interfacial structure and properties.⁶⁹ Overall, the use of physics- and ML-based approaches for predicting complex structure-function and interface-function relationships is poised to accelerate the design, synthesis, and scale-up and processing of materials. Potential impacts include improved target functionality; reduced uncertainty; increased efficiency; reduced energy consumption; and accelerated manufacturing of scalable, durable, lightweight, and recyclable materials for next-generation energy technologies.

3. Position single or multiple nanomaterials for quantum electronic and photonic technologies

The application of low-dimensional materials in unconventional energy and quantum devices requires control over their positioning at the nanometer scale, and yet, often with arbitrary complexity over the macroscale. For example, many quantum computing schemes require the positioning of charge and spin qubits with single-digit nanometer precision. This control can be achieved by matching bottom-up assembly with top-down fabrication methods to integrate the best of these different techniques. For example, lithographic definition of topographical features has been used to direct the assembly of block copolymers and inorganic nanomaterials. Figure 36 shows examples of the deterministic assembly of colloidal nanocrystals with tailored size, shape, and composition using size- and shape-engineered, functional and/or sacrificial templates. In these examples, the arrangement of nanomaterials allows the sculpting of the interaction of light with matter. These strategies have been used to position single upconverting nanocrystals (which absorb infrared light and emit in the visible) in metal nanohole arrays, and to create upconverting nanocrystal–metal nanorod heterodimers, both of which use the plasmonic character of the nanostructured metals to amplify the upconverted luminescence. Topographical templates also have been used to assemble artificial metamolecules, in which nanocrystals serve as the building blocks of oligomeric structures and can form closely packed and open architectures. For metal nanocrystals, these assemblies have arrangements that “sculpt” rotation- and polarization-dependent, hybridized electric and magnetic plasmonic modes. The development of computational inverse design techniques motivates the establishment of assembly methods to realize more complex and arbitrary assembly arrangements.

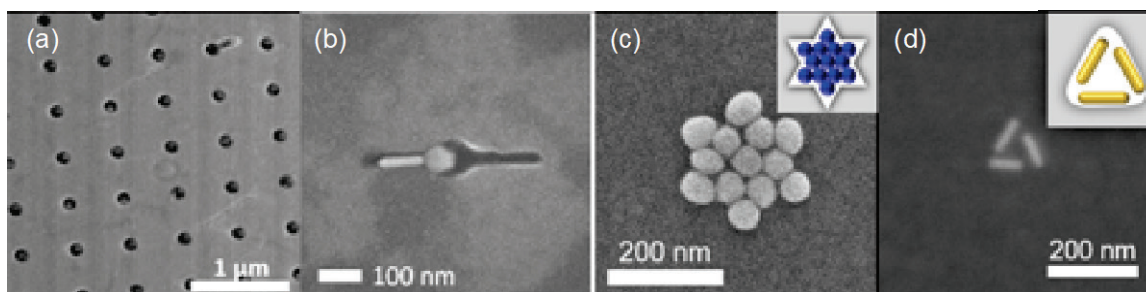


Figure 36. (a) Assembly of upconverting luminescent nanophosphors (UCNPs) in gold nanohole arrays⁷⁰ and (b) gold nanorod–UCNP heterodimers, both of which amplify luminescence efficiency.⁷¹ (c) Close-packed gold nanocrystal oligomers⁷² and (d) open gold nanorod trimers, with rotation- and polarization-dependent, hybridized electric and magnetic plasmon modes.⁷³ Sources: (a) Reprinted with permission from [M. Saboktakin et al., ACS Nano 7, 7186–7192, 2013](#). Copyright 2013 American Chemical Society. (b) Reprinted with permission from [N.J. Greybush et al., ACS Nano 8, 9482–9491, 2014](#). Copyright 2014 American Chemical Society. (c) Reprinted with permission from [N.J. Greybush et al., ACS Nano 11, 2917–2927, 2017](#). Copyright 2017 American Chemical Society

4. Improve efficiency through manufacturing and the material lifecycle

A paradigm change is needed by which the product and the process-to-product are designed for optimal efficiencies to enable critical reductions in energy consumption and emissions. For example, the amount of energy required per unit of output (J/kg, e.g.) needs to be minimized. Each step in a multi-step process can be a target for improved efficiency, or a series of processes can be consolidated into a more efficient process. In a case where the process itself cannot be rendered more efficient and net energy is wasted, that wasted energy, often heat, could be recovered. Similarly, to reduce emissions, the amount of energy used for the process or the nature of the fuel itself can be targeted, and a feedstock can be chosen for “atom efficiency,” so that byproducts and side-reactions are minimized. Beyond reductions in energy and carbon intensity, manufacturing with intention would imply extension of the concept of smart design to “use intensity.” This is a very broad concept that includes enhanced materials properties for greater longevity or other performance metrics, reducing the need for early replacement or even minimizing the quantity originally required for a given function. Ultimately, this concept moves manufacturing toward a circular economy (reuse, repair, recycle).

Conclusion

The promise of precision synthesis to transform manufacturing lies in the ability to take full advantage of the intimate connection between the function of a material and its architecture from the atomic to the mesoscale. Realizing that promise depends on the development of autonomous synthesis and processing capabilities. This approach should take advantage of recent advances in high-throughput synthesis, identification of structure-function signatures, and development of physics-based computational models. Tackling the fundamental challenges of pathway engineering is necessary to access kinetically trapped metastable states and construct experimental and computational databases needed to constrain kinetic and thermodynamic parameters. Doing so will offer the opportunity to rapidly predict efficient reaction pathways to target architectures, as well as routes to control them, with optimized functions for energy applications. To be able to use these autonomous capabilities for atomically precise manufacturing of materials with designed structure and function, it is highly desirable to take advantage of newly developed scalable synthetic routes, advances in understanding chemical drivers and physical forces underlying synthetic routes, the tremendous progress in atomic-scale spatial and compositional control of nanomaterials, and recent knowledge of structure-function relationships across scales. The new scientific challenges lie in designing materials for multiple functions, designing and controlling interfaces, exploiting surface chemistry as a means to direct synthesis and tailor function, managing defects, achieving massively parallel atomic-scale positioning of materials, and gaining a predictive understanding of the synthesis-structure-function relationship. Success in these endeavors affords the opportunity to realize application-specific hierarchical, multifunctional materials, as well as a fundamental understanding of their emergent properties over multiple scales; achieve manufacturing-scale production of quantum information, microelectronic and photonic devices; and reduce energy and emissions through manufacturing and the materials lifecycle.

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Panel 2: Processing and Scale-Up Science

Introduction

Bringing a new technology to the world requires the ability to process both the starting constituents and manufacture final products efficiently at scale. Reducing the cost and environmental footprint of manufacturing fuels, chemicals, and materials remains a significant scientific and engineering challenge. Indeed, enabling a material to be produced in previously unattainable quantities or at much lower cost can be as challenging as the discovery of the material itself. Moreover, the often-complicated processing steps considered acceptable in prototyping must be simplified to enable volume manufacturing of final products, by either scaling up or numbering up. However, deep understanding of the science underpinning scale-up across length scales and the associated processes are often limited, making scaling up costly and slow.

The intent of this panel was to identify where new basic science can best advance processing and scale-up of materials important for the nation and world. This effort includes assessing the state of the art in processing and scale-up science, describing how processing may evolve in a future state with new raw material and energy inputs, delineating science gaps, and identifying where basic science will make the greatest impact on processing and scale-up.

Recent Advances

Benign and efficient energy processing

As industries move to minimize waste and the impact of manufacturing on energy consumption and the environment, new scientific understandings are needed to simplify processing, reduce carbon inputs, increase product life, and reduce the use of expensive components in processes. Among all US industrial processes, petroleum refining (3.9 EJ)¹ and chemical manufacturing (4.8 EJ)¹ consume about 40% of the energy used as fuel in the manufacturing sector (21.0 EJ) (all 2014 data). A large fraction of energy and waste is in separations and cleanup. Potential new ways of driving manufacturing processes include using alternative energy sources, reducing energy use by improving understanding of the effects of processing conditions on structure, and reducing waste through improved catalysis. The latter is the subject of a previous Basic Research Needs report.²

Emerging ways to drive processes using energy sources beyond conventional thermal routes include light plus electric fields,³ mechanochemistry,^{4, 5} sonochemistry,⁷ microwave heating, and low-temperature plasmas,⁷ in addition to electrocatalysis.⁸⁻¹⁰ Most conventional metallurgical processes, such as melting and sintering, use excess material and energy because of their inherent inefficiencies. Low-thermal-budget processing is an emerging opportunity for manufacturing of metals, refractory metals, and ceramics that is less energy-intensive and enables integration with thermally sensitive materials. For bulk materials, emerging low-thermal-budget options include cold sintering,¹¹ which uses a liquid-phase flux to assist in densification without high heating, and flash sintering,¹² which applies an electric field to enable sintering and reactions¹³ to take place through rapid joule heating,¹⁴ and electrically driven mass/charge transport. Scientific underpinnings that dramatically reduce the number and complexity of processing steps, energy use, and waste scrap are critical. Nascent strategies based on synthetic biology also offer potentially efficient pathways to create highly functional hard and soft materials.^{15, 16}

To reduce process waste, improved catalysts are needed for chemical processes, and their preparation methods are critical in defining performance. Conditions that control the molecular interactions can dominate the morphology and topology of the active/functional sites.¹⁷⁻²² The inability to monitor and control conditions at larger scales remains a challenge, as differing induced charges on particles and surfaces alter deposition at scale, including the presence of impurities and the chemical nature of active

phase precursors. Even exo- or endothermicity, which are easy to control at small scales but more difficult in a manufacturing setting, can diminish the ability to translate catalysts from laboratory to manufacturing scales. Recent examples of challenges to scale-up can be found in the literature.^{17–22}

Alternative carbon sourcing

Fossil carbon, having an energy content of 2.8 EJ (2010 data), is used as the feedstock of many basic chemicals and plastics.¹ Renewable and carbon sources that are from waste streams (industrial waste gases, municipal solid waste, manures and agricultural wastes, and wastewater sludges) present new opportunities. A move toward waste carbon as a feedstock necessitates distributed processing, which requires safe conditions, integration of process steps (combining reactions and separations), and the ability to use renewable energy. One opportunity is the merger of chemical and biological catalysis to form new processes for waste reuse. One example of moving a fundamentally new process from laboratory to manufacturing in a relatively short time frame has been the biological processing of waste gases (Figure 37), which can be combined with chemical processing to form additional compounds. In this manner, biological systems efficiently couple one-carbon compounds into two-carbon compounds—a difficult feat with synthetic chemistry—and chemical processes are used to convert a two-carbon alcohol to fuels and chemicals, which is difficult for biology. At a large enough scale, alternative carbon sources could even displace nonrenewable hydrocarbons used as fuels for transportation.

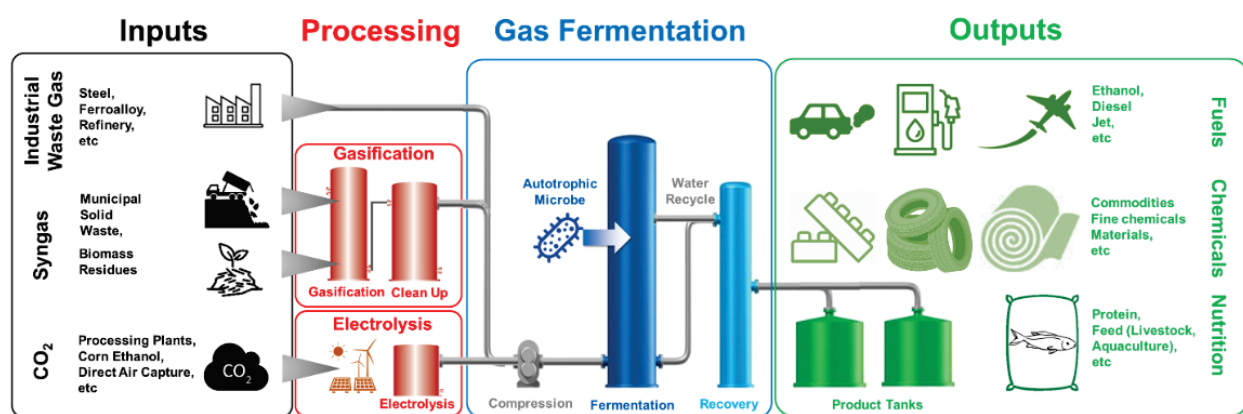


Figure 37. Replacing fossil carbon with renewable or waste carbon as the feedstock for bulk chemicals could offset more than 3 EJ of fossil resource consumption. A continuous biological process (fermentation) has recently been commercialized that processes CO-rich industrial waste gas from sources including unrecycled plastic, wet carbon sludges, and industrial waste gases into ethanol. The alcohol can be chemically upgraded to high-value products in an integrated catalytic process. The combination of biological and chemical processing opens new opportunities to produce chemicals and fuels with higher conversion and energy efficiency. *Source:* Image used by permission of LanzaTech.

Far-from-equilibrium processing

Far-from-equilibrium processing bridges the gap between renewable/lower-energy processing and self-assembly/multiscale processing. Some far-from-equilibrium energy-intensive processes have transitioned to lower-energy manufacturing through recent advances in novel techniques, e.g., spark plasma sintering, additive manufacturing (AM) of refractory materials. These new processing advances direct energy and materials only where they are needed on specific time scales and gradients, e.g., controlling grain structure.²³ They have the potential to further precisely control local conditions to form compounds with precise chemistry, novel structures, increased energy efficiency, minimized defects, and controlled phase composition and metastability across length scales.

Atomic-level understanding of the properties of matter far from equilibrium and under intense process conditions is a challenge across all classes of materials. Manufacturing advanced metals and ceramics is energy intensive, often involving high temperatures, long processing times, rapid quenching, radiation, and/or high pressures. Their structure–property relationships are often poorly understood,^{24,25} in part because the very structures that provide desirable properties are significantly far from equilibrium. As new processing methods emerge, such as rapid solidification AM, the energy landscape changes drastically; and understanding must be gained to enable navigating those regions of metastability. Future materials, such as those that might provide exceptionally high strengths or ion mobilities, will require more radical departures from equilibrium states and the combination of novel compositions and innovative processing to arrive at the desired end state. High-performance computing and advanced characterization will be needed for composition and processing co-development. Extreme processing methods are unlikely to up-scale easily, as precise local control of process conditions and metastability across length scales is challenging; and once promising composition and energy spaces are identified, there will be significant needs for transforming manufacturing science.

Additive manufacturing, bottom up, self-assembly, and multiscale processing

Processing research involving AM and other types of powder or droplet spray-based advanced forming methods minimizes materials waste by producing products in their final shape with minimal (or no) machining required. However, there is a need to both better understand and control microstructural evolution, including grain size and orientation, to improve their strength and ductility, especially in AM.

For high-quality oxide and compound semiconductor thin films, it is challenging to rapidly accelerate the deposition kinetics of molecular beam epitaxy, chemical vapor deposition, and sputtering, while maintaining the desired control over features such as epitaxy, orientation relationships, crystalline quality, and interface chemistry. For example, pulsed laser deposition is one of the best methods for producing high-crystalline-quality oxide films; however, it is one of the most difficult to scale up.^{26–30} Other physical methods—such as sputtering, or chemical methods like plasma-enhanced chemical vapor deposition³¹ or aerosol-assisted chemical vapor deposition³²—are modestly scalable. However, they become more challenging when multiple cations or anions are required in the final film, necessitating the use of multiple targets or precursors, or when the film must be relatively thick. Electrochemical methods are of growing interest, for example in manufacturing the ceramic cathodes used in lithium-ion (Li-ion) battery electrodes (see the sidebar). They have been demonstrated to grow both ceramic oxides and semiconductors in relatively ordered forms.³³

Sidebar: Scale-up of Electrodeposited High-Performance Battery Electrodes

Current status

A significant driver of Li-ion battery costs is the many energy-intensive steps involved in electrode production, including raw material extraction, electrode material synthesis and purification, and electrode fabrication. In addition, despite considerable efforts, the performance of Li-ion batteries is still far from ideal, in part because the internal structure of the slurry-cast cathode and anode consists of active material dispersed in particulate form mixed with electrochemically inactive additives.³⁴

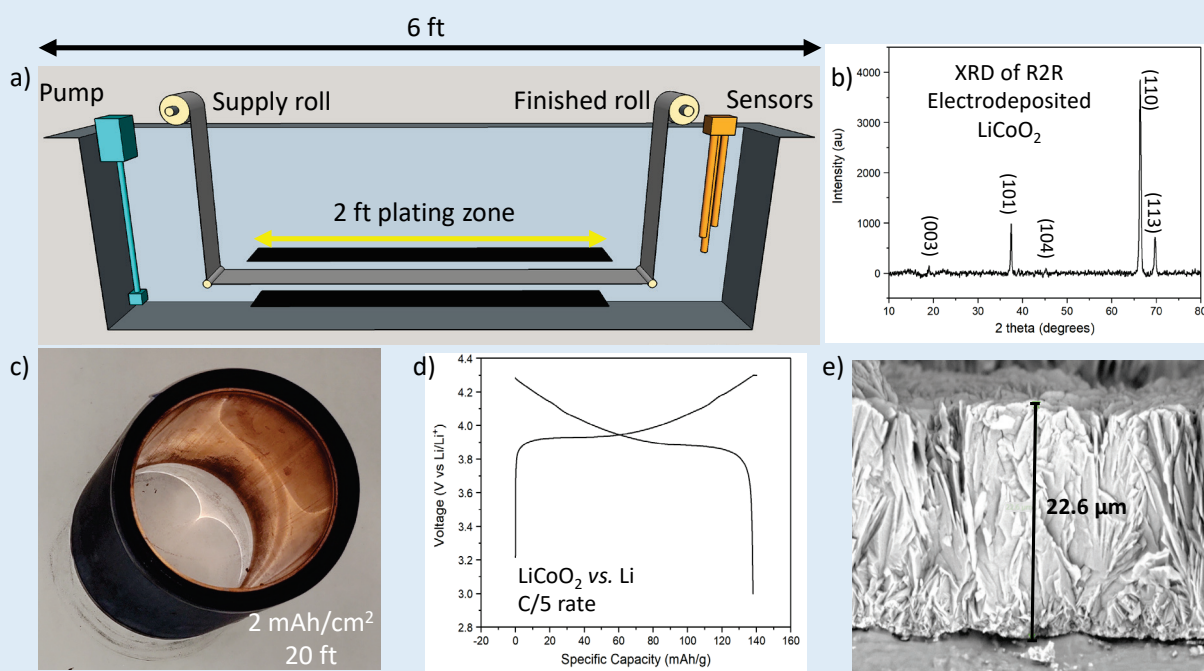
Opportunity

Electrodeposition is a well-known path to growing ceramic layers with tightly controlled crystallographic orientation and porosity.³⁵ The electrodeposition of high-performance LiCoO₂ with particular relevance for energy consumption during production was recently reported.³⁶ The material was grown from low-grade precursors at temperatures ~500 °C below those conventionally used to fabricate LiCoO₂, yet the process resulted in Li-ion

cathodes with better-defined crystallographic orientation and higher active material densities than are found in conventionally formed electrodes.

Scale-up

Recently, pilot-scale roll-to-roll electrodeposition of a high-performance LiCoO_2 -based cathode was demonstrated by Xerion Advanced Battery Corp. The densely packed flake-like morphology of LiCoO_2 produced via this roll-to-roll process is presented in the image at bottom right in the figure. The chart at the bottom center of the figure illustrates the charge and discharge profile of the LiCoO_2 electrode, showing the expected capacity of about 140 mAh/g despite the use of low-purity electrodeposition precursors. The controlled crystallographic orientation places the fast-diffusing lithium plane perpendicular to the substrate, resulting in electrodes with high power densities, even when the active material is thick. These results indicate the promise of electrodeposition as a new paradigm for manufacturing high-performance electrodes for energy storage.



(a) Schematic of the roll-to-roll process. (b) X-ray diffraction peaks. The near absence of the (003) peak indicates the strong texturing of the film. (c) Image of 20 ft roll of LiCoO_2 coated on metal foil. (d) Charge-discharge of electrodeposited LiCoO_2 . (e) Scanning electron microscope image of cross-section of the LiCoO_2 . Source: Images used by permission from John Cook, Xerion Advanced Battery Corp

Scientific Challenges and Opportunities

The scientific opportunities present in processing and scale-up are considerable. As manufacturing strives to become more environmentally benign and energy efficient, they must move beyond use of nonrenewable carbon sources, create materials that derive function through far-from-equilibrium internal structure, and learn to build materials from the bottom up and via self-assembly. It is increasingly apparent that major scientific advances will be required in characterization, modeling, and new processing methods that enable exquisite control of surfaces, interfaces, and defects. This section describes the scientific challenges and opportunities identified in these areas.

Metrology and Characterization at Relevant Dimensions for Scale-up

Modern manufacturing requires tight control of conditions and real-time measurement of the properties of a product as it moves through the manufacturing process. Collecting and analyzing the data at rapid time scales requires the development of new characterization, data analysis, and modeling methods. New characterization methods that are capable of collecting large, relevant data sets will increase yield and accelerate scale-up, especially when coupled with high-fidelity simulations that facilitate optimization of manufacturing processes.

Polymers and polymer composites are examples of where characterization at relevant dimensions would have significant impacts on scale-up. Although there has been progress in understanding the relationship between processing and properties for simple polymer systems, researchers lack the ability to characterize, in real time, the microstructures formed under high shear and rapid heat-transfer processes during manufacturing of high-performance polymer blends and composites. This lack of information significantly limits the ability of industry to predict the relationship between properties and processing in polymers and composites. Similar issues are present in manufacturing of almost all classes of materials.

Open questions remain as to how to characterize the three dimensional properties of a material in real time in a manufacturing process. Can microstructure, density, thermal conductivity, electrical properties, and elastic modulus be determined with high spatial and depth resolution using non-contact, spectroscopic, or acoustic methods?³⁷ New understandings of materials physics, particularly at the intersection of light–phonon–matter interactions, may enable such characterization. What are the opportunities for new x-ray–based characterization approaches? At the even more local scale, can the concentration of atomic defects (good and bad) be determined in real time in three dimensions? Can rich information provided by advanced metrology methods be processed and interpreted in real time to guide manufacturing processes? Success will have a deep and meaningful impact on advanced manufacturing.

Multiscale Modeling, Measurements, and Systems Design

Manufacturing of fuels, chemicals, and materials requires that synthetic methods that span from the laboratory to pilot, and ultimately, industrial scales. Translating laboratory discoveries into industrial manufacturing remains a challenge.

Multiscale approaches. Commonly used experimental and modeling techniques are scale-specific (in both length and time). For materials and products, separate approaches are used to design and predict properties and to understand behavior and measure performance. Integrated multiscale physical and chemical approaches are needed to address these phenomena, particularly for complex materials. The availability of data mining, machine learning algorithms, and new computational tools capable of working with up to 300,000,000 atoms will enable new insights and knowledge not available before. Integrated multiscale modeling (such as thermal evolution and reaction gas distribution/pathways) of coatings and advanced atomic-level characterization capabilities would further our understanding of surface science. Modeling across large time regimes (say fs to s) requires combining dynamics and kinetics to account simultaneously for multicomponent transport, elementary reaction steps (e.g., microkinetics), and physical events (e.g., crystallization, aging, sintering). New approaches are needed for evaluating phenomenological kinetics at larger scales, modeling transient data to temporal scaling, and connecting equilibrium and far-from-equilibrium states (e.g., numerical solutions of the related, stochastic partial differential equations).

Far from equilibrium. Most multiscale approaches are largely limited to equilibrium processes, and the need for modeling of nonequilibrium systems at all scales prevails. Modeling the changes of properties caused by environment and process conditions may provide insights that can be used for phenomenological models for far-from-equilibrium situations. Output from models will offer input for

molecular models for more precise atomistic calculations. Currently, the automotive industry uses a similar technique in which empirical constitutive laws provide limited empirical access to anisotropic and heterogeneous materials. The challenges for understanding microstructure and texture and their evolution during processing and manufacturing remain.³⁸

Sensors across scales. Future manufacturing requires the identification, adaptation, or development of sensors that can be used across scales for monitoring events, processes, and components. Tools need to be developed to extract and make sense of large data sets that can be used in industrial settings. Since the data sets will come from diverse sources and from an extensive variety of sensing instruments, extraction of information from multivariate measurements typically will need to be robust against noise and errors in the measurements. Advances in analytic techniques will need to account for the complex structure of multivariate measurement errors. A better understanding of the origins of errors for a sensor or measurement system is needed. Additionally, improved data analysis tools need to be designed to treat the errors in an optimal way.

The optimal application of realistic error characterization and error-in-variable modeling reduces the cost-prohibitive replication process that is required to create surrogate estimates for “true” noise characteristics. Integration of such methods with multiscale models can lead to more efficient extraction of information and improved decision-making.

Surface Science and Interfaces

A deeper understanding of surface science and interfaces would greatly enhance processing and scale-up and would transform manufacturing. Currently, the ability to realize the full benefits of existing computing power and advanced models is limited due to a lack of input data and constitutive relations of the physics of surfaces. Such advances would not only enable new materials by control of surfaces and interfaces (hybrid materials and composites), but improve traditional technologies (e.g., filtration, biofuels, chemicals, catalysts, powder metallurgy). The scientific challenges that arise are addressed separately below for surfaces, interphases, and diffusion/transport.

Free surfaces. Powders, slurries, porous materials, fibers, and membranes possess large surface-area-to-volume ratios compared with bulk materials. Gaining atomic-level control of surface species and layers could allow for the generation of nonequilibrium surfaces, access to new rapid solidification regimes, and precision processing control for efficient synthesis and rapid scale-up of materials with large surface-to-volume ratios, resulting in new processes and materials with unique microstructures and properties. Researchers lack an understanding of rapid solidification, solute segregation scales, limited diffusion lengths/paths, surface chemistries and rates of formation, nanoscale structures/limitations, and their impacts on the final consolidation aspects and rate-controlling processes. Deeper understanding of the interactions of nanoscale dispersions, including the factors that control solubility and stability, would enable deterministic control of particle-fluid interactions; and their rheology, printing, drying, and sintering could be tailored on demand.

Interphases. For particles suspended in liquid phases and heterogeneous soft materials, interfacial properties and processing methods (e.g., application of shear, thermal, or other forces) influence their local solidification kinetics, microstructural development, and resulting interphase properties. The determination of the dominant atomic and molecular interactions that govern interphase morphology and topology is essential. Interphases can also be controlled by the chemical composition of active precursors (e.g., reactivity, hydrolysis, surface charges, charge density, structure) and liquid phase, including presence of impurities.

Diffusion/transport. Surface diffusion, liquid, and solid-state diffusion and permeability data are important for materials processing; however, these data sets are limited. Diffusion affects operating conditions, structure and performance, control of transport, and durability. For example, the lack of precise control of the surface chemistry, composition, thickness, and spatial distribution of oxide powders and metal electrodes hinders the performance of rechargeable batteries. Improved understanding would benefit the development and manufacturing of advanced membranes and electrodes, accelerating the translation of electrochemical materials from academic labs to real-world applications.

Defect Control and Tolerance

Once a defect is introduced during manufacturing, it remains throughout the product's life. While self-healing materials are now commercialized,³⁹ “intelligent” materials that can self-repair manufacturing defects and damage remain out of reach. Defect-tolerant manufacturing coupled with the embedding of “intelligence” into a material, which enables automatic detection and repair of defects and damage, would expedite processing and scale-up, decrease scrap rate, reduce safety margins, and reduce the overall energy inputs into manufacturing. The impacts on materials science and processing would be better life prediction, reliability, and part maintainability to reduce life cycle cost and energy.

Defect control. Many materials made at lab scales cannot be made at industrial scales due to defects inherent to scaled-up manufacturing processes. While defects are inherent to any material, and some defects highly desirable, what remains needed are better processes to control defects at manufacturing scale. Success would enable new energy-efficient and high-performance materials that exhibit unprecedented properties and performance (e.g., high specific strength, fatigue life, electrochemical energy storage capacity). For example, scalable pathways for achieving the theoretical performance of current bulk materials could be enabled. Defect control requires understanding processing pathways to enable hierarchical materials and metastable bulk materials to achieve spatially resolved microstructural heterogeneity on demand.

Intelligent materials. Unifying principles for new classes of intelligent materials are needed that address fundamental questions: What is the uncertainty propagation during processing as opposed to material architectures? How are defects measured and self-reported for repair? What is the specific micro-defect correlation to material performance (e.g., grain size control)? And is sufficient understanding currently available of microstructural evolution and degradation under loads, or is a better understanding of stress-induced cracking and materials fracture needed, potentially including the characterization of microscopic stress fields, tribo-emission spectroscopy, and quantum molecular dynamics modeling of fracture?

This level of defect control would enable near-net shape manufacturing of parts with acceptable defect size and distribution, potentially impacting simplified processing for conventional ceramics, novel powder techniques, and possibly machining/finishing of very hard, brittle materials. New chemically and physically driven processes could be developed that have fewer processing steps, alongside novel machining and finishing technologies, which also would enhance modeling of the impacts of defects on properties and lifetime. Novel inspection techniques that allow in-line detection of defects would impact new materials/processes/systems for energy efficiency, e.g., ceramic matrix composites, fuel cells, new physics/chemistry-based processes, and new energy cycles.

Efficient Processing via Combinations of Input Energies

From hot-rolling of metals to high-pressure, high-temperature reactions, industry is familiar with combining heat and pressure to accelerate and direct chemistry. The results reflect a combination of thermodynamics (phase behavior, equilibrium) and kinetics (activation barriers, volumes of activation). Less traditional combinations also can provide combinations of energy inputs whose sum exceeds that needed to activate desired reactions and enhances the selectivity of the process and the longevity of

catalysts. A few examples are light plus electric fields,³ mechanochemistry,^{4,5} sonochemistry,⁶ and electrocatalysis.⁴⁰

Targeted energy. Alternative sources of energy—such as sonochemistry, low-temperature plasmas, and mechanochemistry—are capable of focusing energy in small volumes. While the direct excitation of a specific bond within a molecule is generally not possible owing to rapid thermalization of the absorbed energy prior to reactions,⁴¹ steep spatial gradients within a reaction zone can be constructed to deliver excited reactants or remove excited products rapidly enough to enhance reaction selectivity. Systems with very short, down to millisecond, contact times may be particularly useful.^{42,43} In addition to a lack of mechanistic and kinetic information, materials development for catalysts used in nonthermal plasma systems is needed.

Methods to study reaction transients⁴⁴ are needed but must be analyzed carefully to bear on steady state performance. Approaches that study transients in the course of steady state operation include isotopic transients,⁴⁵ and modulated excitation spectroscopy^{46,47} can afford information across the evolution, use, and deactivation of catalysts.

Electrochemical/electrocatalytic. Electrochemical processing is not new. More than 100 years ago, the Hall-Heroult process reduced the price of aluminum from that of silver to that of a commodity. In the future, the role of electrochemistry may expand to provide additional levers to control reactions. While electroplating of metals in film form is widely practiced, more recently, it was discovered that electrochemical processing can control, with great precision, the doping and crystallographic orientation of broad classes of organic and inorganic materials in both thin and thick films.^{10,36,48,49} Crystallographic control is of particular importance given that many of the most important materials for energy storage and production are highly anisotropic and exhibit advanced properties only when highly textured with appropriately small defect densities. Electrocatalysis also can extend levers for effecting chemical transformations (Figure 38).^{9,40}

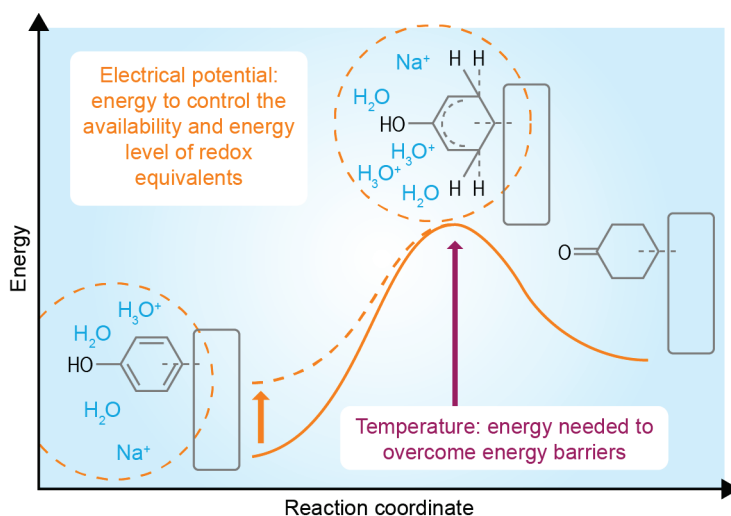


Figure 38. Applied electrical potential is an energy input that regulates energy levels and the availability of redox equivalents. Thus it, along with temperature, provides a strong control lever for chemical reactions. Source: Image provided by Pacific Northwest National Laboratory

Targeting the diversity of bonds required to form desired materials via an electrochemical/electrocatalytic pathway remains an open challenge and will require development of the electrocatalyst, design/selection of the precursor, and electrolyte engineering. Characterizing and controlling the electrochemical processes in a manufacturing environment is complicated by the presence of the liquid and sometimes

corrosive electrolytes and the diversity of chemical species present in the system. Finally, of particular relevance for manufacturing, the discovery of scientific principles to broaden electrochemical processing windows is needed (e.g., the use of surfactants).

Other forms of light. Artificial light-driven processes are another means to use renewable electrons to drive reactions. The key challenges will be maximizing the efficiency of converting electrical energy to light; improving photon transfer; and learning to control reactivity through reactor configuration, light flux, and mass transfer. Nearly monochromatic light-emitting diodes can be used to study the effects of specific light wavelengths on reactivity. Still, an understanding of the effects of temperature, dark reactions, and characterization of mass transport is needed to optimize light-driven process.

Overall, improving the efficiency of transformations via combinations of input energy can reduce the carbon emissions of many processes. In some cases, there is an added benefit of providing materials with improved or even otherwise unavailable characteristics. Finally, using combinations of input energy opens new windows for processing in decentralized facilities using renewable electricity.

Summary

The challenges of moving innovative discoveries from the laboratory bench to fully integrated industrial-scale processes are significant and present an area of science that is often neglected. A deep understanding of the basic science of processing and scale-up of materials synthesis, shaping, and reuse is important for the nation and world. Deeper understandings of how to effectively and efficiently use, at scale, new raw material and energy inputs, and of how to tightly control the chemistries, nano- and microstructures, and compositions of the materials produced will make great impacts on processing and scale-up. The science needs identified in the panel discussion point to a need for operando characterization (PRD 2), multiscale models enabling adaptive control (PRD 3), and energy-efficient manufacturing (PRD 4).

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Panel 3: Systems Integration Science

Introduction

Systems integration science is a broadly defined topic area that requires a collective understanding of its definition. The panel spent considerable time discussing the nuances of the definition. “System” can refer to a complex set of interacting components in a product or device, or to a complex environment in which a product must function. Systems can be thought about at various scales, including the nanoscale, mesoscale, or macroscale. The boundaries defining a system often expand at higher technology readiness levels. Systems integration often involves multiple objectives and trade-offs between differing performance aspects. Successful application of fundamental research to systems integration would increase the probability of success for technology applications and reduce the time required to move products to market. Systems integration holds the possibility to transform manufacturing by enabling resilience and agility in molecules, materials and processes.

The current status of systems integration is that a number of key gaps cause technology development to stall or fail. Importantly, basic experimental and modeling research can address these gaps. Success in those areas will enable breakthroughs in manufacturing, while failure will manifest itself by placing unnecessary limitations on defining required chemical or material properties. Some examples of gaps include lack of reliability, lack of material and chemical compatibilities, and lack of data and models. A lack of reliability in realistic environments often is due to issues that were unanticipated in the initial experiment. Typically, the issue would be that the initial work focused purely on idealized reaction environments, settings and data sets. This approach significantly limits the ability to harness synergistic or emergent phenomena that arise only in interacting systems. Second, a lack of compatibilities among molecules, materials, or operations in interacting subsystems results in a lack of understanding of the structure-process-property relationships in truly multiscale ways (e.g., from nanometers to kilometers/ from atoms to devices). Third, the lack of data and models relevant to manufacturing conditions contributes to the production of data that are of value from an academic perspective but cannot provide answers economic and energy savings questions that ultimately are needed for development and implementation.

Basic science challenges, opportunities, and needs exist that can guide research and subsequent implementation for manufacturing. These do not necessarily appear in a linear fashion but can best be envisioned in a collaborative environment encompassing mutual awareness between low and high technology readiness level activities. In this environment, basic research would provide novel insights and imaginative alternatives informed by industrial processes. Based on discussions, panel 3 presented three aims: (1) identify tools and scientific challenges that can be addressed by fundamental research which are timely and will impact one or more of the gaps; (2) focus on approaches that are specifically motivated by systems integration needs, although there may be commonalities with other aspects of manufacturing-related science; and (3) find themes that resonate across multiple manufacturing sectors.

The following points illustrate how the integrative nature of these endeavors has important implications for basic research for systems integration understanding.

- (1) *Improved measurements and diagnostics under process-relevant conditions.* Examples include opportunities for manufacturing-relevant measurements; design, development; and implementation of high-speed and real-time on-line and in-line sampling, monitoring, and survey measurement techniques; and development of enhanced-fidelity low-cost “desktop” instrumentation and methods to enable faster data turnover.

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- (2) *Methods to assess and predict chemical products and materials performance under non-ideal conditions and during operation in complex environments.* Because local defects, for example, have impacts on multiple scales, there is a great need for an understanding of the mechanistic and kinetic basis for aging and degradation along the mesoscale (from atoms to machined parts). Furthermore, the ability to identify the robustness of synthesis conditions and operating environments will enable the determination and quantification of robustness both in a system's parts and as a whole.
 - (3) *Methods to integrate data from diverse sources.* Data collected both at the mesoscale and from different methods in the same process step enable the down-selection of experiments with a degree of confidence, while also extending the scope of the systems-based model predictions.
 - (4) *Improvements in systems integration science will directly lead to efficiencies in manufacturing and production processes.* Systems integration spans the length and time scales from basic research to manufactured processing and production. When data collection, evaluation, and feedback on all these scales are possible, the resulting manufacturing confidence level and qualification surety will enable broader sets of variables that can easily be reduced to confident choices of reactants, materials, and processes in energy-intensive manufacturing settings. These improvements also will catalyze conceptually new technologies in energy-intensive and energy-creating sectors.
 - (5) *Methods for integrating multiple devices or components that function in complex physical environments.* These methods will be necessary for deploying new technologies in the real world with minimized incompatibilities, minimized defects, and ensured robustness, thereby allowing for energy efficiency and economic benefits to a system process.

Basic Research Pathways

Descriptions of basic research pathways encompass “bottom-up” fundamental research tied to “top-down” systems processes. Examples include multicompositional and multifunctional materials and structures, which comprise many components made of different materials and chemistries to meet use requirements. Manufacturing these kinds of products typically require multi-step processes to attain a final high-yield pure product or the joining of many different parts for a final manufactured component. Chemical production processes, for example, require numerous steps, thus presenting the basic research challenge of controlling and tailoring reaction pathway steps to obtain a product with high yield and compositional purity along with maximized performance and minimal waste. In materials processing, much basic research is focused on the composition, structure, and function of a material, so that a component or system consists of a single material with compositional and functional gradients without the need for joining. In multicomponent systems, a system usually comprises many components to meet the use requirements.

A second challenge in connecting early-stage research to manufacturing is that functioning products at a system level typically make trade-offs among multiple performance measures rather than maximizing a single quantity. Advanced modeling and simulation results in an increase in the rate of data acquisition and improved quality of data. To enhance system performance and to avoid failures at the system level, materials and chemical processes should be modeled, tested, and characterized under conditions as close to the use conditions as possible.^{1,2} For example, a key to success in rapid design and manufacturing is that performance be derived and predicted from as-built (and not as-drawn) additive manufacturing (AM) parts based on measured microstructural properties. Similar conceptual challenges are common in more chemically oriented materials. There is significant interest in tying modeling to real-world manufacturing conditions to enable kinetic pathways to thermodynamic conclusions.^{3,4} Opportunities exist for new discoveries using high-throughput capabilities, machine learning, artificial intelligence, and large-scale BES user facilities.

Finally, it is important to acknowledge that new manufacturing technologies, such as AM, require research and development to minimize the uncertainties in the resulting products. This kind of research involves experimental measurements and large simulations, which result in huge quantities of data to be stored and analyzed. Challenges exist in using appropriate data handling methods to address this situation.⁵ Two thrusts were identified that address the scientific challenges, research directions, potential scientific impact, and energy technology impacts associated with these research pathways.

Thrust 1. Understanding interacting phenomena in integrated systems

A scientific challenge the panel identified is that complex interacting systems frequently feature emergent phenomena across many time and length scales. Multimodal characterization and multiscale modeling and prediction using many simultaneous length and time scales can counteract this issue. As a result, the community will develop the ability to deliberately incorporate complexity in prediction and understanding into their data sets.

Second, interactions occur between components, at interfaces and at the mesoscale, among additives and impurities, and from operating environments.^{6,7} Moreover, all of these interests can be strongly influenced by nonequilibrium processes. Basic chemistry and physics can develop a fundamental understanding of these processes, and manufacturing can exploit the emergent phenomena from these studies. The impact will be the ability to integrate modeling and prediction to enable advanced materials and chemical synthesis and device-level operation.

Additionally, it is important to understand impacts of chemical contaminants in a system. This capability is important both in materials processing and in chemical synthesis and scale-up. Modeling across the mesoscale enables a fundamental understanding of materials synthesis, design, and formation necessary for the successful design of reliable materials and chemical processes for transformative manufacturing. First principles modeling will be enabled by extension to broader scales (e.g., ab initio molecular dynamics) to model real-world chemical systems (e.g., contaminants, co-solvents,) and by learning how chemical kinetics influence the relevant processes. The use of density functional theory modeling and molecular dynamics simulation-based programs will enable chemical design and structural performance prediction prior to expensive and environmentally impactful industrial process scale-up. In fact, it affords a more straightforward path to chemical synthesis optimization than traditional experimentally based methodologies.

Additionally, information from mesoscale modeling will ultimately enable system-wide quantification of manufacturing uncertainty. Currently, much modeling is done by invoking a long list of simplifying assumptions that enable inexpensive methods of computation. Advanced computational methods development will be needed for this focus to enable more complex, real-world condition predictive modeling.^{8,9}

Third, the functions of a system are often far more than the sum of their parts. Research into the fundamental mechanisms of interactions among system components will result in a strong impact in efficiency in manufacturing. New capabilities from this knowledge base will be enabled to leverage diverse and incomplete data collections from multiple time and length scales.

Success across this thrust is key to a strong impact on energy technology. In particular, a healthy systems integration science effort will require the tightening of the current connection between fundamental research and later process development. The goal and result will be the accelerated development of new technologies in energy-intensive sectors via efficiencies not possible in a traditional linear relationship between basic research, applied research and development, and industrial processing.

Thrust 2. Harnessing complexity to create robust and resilient systems

A scientific challenge identified by the panel is that the limited control of assembly and complexity in integrated systems hampers the performance of existing molecules and materials and the conception of new processes. Research into the development of methods for predicting systems performance or failure will need to link components and phenomena over the entire lifetime of the system. Success in this area will exploit phenomena of interacting materials, chemistries, and components, thereby opening the envelope of composition, structure, and performance. Another challenge is the requirement of control of chemical and materials synthesis at multiple scales. Such control will elucidate a wide palette of options for creating processes and materials. The scientific impact will include self-correcting, resilient systems that create a diverse set of paths to new materials.

An example of these challenges comes from work on atomic-scale toughening and strengthening mechanisms based on engineered multiscale microstructures. Intermetallic alloys can possess exceptional soft magnetic properties, including high permeability, low coercivity, and high saturation induction but nonetheless exhibit poor mechanical properties that make them impractical for bulk processing and use at ideal compositions. Laser-based additive manufacturing, however, has been shown to be useful for traditionally brittle Fe-Co and Fe-Si alloys at near-ideal compositions for electromagnetic applications. Molecular dynamics simulations provided insight into the impact of a nanoscale subgrain substructure on Fe-Co deformation behavior.¹⁰ Furthermore, control of the atomic structure, as measured by the extent of the embrittling chemically ordered phases, has been demonstrated in intermetallic alloys through AM and characterized using high-fidelity neutron diffraction. As a layer-by-layer rapid solidification process, AM was employed to suppress the extent of chemically ordered (B2) phases in a soft ferromagnetic Fe-Co alloy (Figure 39).

Another challenge is developing the ability to control the envelope of performance and robustness in a system rather than merely maximizing a single parameter. This will enable the creation of a system that is inherently robust or self-correcting and thereby allows results far beyond the sum of its individual parts.

The potential impact on energy technologies is bold. First, success in this thrust will create pathways to materials that are “born qualified,” enabling the confidence, efficiency and speed necessary for manufacturing surety.¹¹ Second, it will enable the development of methods necessary for a robust circular economy, resource reuse, and extended material lifetimes.

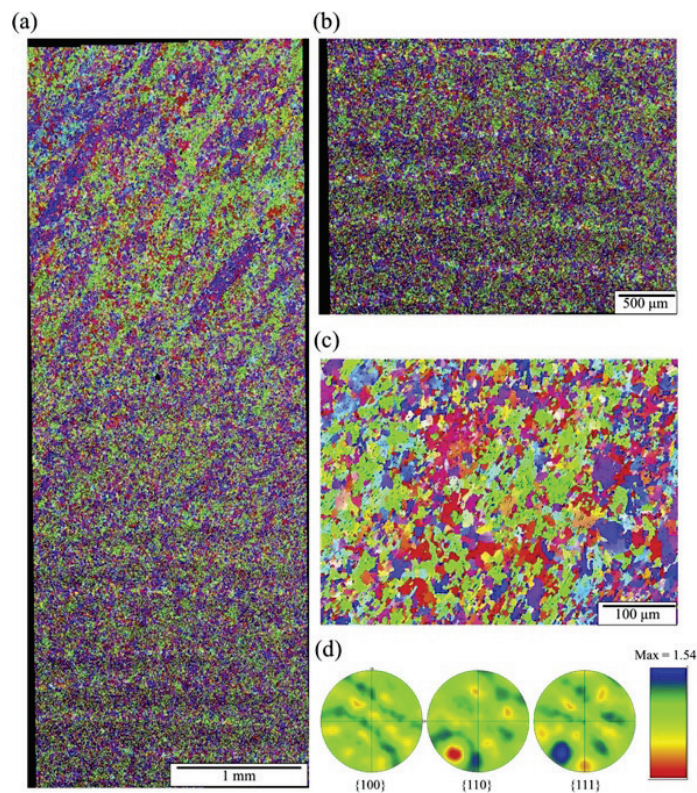


Figure 39. Cross-sections of a representative as-built Fe-Co thin wall specimen processed by laser engineered net shaping (LENS). (a)-(c) Electron backscatter diffraction maps showing the as-built grain structure at various magnifications and locations throughout the specimen height. The grain structure was predominantly characterized by a fine equiaxed morphology. Specimens did not possess any significant macroscopic defects. Source: A.B. Kustas et al. *Addit. Manuf.* 28, 772–780, 2019.

Conclusion

Progress on the fundamental research challenges defined by systems integration science has potential to dramatically improve the agility and resilience of manufacturing processes. Articulating and attacking these challenges will require multidisciplinary efforts that span technology readiness levels. One theme that is common to many aspects of systems integration science is the need to increase the complexity of materials and processes that are considered in fundamental studies. Although this increase in complexity can be challenging, it is vital because of the phenomena that arise only in complex systems and for systems operating in complex and variable environments. A second theme is the need to integrate data from diverse sources. The concepts of multiscale modeling have existed for a long time; but in many cases this concept focuses on using data from smaller, faster, or simpler scales to make decisions at longer, slower, or more complex scales. Successful systems integration science will require methods and researchers who can seamlessly generate and combine data from all scales to enable focusing of experimental efforts and process development.

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Panel 4: Sustainable Manufacturing

Introduction

Around half of global annual industrial greenhouse gas (GHG) emissions are released during the production of steel, cement, paper, plastics, and aluminum. Of the remaining emissions, most can be attributed to the production of chemicals, glass, and the processing of all the above materials into products.¹ Profit motive alone has been sufficient to prompt a steady improvement in the energy efficiency of primary production, largely driven by the desire for reduced energy costs. However, further gains have been stalled, as production of many of these materials has approached practical limits using today's best manufacturing practices.² Therefore, new discoveries in basic science that lead to disruptive innovations are needed to dramatically alter the landscape of opportunity in sustainable manufacturing. There are early signs that such approaches could include the electrification of materials and chemical production, with electricity coming from zero-emission and renewable sources; and reduction of the demand for new materials and chemicals by sustaining or upgrading the value of existing materials through advances in chemical recycling and upcycling. Understanding the impacts of such advances on sustainable primary production and remanufacturing in circular manufacturing systems and associated water use is also important.

Current Status and Recent Advances

The availability of renewable electricity at low cost has created an opportunity to electrify chemical and materials manufacturing.³ By electrifying chemical reactions at scale, it may be possible to reduce carbon for these processes, which currently account for 7% of global GHG emissions. In a future electrified chemical industry, sustainable, ubiquitous feedstocks—such as water, CO₂, and nitrogen—could be converted into high-volume chemicals (Figure 40) such as ammonia,⁴ ethylene,^{5,6} and epoxides.⁷ A key scientific challenge to realizing this vision is understanding how to synergistically harness various forms of energy, including thermal, electrical, radiative, and mechanical energy, to achieve a desired chemical reaction under benign conditions. Basic energy research that overcomes this challenge will involve understanding the coupling of kinetics and transport, with full accounting of thermodynamics. Furthermore, rare species and events at the electrode–electrolyte interface will need to be fully understood, requiring advances in experimental methods of probing these, as well as computational methods for predicting them. More advanced methods of discriminating molecules at an electrode surface will be needed, as will discriminating bonds within a molecule, providing selectivity in electrosynthetic transformations. If researchers are successful in these scientific efforts, a foundation will exist on which stable electrosynthetic systems can be built; this is, in part, because instability is often a series of rare molecular events that, when put together, lead to failure of a system. Through deeper understanding of how to electrify chemical synthesis, mild operating conditions may be achieved, which are conducive to distributed manufacturing close to where renewable energy is generated.

Similarly, for materials manufactured in high volumes—such as cement, the most widely produced man-made material in the world (4.1 billion metric tons per year)⁸—GHG emissions are staggering. For example, as of 2018, each 1000 kg of cement produced net emits nearly 608 kg of CO₂, excluding CO₂ from on-site power generation,⁹ and several gigatons of CO₂ per year will be released in creating new infrastructure, highlighting the opportunity for reducing CO₂ formed during the production of cement.

Current efforts along those lines include carbon capture from flue gases, use of alternative fuels, or development of supplementary cementitious materials. However, flue gas from cement plants is considered too impure for economical carbon capture through amine scrubbing. Advances in selective, cooperative, and high-capacity CO₂ adsorbents (e.g., metal-organic frameworks and other microporous materials) and membrane-based separations with greater tolerance of impurities may provide new opportunities to address the intolerance and high cost of conventional amine scrubbers. The current use of

alternative fuels, such as tires or end-of-life plastics, to power energy-intense thermal processes, while practical in lowering costs, does not mitigate primary emissions tied to converting CaCO_3 to CaO . Furthermore, the use of supplementary materials in concrete has had a limited impact on the carbon emissions from Portland cement and may ultimately compromise its physical properties.

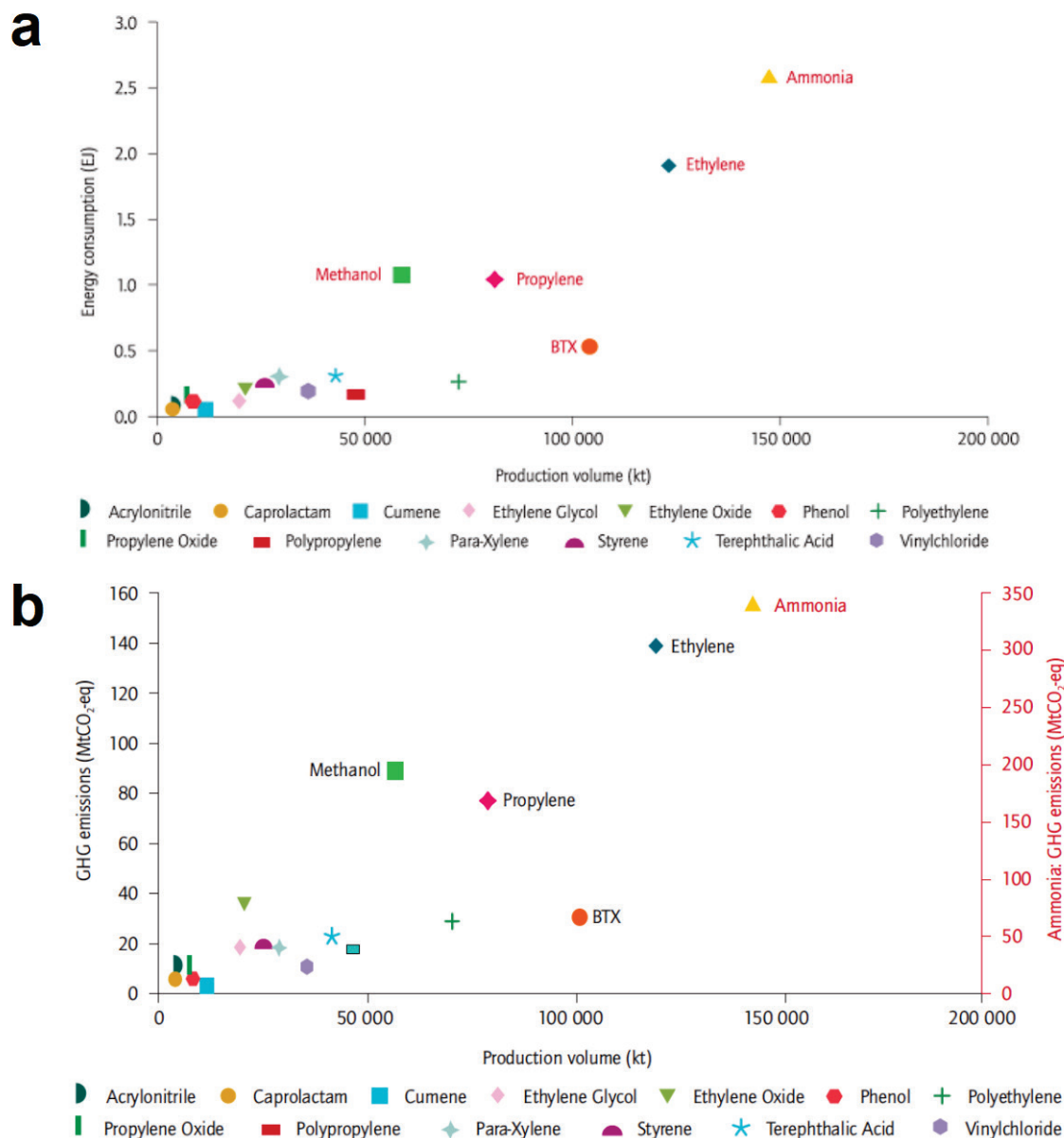


Figure 40. (a) Global energy consumption versus production volumes of the 18 largest-volume chemicals, 2010. (b) Global GHG emissions versus production volumes of 18 largest-volume chemicals, 2010. Source: Reprinted with permission from [Technology Roadmap, Energy and GHG Reductions in the Chemical Industry via Catalytic Processes](#), International Energy Agency, June 2013.

Deep decarbonization of cement manufacturing may therefore require the development of new methodologies that lower the energy intensity and emissions in converting CaCO_3 to CaO , as well as high-temperature calcining and sintering processes downstream in production. To this end, low-temperature electrochemical methodologies may be particularly impactful, e.g., in decarbonating CaCO_3 to Ca(OH)_2 through the design and development of new electrolytic cells and integrated processes.¹⁰

$\text{Ca}(\text{OH})_2$ is an intermediate in the production of many widely used cements and therefore may serve as a strategic entry point for the electrification of cement production.¹¹ If $\text{Ca}(\text{OH})_2$ were produced using low-cost renewable electricity, it might be possible to alleviate both chemical and thermal sources of GHG emissions associated with conventional cement manufacturing practices.

Advances in electrochemistry also are poised to revolutionize steelmaking by lowering CO_2 emissions. Steel is a key material in global manufacturing systems. Its uses span commercial and residential buildings as well as vehicles for transportation, industrial equipment, and various consumer goods. It is also vital to national defense. Shipments from domestic steel mills total ~100 million tons annually. However, the manufacturing of iron and steel is energy-intensive. In 2014, ~1.6 EJ of primary energy was consumed for that purpose, accounting for ~5% of total energy consumption in US manufacturing that year.¹² Current methods for producing iron used in steel involve high-temperature reactions between iron oxide and coke in a blast furnace. This and other steps in the process result in ~0.1 gigatons of CO_2 emissions in the United States annually¹² and 2.6 gigatons of CO_2 worldwide.¹³ While iterative processing and furnace optimization provide near-term opportunities for energy savings, over the long term, it may be necessary to ensure the sustainability of domestic steelmaking through electrification of key processes.

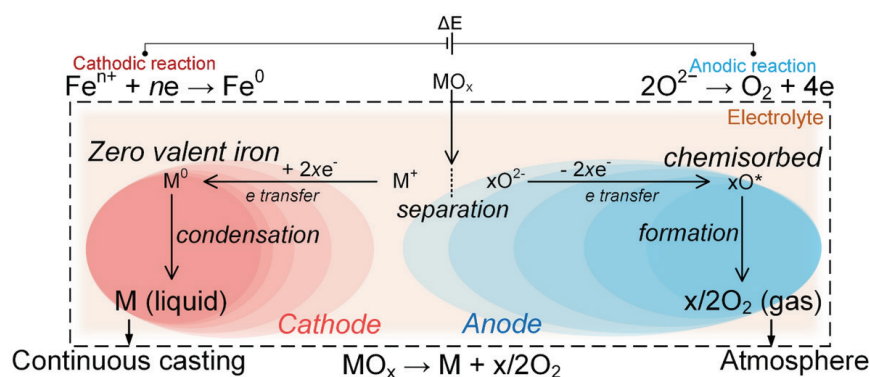


Figure 41. Illustration of the direct electrochemical conversion of a metal oxide (ore) to liquid metal and oxygen gas using electrolysis, which requires two electrodes (a cathode and an anode) and an electrolyte. Such a process could be used to reduce carbon in the production of important metals, including iron for steel-making and rare earths for magnets and other materials. Source: [M. Esmaily et al. *Sci. Rep.* 10, 14833, 2020](#). Licensed under the [Creative Commons Attribution 4.0 International License](#).

but also resistant to transformation or decomposition in the presence of both the molten oxide electrolyte and oxygen gas. The anode should further be resistant to deleterious material reconstructions at applied potentials and at high temperatures. Harnessing the predictive power of high-throughput density functional theory–based (DFT) computational screens for metals and alloys of arbitrary composition and hierarchical complexity may prove useful in identifying new classes of anodes for molten oxide electrolysis. Similar screens may further provide insights into the design of molten electrolytes that are stable to reduction and lower the overpotential.

To develop an understanding of process across scales requires the development of advanced spectroscopic and microscopic capabilities with x-rays, neutrons, and electrons for identifying key intermediates in the redox-driven oxygen atom transfer processes at the anode interface, and tracking of material evolution in space and in time to understand how kinetics and transport influence the efficiency. Operando techniques unraveling the full complexity of the reaction coordinate would benefit from advanced cell designs for such studies, given the extreme environments inherent to the process. Knowledge gleaned for how to direct these primary energy and materials conversion processes in electrolytic cells may be further leveraged for the development of a scalable, practical, and efficient process. To that end, machine

To that end, new classes of molten oxide electrolytic cells have shown early promise in deoxygenating iron oxide, generating metallic iron or its alloys (Figure 41). Molten oxide electrolysis into liquid metal and ideally oxygen gas hinges upon the development of inert anodes capable of sustained oxygen evolution.¹⁴ This requires a deeper understanding than is presently available of how the anode might be designed as both conductive and mechanically robust,

learning could prove useful in establishing a chemistry- and materials-aware framework to co-develop electrolytic methodologies and processes through modeling, simulation, and visualization of complex interdependencies of system variables across scales. Foundational discoveries in basic science regarding how to deoxygenate metal oxides to metals and metal alloys in molten electrolytic systems may find use more broadly, e.g., as means to convert rare earth oxides to rare earth metals.

With regard to the sustainability of battery manufacturing, both lithium carbonate (Li_2CO_3) and lithium hydroxide (LiOH) are used as precursors for the production of cathode powders and metal anodes used in the manufacturing of current lithium-ion and future lithium metal batteries for electric vehicles, aircraft, and the grid. These salts are also used to produce ceramics and glass. Domestically, raw materials for Li_2CO_3 and LiOH are sourced from mines such as those in Clayton Valley, NV, and King's Mountain, NC. LiOH is typically produced using a salt metathesis: $\text{Li}_2\text{CO}_3 + \text{Ca}(\text{OH})_2 \rightarrow 2 \text{LiOH} + \text{CaCO}_3$. This process generates dilute aqueous solutions of LiOH (~3%), from which LiOH is obtained after concentration and crystallization over long periods. When lime is used in this process, calcium impurities are substantial; and removing them increases the time and cost of LiOH manufacturing for key industries that require high-purity LiOH in high volumes. Moreover, a further complication in LiOH manufacturing comes with scale as a result of the voluminous nature of CaCO_3 precipitates.

Strategies for efficiently producing LiOH directly from raw material sources, or more effectively converting Li_2CO_3 or other lithium salts to battery-grade LiOH , are needed to address the sustainability of lithium feedstock production for use in manufacturing and energy systems. By analogy to the sustainable processes identified for the electrification of cement production, electrification of LiOH production from LiCl , Li_2CO_3 , Li_2SO_4 , or other lithium salts from concentrates, leachates, or brines would require the development of new and selective electrolytic systems. It would also require advanced membranes and electrolytes with which it might be possible improve the efficiency of the process. If such advances are realized, highly pure lithium feedstocks may be put to use in the production of lithium metal for future manufacturing of higher-energy-density batteries than are currently available. There are also substantive opportunities to realize circularity in supply chains for battery manufacturing, e.g., in recovering valuable and in some cases critical raw materials, such as cobalt, nickel, and other mineral resources (Figure 42).

Sustainable manufacturing increasingly must deal with not only the means and methods of production but also end-of-life. Modern society has largely adopted linear product life cycles: “make–use–dispose.” For plastics and polymers generally, this approach is particularly wasteful (Figure 43). Approximately 40% of post-consumer waste for plastic packaging is landfilled, functionally isolating it from domestic manufacturing systems.¹⁵ Another 25% is incinerated for energy recovery,¹⁵ which generates GHGs and may release toxic combustion products into the environment near incineration facilities. Some plastics, such as polyvinyl chloride, are not incinerated because of the difficulties in managing the impacts of toxic combustion products. Recycling rates are low and vary widely by polymer. More often, the recovered materials are sorted so that only the most easily recycled streams are processed for reuse, e.g., clear plastics of linear and high-molecular-weight polyethylene terephthalate or high-density polyethylene. Intrinsic recycling opportunities for different plastics are tied to the foundational polymer chemistry and the lability or reversibility of constituent polymer bonds, as well as extrinsic factors such as the prevalence of additives or impurities in the polymer. The latter are typically sorted out using manual and automated processes at recycling facilities, e.g., based on color, density, product class, and other sortable features. Assuming the plastics can be cleaned and sorted, mechanical reprocessing leads to a variety of chain degradation mechanisms that lower the overall molecular weight of the material, which negatively impacts other physical and chemical traits of the polymers and renders them significantly altered.

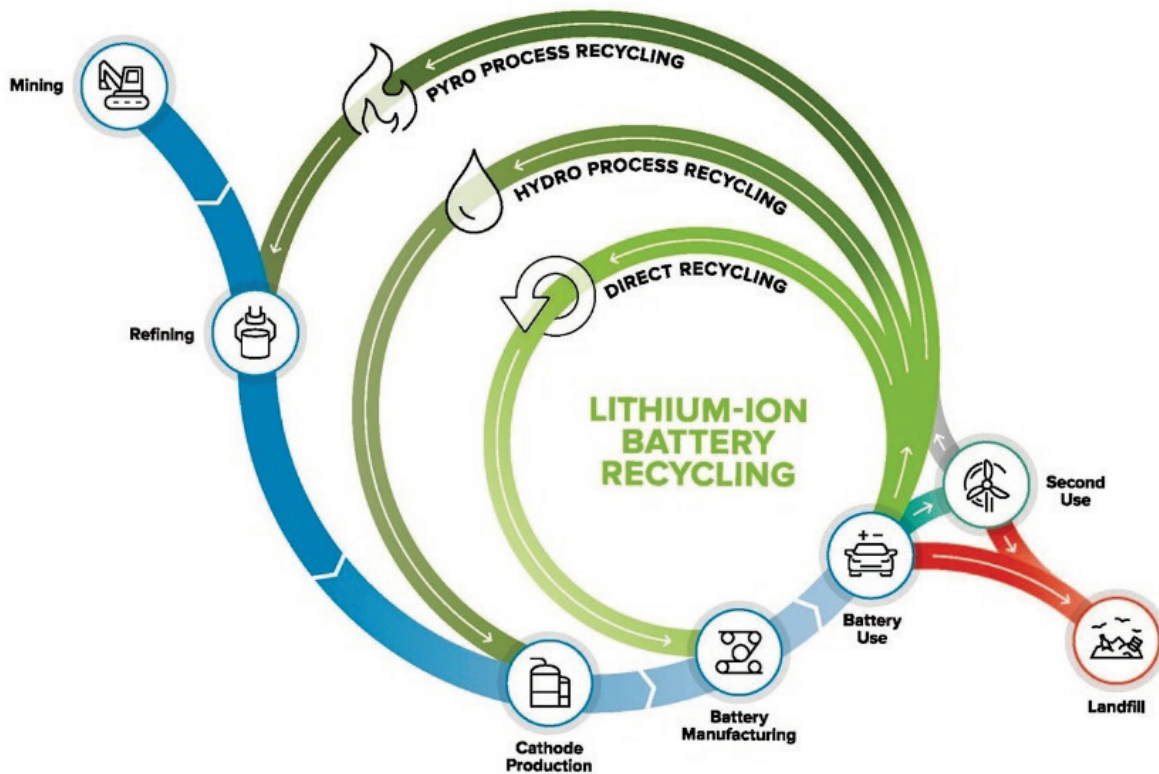


Figure 42. Batteries may be recycled after removing cells from their packs, after which constituent elements are extracted. Most efforts today are focused on the cathode, which contains most of the difficult-to-obtain raw materials such as cobalt, nickel, and lithium. Recycling becomes challenging when cathode chemistries vary. Using today's best practices, only 30–40% of a battery's embodied mineral resources are recovered. Advances in recovery and refinement are needed to realize circularity in battery manufacturing. Source: Image provided by Argonne National Laboratory.

Because of the tendency for polymers to be downgraded during recycling, only ~2% of all recovered plastic waste is returned to the same manufacturing loop for similar product classes. More often, the waste must be mixed with virgin materials to meet manufacturing specifications for conversion into products. The remaining waste typically leaks into the environment, where it may have substantial environmental impacts, largely associated with degradation products and the leaching of toxic additives from the processed materials over time. Whereas energy and monetary investments in a product are lost when the product is disposed of, these investments are retained with circular life cycles that reuse product components, chemicals, and materials.^{16,17} Creating such circular life cycles requires improved understanding and scientific advancements in both materials and the processes related to reuse (e.g., reuse as-is, remanufacturing, and recycling of chemicals and materials associated with end-of-life products).

Global polymer flows, millions of metric tons per annum, 2016

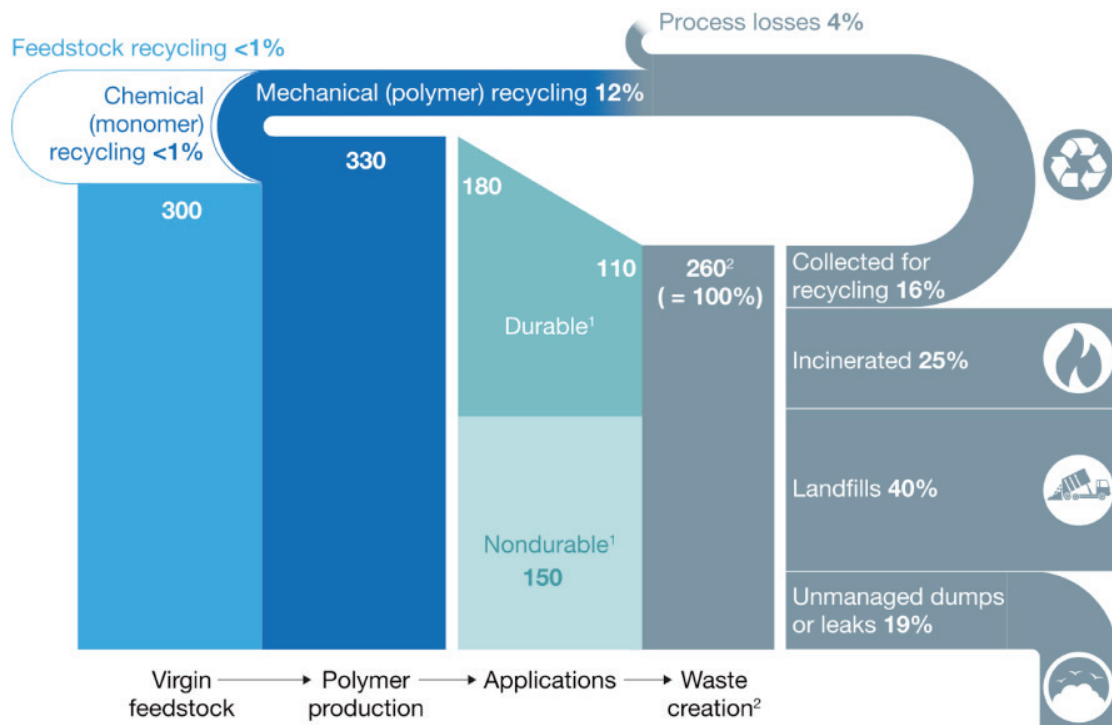


Figure 43. The majority of used plastics go to landfills and incineration, where materials are lost forever as a resource. Only ~16% of plastics waste is reprocessed to make new plastics; the “leakage” into oceans is primarily due to lapses in landfill management or a complete lack of waste-disposal systems. From a resource-efficiency and conservation perspective, this analysis suggests that a huge amount of potential value is currently being lost—value that could instead be captured by better approaches to reusing plastics waste. There are three principal approaches to the reuse of plastics: mechanical recycling, chemical recycling, and processing the plastics waste back to basic feedstocks. All these approaches have suffered from a vicious cycle in which a lack of raw materials due to low rates of recovery of used plastics has limited their growth and dampened interest in their further development and investment; this could now be reversed. Mechanical recycling takes used plastic and physically processes it back to resin pellets, which results in substantial downgrading of the materials due to cleavage of the polymer chains in the process. A key challenge is finding how to preserve the performance quality of resins through recycling steps and avert the deterioration that currently occurs—which may be possible through chemical recycling via which polymers are reverted back to monomers and thereafter refined. *Source:* Exhibit from “No time to waste: What plastics recycling could offer,” September 2018, McKinsey & Company, www.mckinsey.com. Copyright 2020 McKinsey & Company. All rights reserved. Reprinted by permission.

To circularize product life cycles, several challenges related to the design, recovery, and re-processing of products, chemicals, and materials must be addressed. End-of-life product collection processes often allow materials and chemicals to be mixed (e.g., mixed plastics and steel mixed with aluminum).^{18,19} In addition, used chemicals and materials are often contaminated, e.g., food waste on plastic and grease on steel. Products in need of recycling often are made up of several types of polymers, wood, metals, glass, and other materials. Even plastic products may come as mixtures of two or more resins, blends or laminates (e.g., flexible packaging for food and beverage). During use, plastics are oxidized, which subsequently makes them more susceptible to thermal and mechanical degradation during reprocessing. Residues and other contaminants retained in the polymer waste can diffuse and redistribute when processed from melts; that process can lead to further degradation of properties such as decreases in

elongation at break, fracture toughness, melting point, or degree of crystallinity, and even changes in appearance. Trends toward circularization of manufacturing systems have pointed to the need for remedies, which could come via chemical recycling. Although there are current research directions in chemical recycling toward valuable chemical feedstocks (i.e., upcycling), examples of recycling to monomers are rarer. A complete understanding of the opportunity to integrate monomer-to-monomer circularity in polymer manufacturing is needed to determine the impact of such innovations on sustainable manufacturing beyond what already has been accomplished for polyethylene terephthalate.

Scientific Challenges and Opportunities

A molecular-level and structural-level understanding of chemicals and materials is needed to sustainably synthesize them in primary production, e.g., via electrification, and even circularize their reentry in manufacturing systems. Such knowledge is required to design chemicals and materials for processing, use, and reprocessing for their use across multiple, circular life cycles. For example, in the design of the next generation of circular polymers, monomers will need to be chosen not only to deliver specific thermomechanical properties for different product classes but also to deliver on the promise of circularity through energy- and atom-efficient polymerization and depolymerization.²⁰ Hence, catalysts enabling selective depolymerization will be impactful. If the science of pairing catalysts and polymers becomes fully developed in time by nurturing these activities now, it may be possible to kickstart a circular economy of polymers and composites. For the purposes of reprocessing, new methods and approaches are needed that are less sensitive to impurities in feedstocks from recycling streams that may be contaminated or have mixed content. These new methods/approaches also may need to adapt to time-varying changes in feedstock impurities.

A key barrier to effective circularity is the difficulty of recycling heterogeneous waste streams at end-of-life.^{21,11} The creation of materials/chemicals for a given use often involves the creation of a mixture (or alloy), e.g., a polymer mixed with plasticizers and colorants, or iron alloyed with chromium and carbon. The next use cycle may require a different mix/alloy, and it is generally energy-/carbon-intensive to return the materials/chemicals to their unmixed (pure) state. Key scientific challenges include the design of new chemistries for metal alloys and polymers that are resilient to contamination/material mixing during end-of-life processing; development of manufacturing processes that are resilient to suboptimal chemistries from heterogeneous waste streams; and novel low-energy, high-throughput multi-phase separation techniques that can, for example, prevent liquid copper from wetting steel scrap or remove trace iron from aluminum melts. To address these scientific challenges, potential research directions include the study of recycling-friendly materials, the degree to which material properties can be achieved through processing of a limited set of base chemistries, and material detection and separation in different (multiple) phases. The potential scientific impacts include a greater understanding of the thermodynamics of separation at elevated temperatures, and the recycling of immiscible polymers. Consequently, the significant positive impacts on the industrial system will include increased recycling rates and the availability of recycled content in the material supply chain; this will reduce the production of GHG emissions, the energy intensity of the process, and the water use tied to recovery and refinement (e.g., separations). In addition, such advances are expected to lead to increased recycling of materials within the United States, enhancing the nation's material independence and security.

The anticipated demand for more circular materials in manufacturing systems may lead to a disruption of conventional approaches to experimentation to accelerate the discovery process. Even leading-edge approaches for engaging in scientific discovery today are capital-intensive and often require years to deliver potential solutions. In the laboratories of tomorrow, it may be that accelerated materials and process discovery for next-generation manufacturing processes will be aided by autonomous (i.e., self-driving) laboratories where automated experimentation platforms are augmented with artificial intelligence (AI) (Figure 44). Here, novel methods based on AI can be used to generate and subsequently

screen experimental procedures for making and even deconstructing materials by hypothesizing about their outcomes based on previous experiments. Some of these methods may be unassisted while others may be designed to keep an expert in the loop. By either scheme, these loops provide unbiased and informed feedback, which may reduce the number of experiments needed for the discovery of circular materials. When these loops are further executed by robots in the lab, making use of fully automated workflows, autonomous experimentation can even shorten the timeframe in which discoveries are made and, in the long term, the costs associated with discovery.

New sustainability goals and commitments, on the part of industry and of governing institutions and agencies, that reflect a broader societal shift toward a circular economy are placing new demands on materials and manufacturing systems. As a result, companies in the chemicals and materials industries must redesign their products to meet emerging sustainability constraints without sacrificing product performance. To address these challenges, machine learning (ML)-based materials design and optimization capabilities²² could enable us to identify chemistries and processing routes that are most likely to simultaneously satisfy a large number of property targets and constraints. Large, publicly accessible materials data sets would enable the training of accurate ML models and intuitive visualizations²³ that could then be used by product designers to efficiently “design for sustainability” using the results of those ML models.



Figure 44. Modules involved in next-generation self-driving laboratories, which are empowered by artificial intelligence for experiment planning, device scheduling, data analysis, and researcher communication. A control software (top right) orchestrates individual modules of the self-driving laboratories. Modules highlighted in gray are directly involved in the experimentation process, either to plan new experiments or to execute recommended experiments. Modules highlighted in yellow improve the user-friendliness and practicality of self-driving laboratories and facilitate long-term data storage or communication with researchers. Source: Reprinted from *Trends Chem.* 1(3), F. Hase et al., [Next-generation experimentation with self-driving laboratories](#), 282–291, 2019, with permission from Elsevier.

These developments will allow materials scientists to rapidly explore and advance the “Pareto frontier” of materials performance, i.e., the materials that offer optimal trade-offs among many conflicting properties of interest. Visualization tools similar in spirit to Ashby charts²⁴ will enable the designers responsible for the insertion of new materials into products to understand the advantages offered by these emerging, ML-designed sustainable materials. This virtuous cycle of rapid materials development followed by rapid insertion of improved materials into products would enhance the competitiveness of American manufacturers by enabling agility in the face of fast-changing consumer preferences and regulations related to sustainability.

Conclusions

Basic energy research in sustainable manufacturing should address the primary means of production of important chemicals and materials, as well as the means to recirculate those resources in circular manufacturing systems. Doing so will provide unprecedented opportunity to lower the intensity of manufacturing on a global scale. For example, manufacturing processes conducted today using energy-, carbon-, and water-intensive thermal processes might in the future be conducted electrochemically, photochemically, or by coupling several types of energy inputs to direct chemical transformations

selectively and with tolerance of less refined feedstocks. Advances in catalysis using these unconventional energy inputs will provide further gains in efficiency. If those are successful, it also may be possible to increase the recyclability of consumer products and, in stride, increase the amount of recycled content of future manufactured products. For example, catalyst design could focus on selectivity in transformations in chemically heterogeneous environments such that manufacturing can produce a desired chemical or material from less refined feedstocks, as is typical of recyclates. Doing so will increase the resilience of manufacturing systems by providing access to critical resources and feedstocks from waste, rather than extracting them to meet demand, which is growing at an unsustainable rate.

Underlying the scientific challenges in sustainable manufacturing are the fundamental processes by which losses, entropic and otherwise, are managed at all stages of production, use, recovery, and refinement for reuse. To understand these losses will require the development of new in situ characterization capabilities, data analytics, and theory to understand from multiphysics models how kinetics and transport phenomena across scales are tied to fundamental processes associated with chemical transformations in bonds and at active sites in complex reactive environments. Similar insights will be needed to address outstanding challenges in reactive separations, which could be the key to refining feedstocks from recyclates for reuse. Through such insights, it may be possible to design and develop a next generation of hard and soft materials that are intrinsically more recyclable by minimizing entropic losses.

It may be possible to accelerate the discovery of more sustainable materials, manufacturing, and recycling processes by automating workflows through advanced instrumentation, data analytics, and machine learning. Fully autonomous workflows are also possible when processes are coupled to robotics and AI is used to inform and control robotic workflow. AI can analyze the outcomes of experiments to refine hypotheses for the governing behavior and interdependencies to ensure that only the most informative experiments are executed in the next round of tests in pursuit of a goal. It will also allow researchers to test more-complex and nuanced hypotheses in energy and manufacturing systems. These can be executed across scales in ways not previously imagined, resulting in a transformative impact on the generation of knowledge and the foundations of science for sustainable manufacturing.

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Panel 5: Digital Manufacturing

Introduction

Digital manufacturing applies computational tools to simulate and control manufacturing processes—from design to final product. The precise control of these processing steps has led to significant advances in design optimization methods as well as the fabrication of materials with spatially tailored properties. New capabilities are emerging to maximize the performance of digitally manufactured materials against multiple constraints (e.g., maximizing one or more properties, while minimizing the amount of material needed). Digital manufacturing methods have recently emerged that enable voxel-level control of the structure and composition of materials across an unprecedented range of length scales and classes of materials.^{1–3} Such methods also provide the ability to construct digital twins that intimately link process and properties.

Despite many recent advances, digital manufacturing has largely progressed in the forward direction only. A pre-programmed input drives a machine toward a physical output (product) that attempts to exactly match the intended design. Processing outcomes can be agnostic to differences in equipment used, operators, time-of-day, feedstock, and combinations thereof. Manufacturing at digital speeds requires methods that elevate beyond automation, toward autonomous operation in which computational design, process actions, and outcome dynamically adapt based on observation and experience. The end result would be transformative manufacturing methods that are capable of making decisions to achieve the intent behind design requirements, i.e., components that are “born certified” rather than strictly following a predetermined set of processes.

Science Challenges and Opportunities

Rapid developments in advanced manufacturing techniques have led to the integration of what would traditionally be considered materials processing with the manufacturing of macroscopic components. This opens up the design space in the sense that material properties can be varied and tailored through multiple pathways: from chemical to microstructural to macroscopic (including shape, topology, and compositional configuration). This emerging capability brings about a concomitant challenge of constitutive solid properties becoming a function of manufacturing, *rendering digital manufacturing inherently multiscale*. Both processing and manufacturing operations typically employ high temperatures, laser exposure, and sometimes also high pressures, which introduce gradients in stimuli, produce far-from-equilibrium microstructures, and induce phase transitions via a complex, nonlinear process. To accurately predict part performance and quality, modeling must capture the multiphysics nature of these advanced manufacturing methods across multiple scales. The relationships among spatio-temporal structure and composition variations and process parameters are not well understood.

Existing state-of-the-art digital manufacturing methods are extremely computationally demanding; practical use of optimization models and uncertainty quantification is currently not possible. Similarly, on-line monitoring and data processing for nondestructive testing in real time are unattainable today. While advanced manufacturing has opened up the design space to unprecedented dimensions, its exploration and optimization will be possible only through fast and efficient multiscale, multiphysics digital models. This represents a serious challenge because even the highest-fidelity, physics-based models available today are prohibitively expensive. Without a guided search, the amount of collected data is too expansive to explore. Additionally, given the limited understanding of advanced manufacturing processes, it is unclear *what data should be stored and how they should be made available and utilized effectively* (e.g., infrastructure for sharing and security of data). Exacerbated by the inevitable presence of defects and uncertainties within performance parameters, the lack of knowledge of how to plan and to optimize digital manufacturing methods compromises the quality of the part.

Topic 1. Multiscale and multifunctional co-design frameworks that leverage digital manufacturing

The first basic research direction that was discussed for digital manufacturing was the creation of a framework that can discover multiscale and multifunctional material systems. Recent developments in advanced manufacturing techniques have enabled the fabrication of a broad range of multifunctional composites and have spanned an unprecedented range of critical feature length scales within complicated 3D geometries that could not be made before. These capabilities enabled the demonstration of material properties and performance that previously were deemed impossible.⁴⁻⁶ Looking ahead, they promise potential breakthroughs in engineered material systems that may substantially outperform current materials and manufactured products.

Two major barriers prevent the achievement of this paradigm shift in digital design: (1) The design parameter space is complex and requires multiscale and multiphysics computation methods to effectively explore, capture, and achieve the desired performance, which is not possible with current state-of-the-art digital design methods. (2) The complex interactions between material composition, microstructure, defects, topology, and physics of the manufacturing process render the intuition-based designs inaccurate and impractical. Moreover, emergent digital manufacturing models are too computationally demanding. Nascent data-driven machine learning techniques offer a potential route for property characterization and prediction via effective data-driven models; however, large data sets are needed for training. These fundamental challenges currently limit the discovery and manufacturing of new materials designs and architectures that may give rise to superlative performance.

Current status and recent advances

Advanced material design and fabrication at the meso-, micro- and nanoscales can lead to dramatic improvements in materials properties and performance that could not have been imagined previously^{2,7-9} over a wide range of disciplinary applications, as shown in Figure 45–Figure 477. For example, the superlative performance of architected materials fabricated at the laboratory scale primarily stems from controlling their composition and structure across multiple scales, as demonstrated by ultralightweight, mechanically resilient hierarchical ceramic nanolattices shown in Figure 46¹ and the heat exchanger shown in Figure 477.¹⁰ By further expanding these materials sets and scalable digital manufacturing methods, one should ultimately be able to create architected materials or systems with optimum functional performance over the entire property space of interest. However, the design criteria needed to achieve their optimal performance are currently unknown.

A recent series of multiscale optimization studies revealed that ideal system performance usually does not require maximizing or minimizing a particular material property; rather, it is a function of the system configuration, which is typically unknown *a priori*.¹¹⁻¹⁴ Rigorous coupled optimization of material and system configurations can lead to orders of magnitude improvements in the overall system performance.¹¹⁻¹³ In contrast, exclusive optimization of system design may produce a combination of material properties that either are challenging to find through computations or do not even exist.¹⁴ A particular challenge remains to effectively proliferate systems-level design requirements down to the material level.

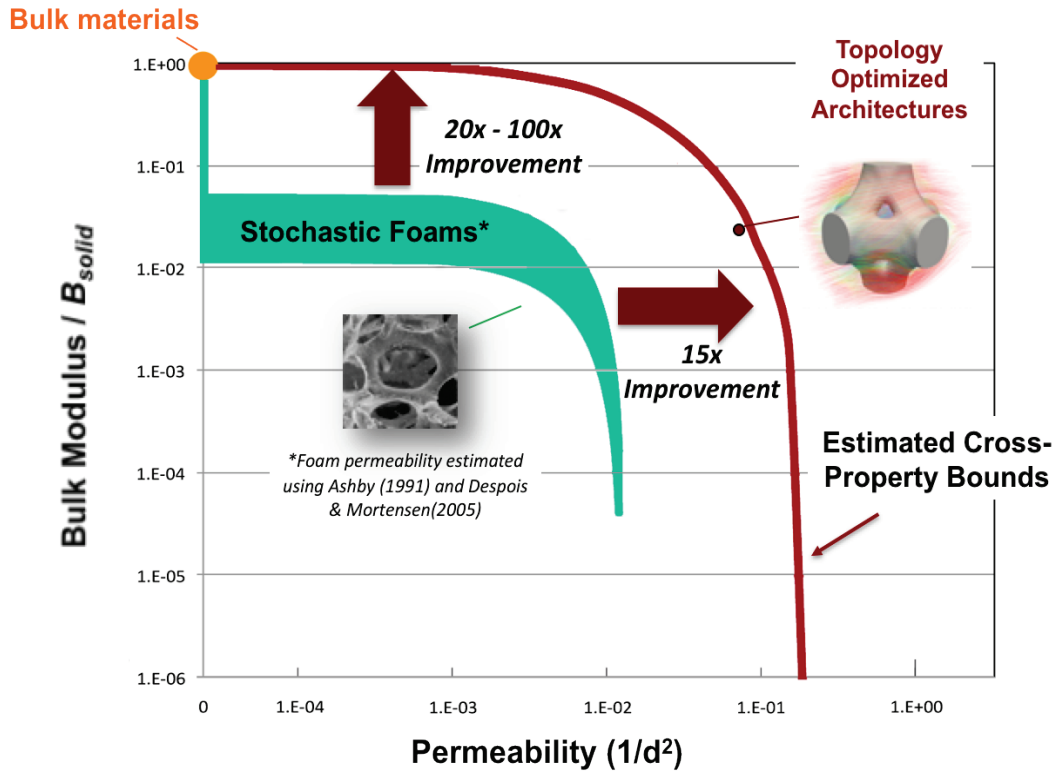


Figure 45. Design for multiphysics and multiple scales can deliver dramatic performance gains that have previously been thought impossible. Source: Adapted from [M. Osanov and J.K. Guest, *Rev. Mater. Res.* 46\(1\), 211–233, 2016.](#)

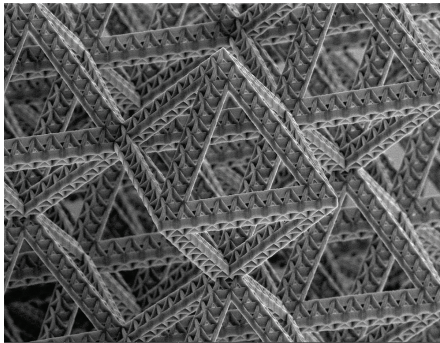


Figure 46. Advanced manufacturing has enabled a realization of an ultra-lightweight, mechanically resilient novel material at nanoscale. Source: L. R. Meza et al. *Proc. Nat. Acad. Sci.* 112(37), [11502–11507](#), 2015.

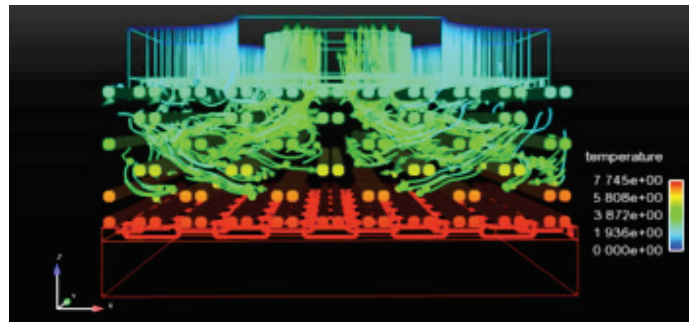


Figure 47. Complex heat exchanger design at material scale. Source: Image courtesy of James K. Guest, Johns Hopkins University.

To construct a co-design framework that can manage the trade-off of conflicting multifunctional requirements, we need basic science advances that enable the modeling, design, and fabrication of optimized systems across multiple length scales. Physics-based models are beginning to emerge that couple across scales and multiple physical phenomena and/or chemistry, and the predictions can be sufficiently reliable to tune the manufacturing protocol and minimize defects.¹⁵ However, the computational resources required for such high-fidelity physics simulations, an example of which is

shown in Figure 48, are extremely costly and do not allow for an iterative approach. Data-driven modeling and machine learning offer a pathway to potentially overcome these limitations; however, insufficient data from new manufacturing technologies coupled with the growing palette of new materials currently limits the successful integration of data-driven techniques.

Scientific challenges and opportunities

A major challenge is the lack of a rigorous pathway to communicate the system-level multifunctional performance requirements down to the relevant material length scale. Materials design is predominantly driven by the required properties, which can serve as surrogates to communicate the material behavior to the system level. However, the optimum material properties are not always intuitively known and are influenced by the system configuration. Digital manufacturing offers a means to encode complex functional relationships that cannot be captured via numerical computations. To achieve maximal performance at the system level, basic science advances are needed that integrate predictive design with digital manufacturing to create new material and/or composite architectures with spatially controlled composition and structure across scales.

A converse challenge is the development of a rigorous methodology to quantify the effects of material design on overall system performance. The current state of the art of material design for a specific set of properties does not adequately inform system performance. The complex and emergent material behavior of multi-material architectures or composites at the system level is lost, along with the potential to tailor material designs. A physics-based multiscale model is needed that rapidly simulates and provides material responses that are propagated to system-level benefits. While existing physics-based models are promising, they are computationally demanding. A potential solution may be to develop new physics-based, data-driven modeling approaches, or novel formulations within existing numerical techniques, that enable low-order and/or multi-fidelity models. To enable data-driven approaches, it is imperative to first establish and define the data to be collected and analyzed. *Developing computationally accessible and reliable predictive models is a critical need for digital manufacturing.*

One important consideration of accurately predicting and understanding material performance and behavior is the type, morphology, and distribution of defects generated by a given digital manufacturing method. A major barrier is the inability to predict and control defects and understand their impact on system performance. Due to the lack of proper description and quantification of how defects affect final material systems, most current efforts focus on simply trying to eliminate defects and achieve the intended “ideal” behavior. However, digital manufacturing offers the potential to exploit and control defect populations, which may prove, in some cases, to be beneficial design features. Rapid defect modeling can guide the optimization of manufacturing parameters (e.g., tool paths, laser flux) to achieve the desired defect engineering. Defects and associated uncertainties can be managed with this co-design framework to ensure overall reliability and safety of the system. This is a critical step toward component validation and qualification.

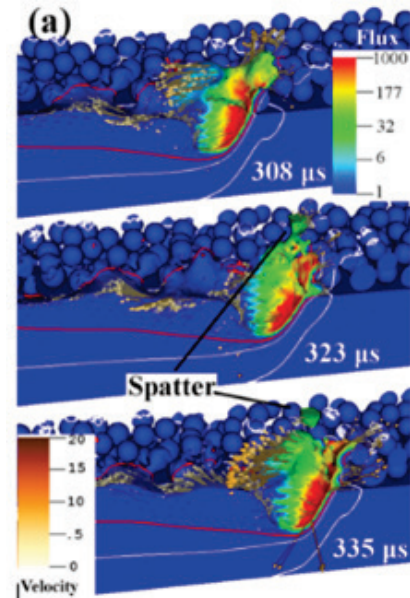


Figure 48. Metal additive manufacturing process simulation is highly complex with many physical interactions and requires an extremely high-fidelity model. Its iterative application for optimization is not computationally attainable. Source: [M.J. Matthews et al. *Acta Mater.* 114, 33–42, 2016.](#)

The final challenge in realizing the full potential of digital manufacturing is the rigorous formulation and development of computational methods that integrate complex multiscale and multiphysics effects with uncertainties to uncover new material and system configurations. Intuitive design based solely on human creativity is unlikely to fully realize the disruptive potential of digital manufacturing. A co-design framework that integrates materials and systems will offer an economical and systematic search of this broad parameter space with confidence supported by uncertainties and defect quantification.

Given the inherent design complexity associated with multiscale, multifunctional material systems, basic research is needed to identify the optimum combination of mathematical and physical insights that augment human intuition.

Conclusion

Several major scientific challenges have been outlined that, if addressed, will enable the revolutionary future of digital manufacturing to be realized through multiscale codesign. First, there is a need for a rigorous computational framework to systematically search the complex multiscale and multifunctional design space and discover new optimal material architectures. This co-design framework should enable the quantified understanding needed to translated system-level multifunctional performance requirements to the materials scale, including the quantification and management of materials parameters, defects, and uncertainty. If implemented, this framework would enable augmented intelligence, which, coupled with human expertise, would lead to disruptive designs and manufacturing methods.

While digital manufacturing is ushering in a new era, the expertise and computational resources required to ensure future commercial adoption at scale remain extremely high. Solving these scientific challenges will enable digital manufacturing of materials and systems with unprecedented performance.

Topic 2. Discover approaches to real-time adaptation and property qualification of digital processes

In parallel with addressing the multiscale modeling and design challenges associated with creating optimized, multifunctional components, this second topic seeks to understand and adapt the dynamic and stochastic processes inherent to many digital manufacturing methods. New approaches are needed to better understand how both instantaneous conditions and processing pedigree impact component quality. The development of in situ assessment of machine and component state relative to a design objective, as well as the exploration of autonomous decision-making during component processing, are paramount.

Current status and recent advances

Although digital manufacturing offers unprecedented control of material chemistry and metrology voxel-by-voxel, the quality of the final component is influenced by many complex multiscale, interdependent factors. The relationships among material, process parameters, and spatio-temporal variations that arise in advanced manufacturing are not well understood. Currently, material performance cannot be predicted well or controlled because of the inability to accurately account for defects.

High-fidelity physics-based models are beginning to emerge that enable the understanding and modeling of materials processes and manufacturing. The existing models are typically complex and take days or weeks to simulate a few seconds of a given process, rendering the current state of the art not suitable to provide rapid insight and feedback to a manufacturing process during fabrication. The computational demands of these models are extremely high and are far from being able to iteratively apply or compute sensitivities to improve the quality of parts. The process design space consisting of control parameters is quite large, and there is a lack of efficient numerical tools to search and determine the most optimal combination of parameters.

One promising approach to highly complex multidimensional problems is machine learning, which requires large data sets that are not always readily available. There is a flurry of research activities in this area across a wide range of advanced manufacturing processes and materials; but the data formats are not easily sharable and there is no common platform that makes the data easily accessible while ensuring security. Given this lack of understanding, it is unclear what data should even be stored. This presents a major obstacle in developing a machine learning approach to understanding digital manufacturing methods.

To collect these data and enable real-time adaptation and qualification, sensors, monitoring, and nondestructive evaluation techniques must be integrated within digital manufacturing platforms. For example, additive manufacturing techniques involve highly dynamic and multiphase processes; and the patterned materials often experience locally extreme environments. Most sensors used today are not suitable for these conditions, driving the need for new sensor technologies capable of real-time, in-line monitoring. In addition to hardware, in situ assessment requires fast and reliable data processing.

Scientific challenges and opportunities

Given the complexity of challenges associated with predicting and pre-defining the design and processing parameters, the quality of digitally manufactured materials, components, and systems involves a high level of uncertainty and is considered unreliable. It is therefore pertinent to develop this capability for real-time adaptive control and qualification to ensure that manufactured parts can be born-qualified.

The emergence of high-fidelity, multiscale, multiphysics models is providing insights into the complexity of advanced manufacturing and processing. In addition, machine learning opens new avenues to fast simulation. Combining data-driven and physics-based approaches, new opportunities exist in developing algorithms that drastically reduce the requirement for large amounts of data. Building on machine learning trained by in-line data, it is expected design and processing parameters can be varied in real-time during manufacturing to ensure reliable part production.

While high-fidelity multiscale models are beginning to show potential, many challenges remain. Physical interactions at the atomistic level give rise to complex behavior at the microstructural and continuum scales which is not intuitive and cannot easily be predicted. Many uncertainties exist at each length scale, with different underlying physics at each one. Recent advances in model fusion and uncertainty propagation can be used to quantify the uncertainties at every scale and their sensitivities to the design features and processing parameters. As simulation speeds and predictive capabilities accelerate, it is anticipated that real-time adaptive control needed to ensure quality will be realized.

Another opportunity area involves the coupling of materials, component, and system level design. Today, material development, topology design, and manufacturing are decoupled. However, a co-design methodology that seamlessly integrates material properties and processing parameters would prove highly valuable. Early work in this area has revealed that topology design is highly sensitive to the material properties and processing and vice versa. Real-time data of materials properties and processing parameters can be used to predictively guide digital manufacturing methods, so that their final products are reliable and satisfy functional requirements.

Conclusion

Developing a closed-loop system that monitors manufacturing processes and part quality in-line and autonomously controls design and process parameters in real time would ensure that digitally manufactured parts are born qualified. This revolutionary breakthrough is now within sight given the emergence of new multiscale multiphysics modeling, machine learning, exascale computing,

optimization, multifunctional materials, scalable digital manufacturing methods, and nondestructive evaluation systems.

Final Remarks

Materials synthesis, manufacturing, and qualification are tightly coupled. By exploiting this coupling through a fully digital framework that integrates fast process models, data from in situ diagnostics, and machine learning algorithms that work in concert, an *adaptive, autonomous manufacturing environment* can be achieved. To realize this vision, many challenges must be overcome, including the development of fast, multiscale multiphysics models, digital surrogates, collection of in situ process data, and predictive, actionable outcomes. Overcoming these barriers requires transformational scientific advances that allow digital manufacturing to provide a scalable pathway from design to production of qualified parts. The critically needed areas of research and the key ideas identified by this panel led to PRD 2: Integrate Multiscale Models and Tools to Enable Adaptive Control of Manufacturing Processes and also influenced the development of PRD 5: Co-Design Materials, Processes, and Products to Revolutionize Manufacturing.

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Panel 6: Crosscutting Themes

In parallel with progress in the fundamental synthesis science, critical scientific advances are needed to enable the translation of new laboratory-scale discoveries to manufacturing scales suitable for meeting consumer demand. Up to the present time, many new material discoveries have failed to advance beyond successful early-stage technology readiness levels; this workshop was intended to identify the basic science research needs to enable progress beyond this point, i.e., transformational manufacturing.

Panel 6 was tasked to identify cross-cutting themes, and to that end the panel participants initially were dispersed across the other topical panels and later reconvened to discuss and capture research needs spanning multiple research areas. This panel report captures these crosscutting issues from each of the original theme panels, many of which were explicitly incorporated into the subsequent PRD panels. Each of the theme-based discussions is presented, followed by an enumeration of key cross-cutting areas.

Theme 1. Precision Synthesis Science

The critical role of defects in synthesis was a major discussion point. To fully achieve future advances, it will be important to develop a detailed scientific understanding of defects with the aim to achieve enhanced detection, control, and/or elimination of defects across multiple length scales at all stages of manufacturing.

Often defects can arise from the raw materials themselves, and as a result, high-purity materials may be used for initial fundamental studies. However, raw material availability and affordability can be an obstacle when the synthesis needs to be transferred to larger volumes. At large scales, it may be difficult to achieve raw material purity comparable with materials used at the lab scale; it is therefore critical to understand the product sensitivity to impurities and how the manufacturing process impacts materials selection and vice versa. Often these issues are not considered during basic research phases and thus could provide an additional barrier to moving novel approaches to higher levels of technology readiness.

Advanced predictive tools can offer support to direct synthetic strategies. Artificial intelligence (AI) and machine learning (ML) approaches to autonomous exploration in materials fabrication were identified as significant future capabilities, but they will require high-quality data sets to enable predictive physics-based models and ML approaches, with AI-enabled redirection on the fly. The data required to achieve this goal may not yet exist, and the computational tools are still very much in their infancy. The solution is the combined development of physics-based theory feeding computational ML advances, using data collected from advanced characterization techniques.

New synthesis, processing, and characterization tools will be needed for this effort to be successful, including digital synthesis, high-throughput precise characterization tools, rapid in situ methodologies, and expanded methods to identify structure and function signatures—accessed and used for real-time feedback control. There is a need for real-time synthesis and characterization tools for monitoring the progression of assembly for feedback into the synthesis process, and a need to adapt the novel methods developed at user facility beamlines to achieve production floor on-line and in-line characterization.

Theme 2. Processing and Scale-Up Science

The Processing and Scale-Up Science discussions also addressed defects (similar in scope to the Precision Synthesis Science discussions), but with the additional consideration of self-healing mechanisms. In fact, it was noted that defects can be beneficial and can be deliberately induced in some cases, although more fundamental knowledge of the defects themselves is needed to fully utilize these approaches. There was much discussion that “out of equilibrium” processes, along with associated characterization and modeling are particularly challenging, but are essential to enable advances in transformational manufacturing.

Physics-based multiscale modeling is used to connect measurements to properties for translation to scale-up. While AI is promising, present AI is insufficient and should be used to complement, not replace, more conventional physics-based models. These methods identify key properties to be measured.

It was recognized that scale-up and process design require large volumes of data. Because processing and rapid interpretation are needed for process control and future use, strategies for reducing large data streams and reduced models are needed to allow more rapid response. For example, models and processes that work at the lab scale for catalyst systems may not translate to the commercial scale. Therefore, understanding how to measure and use operando techniques so that the collected data are useful inputs to models of large-scale behavior will be key criteria for future implementation of these approaches. Moreover, there are needs for developing advanced sampling methods so that operando probes can be made to work for scaled-up processes. For instance, the high depth of penetration afforded by neutron beams in most engineering materials, combined with the time structure inherent in pulsed neutron sources, enables time-resolved and spatially resolved mapping of structural changes under manufacturing or operating conditions (Figure 49). However, in situ and operando studies present many challenges and require the identification of solutions for practical applications. The reason is that real parts have complex geometries that may not be well matched to the available sources; the grain size, morphology, and texture may be complex; large samples and complex sample environments may yield beam pathways that significantly attenuate the beam; and some materials are prone to activation during exposure to a neutron beam.

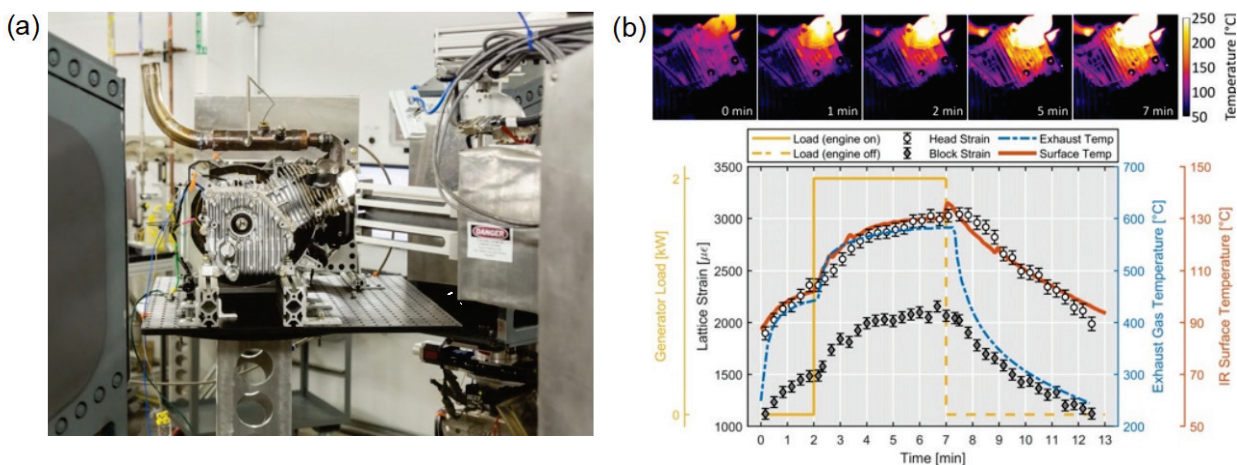


Figure 49. (a) An internal combustion engine manufactured from a novel high-temperature aluminum alloy mounted on the VULCAN instrument at the Spallation Neutron Source for operando characterization using neutron diffraction. (b) The scattering volume is precisely located at defined mapping points within the engine, and the data acquisition was synchronized with the spark timing to nondestructively resolve changes in temperature and lattice strain. Source: Reprinted with permission from M.L. Wissink et al., [Operando measurement of lattice strain in internal combustion engine components by neutron diffraction](#), *Proc. Nat. Acad. Sci.* 117 (52), 33061–33071, 2020. Used by permission under [Creative Commons International License 4.0 \(CC-BY-NC-ND 4.0\)](#).

Theme 3. System Integration

As with other theme panels, within the System Integration area, there was considerable discussion of metrology and measurement capabilities as they relate to manufacturing sciences. The development and utilization of AI, ML, and data science approaches were identified as critical enabling approaches for better design of experiments. However to enable seamless integration of these components so that they will be successful in an increasingly competitive economic environment, it was emphasized that understanding technoeconomic thinking and tensions between various performance and economic tradeoffs could provide an opportunity for driving basic science aimed at transforming manufacturing—

i.e., that “use-based” science needs to be aware of product needs as one inspiration driving materials and process science advances. This concept aligns with some of the discussion from the Precision Synthesis Science topic, in which the role of raw material purity was identified as an area that often plays a significant role in the final manufacturability. Building in those insights right from the discovery may open up entirely new approaches not currently considered.

Historically, basic materials design often looks at optimizing one criterion in isolation. Exposing multiple criteria, throughout the scientific-to-processing stack, may develop new knowledge and opportunities (e.g., energy consumption, life cycle) and lead to more robust outcomes. Such an approach requires and is informed by multimodal data and multiscale simulations and is important for both scientific discovery and diverse downstream uses. Exposing the underlying criteria is necessary for complex design of experiments, multi-objective optimization, and robust design under uncertainty.

Another challenge identified was integrating characterization into the workflows. To that end, advanced characterization techniques are needed to better serve manufacturing science needs: e.g., enabling studies at temperatures and under conditions that are matched to processing and end-use conditions. It also was noted that what is of value is “images vs. digits,” that is, *information* rather than simple *data*. Data are relatively easy to acquire; instead, the challenge is to better extract all the information from the data. Capture of (anti-) correlated failure modes can be of great value in process design.

Once again, the storage of large curated data sets, enabled to be readily accessible for use, was identified as an important issue in System Integration. Organizing such data into usable form poses an immense challenge, but the payoff could be very significant and thus could justify the investment.

Theme 4. Sustainable Manufacturing

Strategies for controlling transformation in matter, from individual bond levels to designing materials with their full life cycle in mind, were a focus during the Sustainable Manufacturing discussions. A circular approach is needed such that all energy input requirements for transformations are defined for optimizing and reducing the energy footprint. To be successful, consideration needs to extend beyond the raw materials and initial product manufacture to include end-of-life and disassembly/recycling processes.

As part of the movement toward improved circularity, there is an opportunity to focus ML on integrating multimodal/multiple criteria initially for optimization, rather than focusing it solely on discovery activities. Additional benefits will result from following materials throughout their life cycles and using information technology approaches. As these areas develop, it is also important to recognize that limitations for recycling of materials are often human oriented—e.g., with regard to cost, inconvenience, and acceptability—and not technical in origin. Factoring these considerations into the development of recycling methodologies is critical to ensure their commercial adoption. Likewise, there is a need for advanced/rapid analytical characterization methodologies coupled with algorithms to reformulate and predict performance in real time to build in flexibility for managing heterogeneous and inconsistent feed materials as a result of circularity and human error in sorting. Going forward, identifying the optimum in designing for performance, while simultaneously designing for end-use recycling/reuse, will be needed to enable more sustainable manufacturing approaches. Advanced knowledge around both materials synthesis/manufacturing and materials performance over time will need to be developed.

Theme 5. Digital Manufacturing

Digital manufacturing is essentially the application of computational systems to manufacturing processes. The key elements involved are multiscale process modeling with topology design, computer-controlled synthesis and processing, real-time process monitoring with advanced sensors, data-supported uncertainty management, and quality assurance. Material-process-product co-design and process control with high

precision are desired features of transformative digital manufacturing. These rely on our understanding of the fundamental physics underlying energy-matter interactions, as well as the driving forces controlling the assembly of basic building blocks across different length scales. There are many opportunities for morphological and topological optimization in this area. This knowledge can be created via a concerted effort exploiting multidimensional, multimodel materials characterization, process monitoring and sensing, and multiscale simulations.

Multidimensional characterization expands the concept of 3D spatial characterization of materials by adding the fourth dimension of time, plus additional processing conditions or service environments such as temperature, pressure, and stress. In the context of transformative manufacturing, multidimensional characterization examines the dynamic evolution of materials structures during the synthesis or manufacturing process. The DOE-BES user facilities provide great resources for in situ/operando characterization of advanced manufacturing (see the sidebar). Electrons have strong interactions with most elements and can be focused into very small probes to afford extremely high spatial resolution in microscopy techniques. Neutron beams have great penetrating power and isotopic sensitivity. Photon-based techniques are highly versatile and often strike a good balance between the sample dimension and the temporal resolution key to dynamic processes. Integrating these capabilities and sophisticated operando systems will enable researchers to interrogate critical materials problems across multiple spatiotemporal scales—particularly those associated with synthesis and processing under far-from-equilibrium conditions, as well as prediction and management of defects and rare events. In addition, many lab-based characterization and metrology tools are essential for measuring different process parameters and materials structures relevant to advanced manufacturing.

Sidebar: The DOE-BES National User Facilities—Enablers for Transformative Manufacturing

There are 12 BES scientific user facilities across the United States. They comprise five light sources, of which one is a free electron laser, two neutron sources (one spallation and one reactor based), and five nanoscale science research centers, which include electron microscopy centers. These facilities are located at the DOE national laboratories and are national treasures that, taken as a whole, have no equal anywhere else in the world. These facilities constitute a unique resource for the characterization of materials and chemical processes at the highest level of spatial, spectral, and temporal resolution, in addition to the synthesis and exploration of matter at the nanoscale. More than 15,000 researchers annually from academia, national laboratories, and industry access these facilities free of charge, primarily through a merit-based peer-review system, provided that the research is nonproprietary. (Proprietary research can also be conducted for a nominal fee). The light sources, neutron sources, electron microscopy centers, and nanoscience centers each have a specific role to play in furthering transformative manufacturing.

The light sources produce x-rays that are many millions of times brighter than those available in laboratory settings. Scientists use these highly focused, intense beams of x-rays to reveal the atomic structure in a wide range of materials, including metals, semiconductors, ceramics, polymers, catalysts, plastics, and biological molecules. Wavelengths can be selected over a broad range to match the experimental need.


The neutron sources provide a versatile probe of the atomic and molecular arrangement, motion, and magnetism in materials. Neutrons penetrate through many centimeters of a solid and so can determine the material structure in situ or provide radiographs of technological devices such as engines and batteries under operating conditions, i.e.,


operando characterization. They have unmatched sensitivity to hydrogen and other light elements and can distinguish different isotopes of the same element.

The nanoscale science research centers provide facilities, equipment, and expertise for the synthesis and study of materials at the nanometer scale—probing single atoms, clusters of atoms, and molecular structures. The scientific theme of these centers is to observe, understand, and predict how these nanosystems function. Together with the electron beam micro-characterization centers, they provide access to cutting-edge instrumentation for direct imaging and micro-analysis of materials at the atomic and molecular levels. In many cases, these instruments now incorporate in situ environmental capabilities, such as gas reaction, tensile, or ion bombardment chambers. Advances in specimen preparation have enabled the atom-level analysis to take place at specific sites in technological materials. These instruments provide structural and chemical information over critical length scales complementary to those probed with neutrons and x-rays.

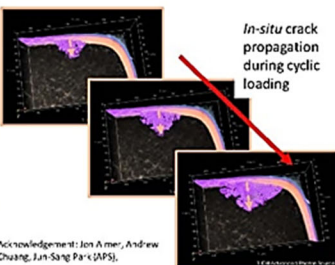
Taken together, these user facilities present an opportunity for developing an understanding of complexity in situ on the relevant length scales across relevant time scales, which will provide an in-depth understanding of how manufacturing processes affect materials' properties and, in turn, the functional performance of a device. This understanding will allow a researcher to build materials “from the bottom up,” atom-by-atom and molecule-by-molecule, so that each atom/molecule plays a prescribed role in producing the macroscopic materials behavior.

Microstructural responses under testing conditions









Deformation mechanism of titanium alloys was studied at 1-ID-E beamline at the APS using an axial-rotational load frame developed by AFRL.



In-situ crack propagation during cyclic loading

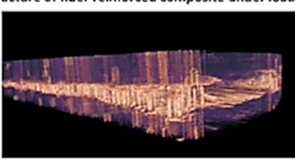
Acknowledgement: Jon Amer, Andrew Chuang, Jun-Sang Park (APS).

Tension Bending Wedge DCB Acoustic emission

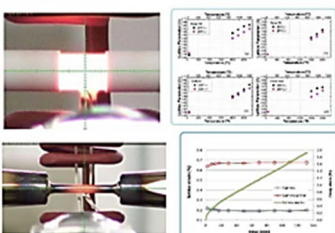

Microstructural responses under various modes of mechanical deformation were studied by in-situ tomography and acoustic emission⁴ at 8.3.2 beamline at the ALS.

Fracture of fiber reinforced composite under loading




Acknowledgement: Dalia Paricson, Harold Bernard (ALS)

Ni-base superalloys for gas turbines were studied at high temperatures (top)² and under both temperatures and load (middle)³ at the Vulcan beamline, SNS.

Engineering diffractometer

Combined in-situ characterization and testing is key to materials innovation



Source: Image is from a presentation at the workshop and was provided by Yan Gao of GE Research.

Digital twinning is a fundamental part of digital manufacturing. The digital twin associated with a manufacturing technology synergistically integrates physical science and data science and provides numerical tools for designing, predicting, and controlling the process. The computational models involved need to be developed or further improved with the experiment data created via multidimensional,

multimodal characterization. Moreover, in-process, closed-loop control systems for manufacturing necessitate the development and deployment of advanced sensors and innovative monitoring frameworks to bridge the factory product line and the virtual space.

Identified Crosscutting Themes

Potential crosscut PRD/themes were identified, such as multi-material architectures, structural process/materials, complex geometric structure control, and uncertainty within models and how it is propagated. On-line monitoring tools also were recognized as a broad need (e.g., data science/cyber security) for digital manufacturing. Underlying all these themes is the dedicated focus on data management required to support and enable the advancement of manufacturing sciences. These concepts are described in more detail in the following pages.

Detection, Prediction, and Management/Control of Small Defects and Rare Events

Current status and recent advances

A major challenge in scaling up of laboratory processes is the increased role of critical flaws, rare events, and trace impurities—perturbations on the meso- to nanoscale that impact properties on the macroscale. For energy materials such as photovoltaic thin films, it is now known that defects introduced during manufacturing may create traps that inhibit carrier mobility and can dramatically reduce device performance. Similarly, in structural materials, small microstructural defects (vacancies, impurities, dislocations) alter the local free energy compared with the macroscale and may result in unexpected properties or trigger local phase transformations.

Scientific challenges and opportunities

There is a critical research need to develop sophisticated computational tools to model this behavior. Once the impact is understood, the next challenge is to control the materials synthesis and processes to the needed fidelity for the final product. Robust science-based modeling tools are required to evaluate between the development of defect-tolerant processes versus defect remediation, elimination, or self-repair. In the latter cases, improved metrology is required. There are also opportunities to implement rapid analysis tools that can be used at a beamline during an experiment with the expectation of steering its progress: see, e.g., the *Cinema* toolkit, <https://github.com/cinemascience>.

Precision Synthesis with Integrated Closed Loop Platforms

Scientific challenges and opportunities

To realize precision synthesis, on-the-fly processing platforms must be built, which require integrated suites of detectors, computation/AI/ML approaches, and stimuli/processing techniques. Development of these platforms requires interdisciplinary approaches that include detectors, software/ML, predictive modeling, and materials synthesis/optimization tools. With spatio-temporal and defect-level control, deterministic synthesis can be achieved across all length scales. Notably, applications in quantum devices, patterned assemblies, and tailored defect-level additive manufacturing (AM) are enabled. Novel “composite” materials with multiple, coordinated functions, can be created. Gaps in length scales will be closed, and high-throughput synthesis can be realized.

Co-design of Nonequilibrium Materials and Processes

Current status and recent advances

Processes inherently occur under the conditions that are far from equilibrium. Nonequilibrium materials and the processes to produce them have traditionally been developed by trial-and-error and through accidental discoveries. Whether it is a chemical transformation, solidification, polymerization, shear

processing, or quenching, such processes rely on changes over time and length scales that are often larger than the characteristic scales over which a material can relax or readjust. When used deliberately, such processes can trap materials in metastable, nonequilibrium states whose properties do not exist at equilibrium. Examples include strong polymer fibers and composites or efficient organic photovoltaic thin film materials, in which molecular orientation is anisotropic and is achieved by processing operations such as extrusion or blade coating, respectively.

Scientific challenges and opportunities

Access to vast amounts of data, predictive physics-based models, and fast computational algorithms provide an opportunity to understand and monitor materials transformations at an unprecedented level of detail, at molecular length and time scales. This information could be used to identify molecular pathways leading to promising nonequilibrium states; that information would then inform process development, product durability, and sustainability over the full lifetime of the product, including disassembly or recycling. The opportunity exists now to manipulate matter down to molecular length scales in large-scale processes. As an example of lithography at the atomic scale (Figure 50), Alemansour et al.¹ have recently demonstrated the use of a single layer of hydrogen atoms as a mask on silicon, which points the way to device patterning with atomic resolution.

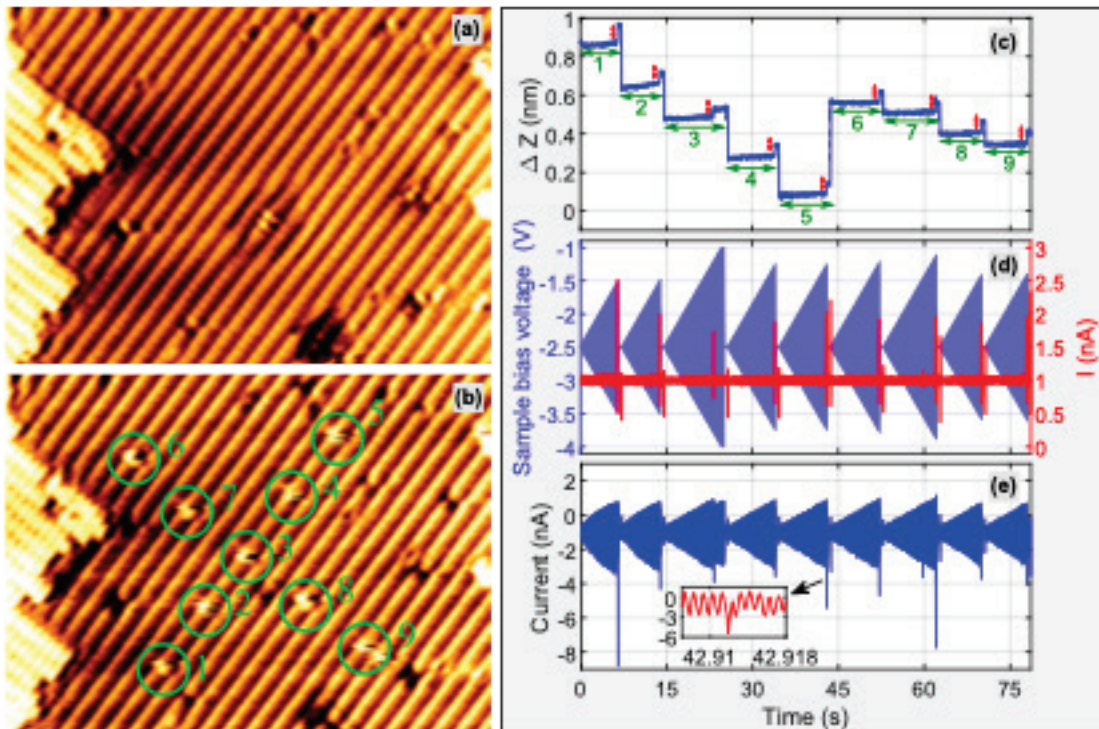


Figure 50. Scanning tunneling microscopy image of a Si(100)- 2×1 :H passivated surface (a) before and (b) after depassivation using voltage-modulated feedback-controlled lithography. Each desorption event is numbered in (b) and its corresponding displacement in the Z direction is shown by the same number in (c). Also, the tunneling current (d) after and (e) before the notch filters are measured. The removal of a hydrogen atom is detected as a jump in the current or the height. Source: H. Alemansour et al. [Controlled removal of hydrogen atoms from H-terminated silicon surfaces](#), *J. Vac. Sci. Technol. B* 38, 040601, 2020.

Supply Chain Transparency for Sustainable Manufacturing

Current status and recent advances

To ensure supply chain integrity around both safety and sustainability issues, there is a need to be able to monitor sourcing and identify suspect or counterfeit materials. Development and tracing of material supply chains also will enable improved understanding of how issues like raw material purity affect final manufacturability. An array of research could help support efforts to address this fundamental challenge—sensors, embedded design principles (holograms, active materials), and security measures like blockchain-type approaches to enable monitoring of the entire length of the supply chain. Such efforts would enable advancement across several different fundamental research areas which could be applied much more broadly than simply to protect supply chains. A major impact would be to preserve the value proposition for recycled materials and/or bio-sourced materials. Having a mechanism to protect and trace supply chains would prevent substitution of these new sources by less expensive/traditional materials and protect nascent industries that ultimately will be more sustainable. The DOE Critical Materials Institute is an example of investment in energy-critical materials processing that pays attention to reuse and recycling. Recently, *ab initio* calculations have been performed by Sutton et al.² to explore the space of compounds that might bind cerium, as an example of a valuable rare earth, and extract it efficiently from minerals such as Ce-bastnašite and Ce-calcite. Figure 51(a–d) shows examples of various configurations of ligands on mineral surfaces that lead to the computation of binding energies. Figure 51(e) shows how the binding energies vary for various types of configurations for a large set of potential ligands, narrowing the range of experiments that need to be performed to identify, in this case, optimum processing for cerium extraction. Such calculations can greatly speed up the search for suitable processes for efficient processing of critical materials. The accumulation of simulation results also is significant in the space of data curation and building libraries of results and techniques for general use.

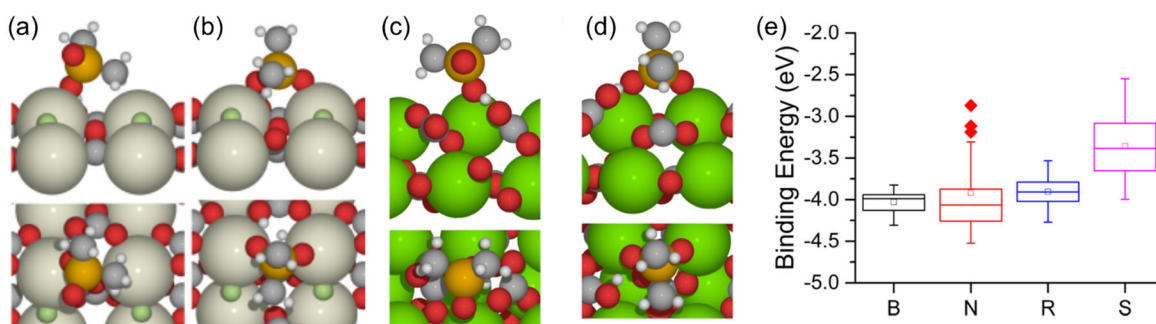


Figure 51. Images (side and top views) of (a, c) monodentate and (b, d) bidentate DMP bound to the most stable facets of (a, b) Ce-bastnašite and (c, d) calcite. Large spheres are Ce (gray) and Ca (green). Medium spheres are C (gray), O (red), F (light green), and P (orange). Small, off-white spheres are H. Each panel shows approximately a single 1×1 unit cell, and images are approximately to scale. The monodentate DMP on Ce-bastnašite is dissociated into an ion pair with the proton on a neighboring surface O (partially occluded by a C atom), whereas the monodentate DMP on calcite is a neutral molecule binding through its OH group. (e) Box plots showing the distribution of binding energies as a function of the binding configuration: (B) bidentate, (N) neighboring, (R) repeated, and (S) staggered. Boxes denote the 25, 50, and 75% values for the binding energies; whiskers mark the upper and lower bounds of the data, excluding outliers. Outliers are points outside the interval centered on the median and bounded by median ± 1.5 IQR, where IQR is the distance between the 25 and 75% lines. If the IQR range extends past the actual minimum or maximum, then the actual minimum or maximum is instead used as the lower or upper bound, respectively. Source: J.E. Sutton et al., [Molecular recognition at mineral interfaces: implications for the beneficiation of rare earth ores](#), *ACS Appl. Mater. Interfaces* 12, 16327, 2020.

Basic Research Needs for Additive and Other Transformational Manufacturing

Current status and recent advances

It is critical to understand a material's behaviors in a dynamic, nonequilibrium system, and the complex microstructure and property relationship both during processing and under use conditions. An *integrated* approach is needed to develop new materials and to break the traditional boundaries between basic and applied research—asking system-level questions during the basic research stage and recognizing the boundaries are blurred in many areas, such as energy storage and AM.

New advances in characterization using scattering methods are needed to meet the required temperature, pressure, and other environmental conditions for samples with the appropriate geometry to generate high-quality data that are suitable to resolve issues related to materials manufacture and use that are generally applicable across a range of experimental needs. As an example, Zhao et al.³ recently identified a behavior at the tips of the “keyholes” that occur during laser melting of metals in AM that may explain how pores are pushed away from the moving keyhole and trapped by solidification (Figure 52). This analysis relied on the use of high-speed operando x-ray visualization of the melting and vaporization that occurs under a high-intensity laser light. The impact on AM is that the generation of porosity via keyhole instability has a well-defined boundary in power-velocity space that users of laser powder bed fusion will be able to avoid.

Scientific challenges and opportunities

There are important gaps in the basic research tools that are needed, both at the laboratory scale and at the national user facilities, to enable high-fidelity characterization under dynamic, nonequilibrium conditions. New strategies are needed for integration of processing and characterization (operando), integrated testing and characterization (in situ) methods, and synchronization and integration of multiple simultaneous characterization methods. There is now a significant opportunity to use advanced computation methods, including AI and ML, to rapidly process raw data and convert data to information that can be immediately input into computational models to modify experiments in real time so that the information generated is relevant to realize advances in manufacturing. It is expected that an enhanced ability to characterize far-from-equilibrium processes will drive the development of computational models in this regime, where essential modeling tools critically needed to inform manufacturing process development are lacking.

Predictive Understanding of Manufacturing Processes

Current status and recent advances

An important element of the transformation in manufacturing to be achieved through scientific research is accelerating the development of predictive understanding for the complex thermo-chemo-mechanical conditions that occur during the wide variety of manufacturing processes. This understanding will come from experimental hypothesis testing, physics-based computational models, and ML methods. The value of establishing such an information infrastructure has been recognized and is one of the central strategies of the AI and ML platforms.

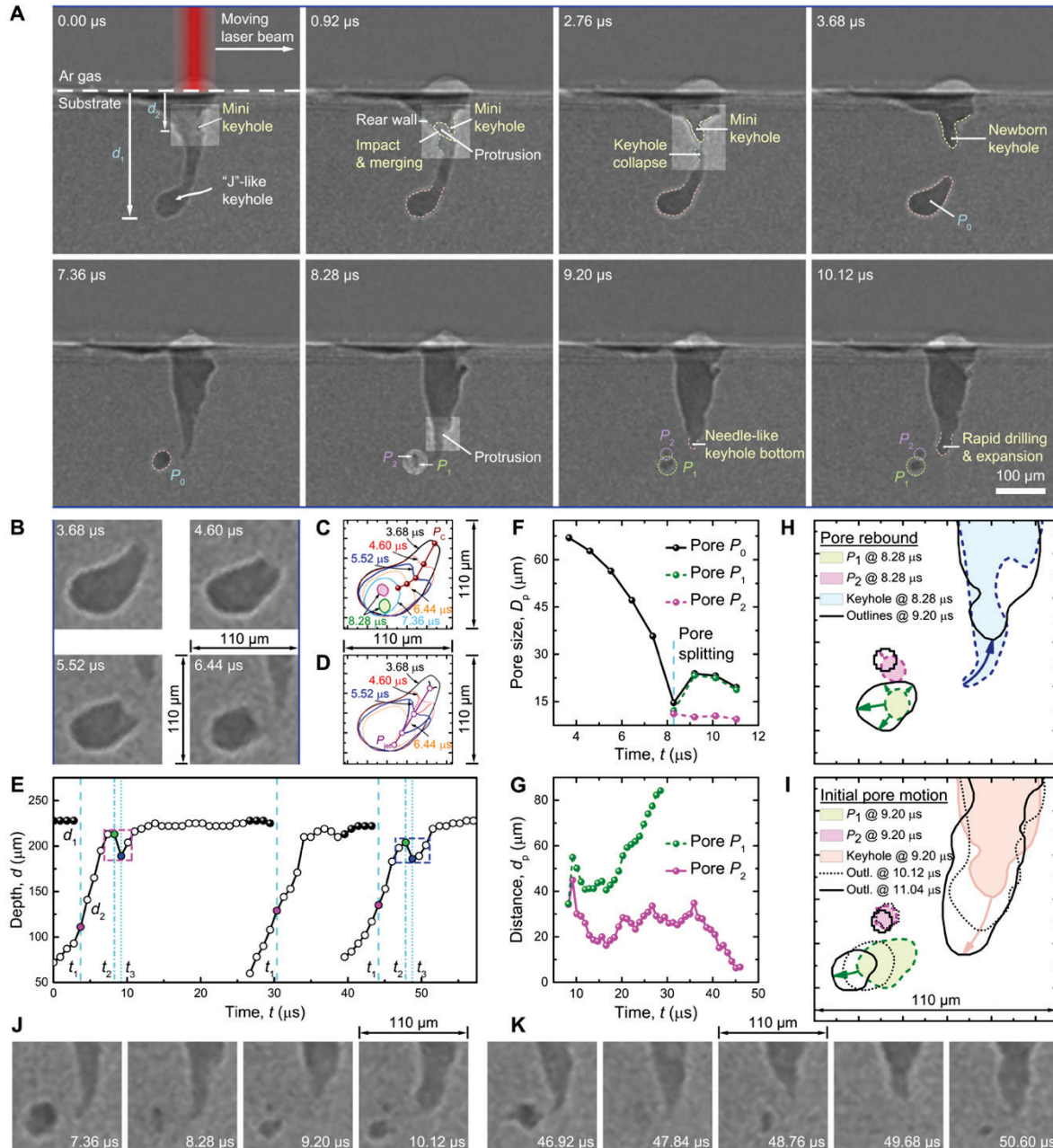


Figure 52. Acoustic wave-driven keyhole pore formation. (A) Megahertz x-ray images of a keyhole pore formation process. (B) X-ray images showing the non-uniform collapse of pore P_0 , driven by a microjet. (C and D) Contours of the pore P_0 and microjet morphologies. (E) Keyhole depths, d_1 and d_2 . Three time nodes are defined: at t_1 (magenta circle), with the keyhole closure, pore P_0 forms; at t_2 (green circle), the primary pore starts to rebound; at t_3 (blue circle), the keyhole depth reaches a local minimum, and the pores start to migrate away from the keyhole. (F) Equivalent pore diameter, D_p , estimated from (C). (G) Distances of pores from the nearest keyhole wall. (H) Formation of a needle-like keyhole bottom, attributed to the coupling between the existence of a protrusion and the rebound of pore P_1 . (I) Initial pore motions caused by the acoustic wave emitted from the needle-like keyhole bottom. (J and K) X-ray images of keyhole pore collapse, rebound, and motion, corresponding to the two abrupt decreases in depth highlighted by the magenta and blue dashed rectangles in (E), respectively. All images were background corrected followed by contrast reversal. Source: [Critical instability at moving keyhole tip generates porosity in laser melting](#), Z. Cang et al., *Science* 370 1080–1086 (2020).

Scientific challenges and opportunities

A comprehensive solution requires information in the form of high-quality experimental data and/or simulations derived from real or simulated manufacturing conditions. This may be realized through rapid, efficient, multiscale manufacturing simulation integrated with advanced 3D–4D+ imaging of microstructures and defects. Making such information widely available to the broader technical community will accelerate transformation in manufacturing by facilitating the reuse of this valuable information by others. There is an opportunity to establish programs to enhance the utilization of data such as outreach initiatives, establishing incentives for sharing results of publicly funded, fundamental research. It will take a combination of education, outreach, and incentivization to realize a transformational culture change in manufacturing. Recent examples of simulation challenges that made high-quality experimental data available for predictive modeling by the community include the NIST Additive Manufacturing Benchmark Test Series, <https://www.nist.gov/ambench>, and the Air Force Research Laboratory Additive Manufacturing Modeling Challenge series, <https://materials-data-facility.github.io/MID3AS-AM-Challenge>.

Manufactured Materials Data Repositories, Schema, Curation, and Sharing

Current status and recent advances

Data are the foundation of scientific progress, and the importance of data is only increasing in the era of high-throughput experiments, big data, and deep learning. New laboratory techniques and industrial processes, including AM, produce multimodal data in unprecedented quantities. Expanded computational resources offer the potential for these data to be stored, accessed, aggregated, and manipulated at scale. Advances in data science, including AI and ML, enable the extraction of useful knowledge from large, complex, and multi-dimensional data sets. Data repositories have been successfully established and populated for homogeneous fundamental data, such as thermodynamic phase equilibria determined by density functional theory (examples include the Materials Project, the Open Quantum Materials Database, and Nomad) and molecular dynamics simulation data (e.g., AFLOW, or Automatic Flow for Materials Discovery). However, these forms of information represent a very small slice of the information required for robust scientific understanding of manufacturing process effects on materials. This understanding will primarily require the ability to understand and predict multiscale microstructure evolution and defect formation, which ultimately control the response of manufactured materials.

Providing the required multi-modal, heterogeneous materials information, coupled with manufacturing process history information, to the broader scientific community is a significant infrastructural challenge. To do so, information repositories will be required to handle highly complex and interconnected data and metadata. There are relatively few examples of repositories for handling such heterogeneous materials information while also making this information available to the scientific community (e.g., the Materials Commons, Materials Data Facility, NIST Materials Data Repository).

Scientific Challenges and Opportunities

Materials science data are notably more diverse in type, size, and complexity than foundational data in some other disciplines, such as computational biology. This is particularly true for the type of heterogeneous, multi-modal, and multiscale data that are needed to understand manufacturing processes and their influences on material response. This diversity presents infrastructure challenges with respect to designing and supporting general-purpose data repository facilities and devising the associated schema that permit the data to be discovered and used. Similarly, assessing the quality of large, diverse data sets is a curation challenge that calls out for automated tools. Inducing the scientific community to make these data available to the broader scientific community remains a persistent challenge. That effort is limited by the lack of incentives/mandates for researchers to take on the tedious and unrewarding task of uploading data and sufficient metadata to make the information useful to the scientific community. Compared with

other fields, government investments in materials information repositories have been modest, compounding concerns about the long-term sustainability of these repositories. Finally, in some instances, data have been and remain intellectual property (IP), in some cases with clear monetization potential. Creating information technology structures, such as anonymization and partitioning, that permit data publication and sharing while safeguarding IP is a logistical challenge.

Despite these challenges, the ability to archive, find, assess, and use data is essential to maximizing the potential of data science, AI, and deep learning. Each of these challenge areas offers an opportunity to advance science in support of transformative manufacturing. Focused and sustained support for information repositories and incentives/mandates for data sharing are necessarily key components of any manufacturing science initiative. The development of methods to extract property and processing information from published work, which can be translated into properly curated data that can be operated on by ML, remains a substantial challenge. In all this, it is essential to combine domain expertise for materials and manufacturing with advanced information science and algorithm development.

Conclusion

Future manufacturing will likely require advanced materials synthesis and design underpinned by incisive characterization methodologies, data analytics, and AI, as well as digital design and simulation techniques. Although there are unique critical challenges in these areas, it is clear there are also crosscutting research themes which, when fully addressed, will advance many fundamental discoveries to higher levels of maturity. Development of embedded feedback between in situ and operando characterization and advanced computational tools will enable improved translation of early-stage discoveries to larger-scale developments (Figure 53). When such tools are combined with active/adaptive learning and other AI tools, rapid advancement of manufacturing science knowledge is possible.

As part of this approach, a new scientific paradigm that fundamentally and deliberately couples discovery of materials with processes to manufacture them at scale is needed to develop the needed predictive capabilities for processing. Such co-design of processing, materials, and product will require a range of new methodologies to understand and control defects, as well as a better overall understanding of the translation of fundamental synthesis parameters into larger-scale techniques. Promoting a more sustainable manufacturing footprint will create additional research needs via the co-optimization of product function with end-of-life considerations for deconstruction and recycling.

Underpinning these efforts is the need for the development and support of the enabling information infrastructure. The advancement of the supporting science will spawn the rapid generation of data that need to be managed for knowledge retention and technology advancement. It is critical to define who owns, curates, and supports the collection and maintenance of these data, and to determine how they will be accessible. An assessment of the data required for manufacturing processes and modeling, followed by repository standardization, curation, and management, will form the basis for translating data into the information needed for transformational manufacturing advancement.

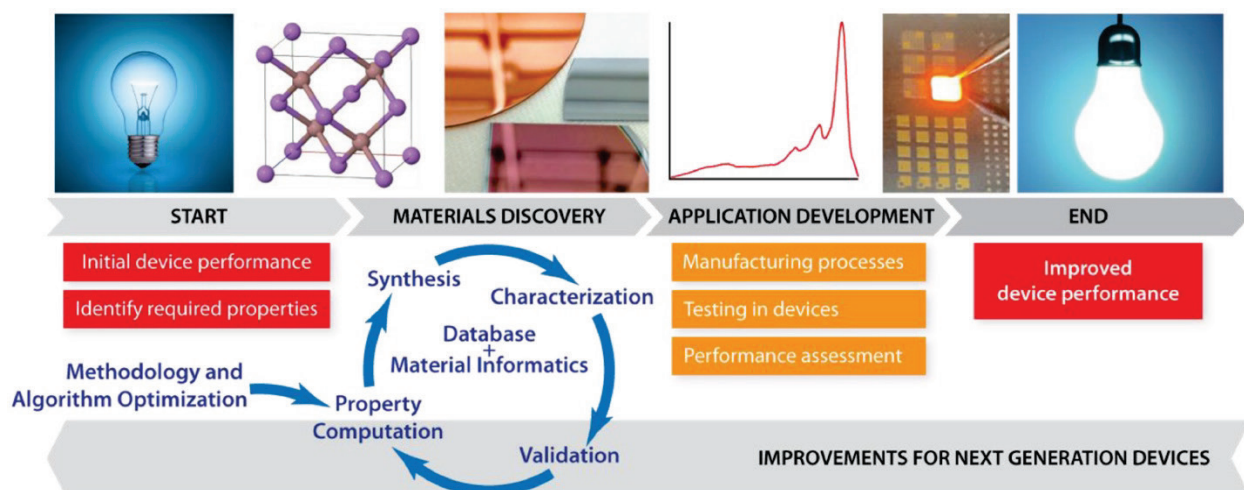


Figure 53. Schematic of the materials-by-design approach to accelerate materials transfer from computer simulations to lab bench and consumer products. Source: K. Alberi, M. B. Nardelli, A. Zakutayev, L. Mitas, S. Curtarolo, A. Jain, M. Fornari, N. Marzari, I. Takeuchi, M. L. Green, [The 2019 materials by design roadmap](#), *J. Phys. D: Appl. Phys.* 2019, 52, 013001. Used by permission under the [Creative Commons Attribution by 3.0 license \(CC by 3.0\)](#).

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3. Zhao, C.; Parab, N.D.; Li, X.; Fezzaa, K.; Tan, W.; et al. Critical Instability at Moving Keyhole Tip Generates Porosity in Laser Melting. *Science* **2020**, *370*, 1080–1086.

Appendix A. Workshop Participants

Basic Energy Sciences Basic Research Needs Workshop for Transformative Manufacturing

Chair: Cynthia Jenks, Argonne National Laboratory
Co-chairs: Ho Nyung Lee, Oak Ridge National Laboratory
Jennifer Lewis, Harvard University

Basic Energy Sciences Team: Linda Horton, Office of Basic Energy Sciences
Bruce Garrett, Chemical Sciences, Geosciences and Biosciences
Bonnie Gersten, Materials Sciences and Engineering
Michael Markowitz, Materials Sciences and Engineering
Katie Runkles, Basic Energy Sciences
Viviane Schwartz, Chemical Sciences, Geosciences and Biosciences

Panel Discussion:

Valri Lightner, Acting Director, Advanced Manufacturing Office (Moderator)
Leo Christodoulou, Boeing
Oleg Gang, Columbia University
William Grieco, AIChE
John Randall, Zyvek

Plenary Speakers:

Simon Bare, SLAC National Accelerator Laboratory
David Weitz, Harvard University
Christopher Schuh, Massachusetts Institute of Technology

Panel 1: Precision Synthesis Science

Chairs: Cherie Kagan, University of Pennsylvania
Paul Nealey, University of Chicago/Argonne National Laboratory

Charles Black, Brookhaven National Laboratory
Geoff Coates, Cornell
Jeffrey Elam, Argonne National Laboratory
Jennifer Hollingsworth, Los Alamos National Laboratory
Christine Luscombe, University of Washington
Jeff Neaton, Lawrence Berkeley National Laboratory
Chris Palmstrom, University of California–Santa Barbara
Jim De Yoreo, Pacific Northwest National Laboratory
Mike Arnold, University of Wisconsin

Panel 2: Processing and Scale Up Science

Chairs: Paul Braun, University of Illinois Urbana-Champaign
John Holladay, Pacific Northwest National Laboratory

Blair Carlson, General Motors
Michelle Kocal, LanzaTech
Krishan Luthra, GE (Retired)
Richard Vaia, Air Force Research Laboratory
David Weitz, Harvard University
Bhavik Bakshi, Ohio State University
Rajiv Mishra, University of North Texas
Magdalena, Ramirez-Corredores, Idaho National Laboratory
Rakesh Agrawal, Purdue University
Randy Cortright, National Renewable Energy Laboratory
Emma White, Ames Laboratory

Panel 3: System Integration Science

Chairs: Yan Gao, GE Research
David Sholl, Georgia Tech

Bill Grieco, RAPID
Tina Nenoff, Sandia National Laboratories
Michael Tsapatsis, Johns Hopkins
Yushan Yan, University of Delaware
Yet-Ming Chiang, Massachusetts Institute of Technology
Bill Huber, First Solar
Eric Lin, National Institute of Standards and Technology
Michael Glavicic, Rolls-Royce
Meagan Mauter, Stanford University
Karena Chapman University, Stony Brook

Panel 4: Sustainable Manufacturing

Chairs: Brett Helms, Lawrence Berkeley National Laboratory
John Sutherland, Purdue University

Dan Cooper, University of Michigan
Tim McIntyre, Oak Ridge National Laboratory
Karthish Manthiram, Massachusetts Institute of Technology
Bryce Meredig, Citrine
Rigoberto (Gobet) Advincula, Oak Ridge National Laboratory/University of Tennessee
Polly Arnold, Lawrence Berkeley National Laboratory

Panel 5: Digital Manufacturing

Chairs: Julia Greer, Caltech
Chris Spadaccini, Lawrence Livermore National Laboratory

Leo Christodoulou, Boeing
Jamie Guest, Johns Hopkins University
John Hart, Massachusetts Institute of Technology
Rob Steinhoff, Kansas City National Security Campus
Tony Schmitz, University of Tennessee
Dan Berrigan, Air Force Research Laboratory

Grace Gu, University of California–Berkeley
Alicia Kim, University of California–San Diego
David Shahan, Hughes Research Laboratory

Crosscutting Themes

Chairs: Elizabeth Holm, Carnegie Mellon University
Anthony Rollett, Carnegie Mellon University
Cathy Tway, Johnson Matthey

Simon Bare, SLAC National Accelerator Laboratory
John Allison, University of Michigan
Tao Sun, University of Virginia
Mitra Taheri, Johns Hopkins University
Juan De Pablo, University of Chicago/Argonne National Laboratory
Andrew Payzant, Oak Ridge National Laboratory
Stefan Wild, Argonne National Laboratory

Invited Participants

Rigoberto Advincula, Oak Ridge National Laboratory/University of Tennessee
Rakesh Agrawal, Purdue University
John Allison, University of Michigan
Mike Arnold, University of Wisconsin
Polly Arnold, Lawrence Berkeley National Laboratory
Bhavik Bakshi, Ohio State University
Simon Bare, SLAC National Accelerator Laboratory
John Berrigan, Air Force Research Laboratory
Charles Black, Brookhaven National Laboratory
Paul Braun, University of Illinois Urbana-Champaign
Blair Carlson, General Motors
Karena Chapman, Stony Brook University
Yet-Ming Chiang, Massachusetts Institute of Technology
Leo Christodoulou, Boeing
Geoff Coates, Cornell University
Dan Cooper, University of Michigan
Randy Cortright, National Renewable Energy Laboratory
Juan de Pablo, University of Chicago/Argonne National Laboratory
Jim De Yoreo, Pacific Northwest National Laboratory
Jeffrey Elam, Argonne National Laboratory
Oleg Gang, Columbia University/Brookhaven National Laboratory
Yan Gao, GE Research
Michael Glavicic, Rolls-Royce
Julia Greer, California Institute of Technology
William Grieco, AIChE
Grace Gu, University of California–Berkeley
Jamie Guest, Johns Hopkins University
John Hart, Massachusetts Institute of Technology
Brett Helms, Lawrence Berkeley National Laboratory
John Holladay, Pacific Northwest National Laboratory

Jennifer Hollingsworth, Los Alamos National Laboratory
Elizabeth Holm, Carnegie Mellon University
William Huber, First Solar
Cynthia Jenks, Argonne National Laboratory
Cherie Kagan, University of Pennsylvania
Alicia Kim, University of California–San Diego
Michelle Kocal, LanzaTech
Ho Nyung Lee, Oak Ridge National Laboratory
Jennifer Lewis, Harvard University
Valri Lightner, DOE Advanced Manufacturing Office
Christine Luscombe, University of Washington
Krishan Luthra, GE (Retired)
Karthish Manthiram, Massachusetts Institute of Technology
Meagan Mauter, Stanford University
Tim McIntyre, Oak Ridge National Laboratory
Bryce Meredig, Citrine
Rajiv Mishra, University of North Texas
Paul Nealey, University of Chicago/Argonne National Laboratory
Jeff Neaton, Lawrence Berkeley National Laboratory
Tina Nenoff, Sandia National Laboratories
Chris Palmstrom, University of California–Santa Barbara
Andrew Payzant, Oak Ridge National Laboratory
Magdalena Ramirez-Corredores, Idaho National Laboratory
John Randall, Zyvek
Anthony Rollett, Carnegie Mellon University
Tony Schmitz, University of Tennessee
Christopher Schuh, Massachusetts Institute of Technology
David Shahan, Hughes Research Lab
David Sholl, Georgia Tech
Chris Spadaccini, Lawrence Livermore National Laboratory
Rob Steinhoff, Kansas City National Security Campus
Tao Sun, University of Virginia
John Sutherland, Purdue University
Mitra Taheri, John Hopkins University
Michael Tsapatsis, Johns Hopkins University
Cathy Tway, Johnson Matthey
Richard Vaia, Air Force Research Laboratory
David Weitz, Harvard University
Emma White, Ames Laboratory
Stefan Wild, Argonne National Laboratory
Yushan Yan, University of Delaware

Invited Observers

Jim Belak, Lawrence Livermore National Laboratory
Carol Bessel, National Science Foundation
John Bobbitt, Savannah River National Laboratory
Chris Bradley, DOE Office of Science, Basic Energy Sciences
Alice Caponiti, DOE Nuclear Energy

Mary Case, Idaho National Laboratory
Santanu Chaudhuri, Argonne National Laboratory
Kerry Cheung, DOE Office of Electricity
Stan Chou, Sandia National Laboratories
Julie Christodoulou, Office of Naval Research
Daniel Clark, DOE Fusion Energy Sciences
Eric Colby, DOE High Energy Physics
Alessandra Colli, Brookhaven National Laboratory
Panos Datskos, National Renewable Energy Laboratory
Steve Eglash, SLAC National Accelerator Laboratory
Chris Fecko, DOE Office of Science, Basic Energy Sciences
Greg Fiechtner, DOE Office of Science, Basic Energy Sciences
Alex Fitzsimmons, DOE Office of Energy Efficiency and Renewable Energy
Dillon Fong, Argonne National Laboratory
Bruce Garrett, DOE Office of Science, Basic Energy Sciences
Bonnie Gersten, DOE Office of Science, Basic Energy Sciences
Matthias Graf, DOE Office of Science, Basic Energy Sciences
Justine Hauptman, Brookhaven National Laboratory
Robin Hayes, DOE Office of Science, Basic Energy Sciences
Craig Henderson, DOE Office of Science, Basic Energy Sciences
Linda Horton, DOE Office of Science, Basic Energy Sciences
Jim Horwitz, DOE Office of Science, Basic Energy Sciences
Tina Kaarsberg, DOE Advanced Manufacturing Office
Bryan Kaehr, Sandia National Laboratories
Robert Kostecky, Lawrence Berkeley National Laboratory
Matt Kramer, Ames Laboratory
Jeff Krause, DOE Office of Science, Basic Energy Sciences
Harriet Kung, DOE Office of Science, Basic Energy Sciences
Randall Lavolette, DOE Office of Advanced Scientific Computing Research
Eliane Lessner, DOE Office of Science, Basic Energy Sciences
John Litynski, DOE Fusion Energy Sciences
Debasis Majumdar, National Science Foundation
George Maracas, DOE Office of Science, Basic Energy Sciences
Alexandra Marchi, Los Alamos National Laboratory
Michael Markowitz, DOE Office of Science, Basic Energy Sciences
Patrick McGrath, DOE Advanced Research Projects Agency–Energy
Gail McLean, DOE Office of Science, Basic Energy Sciences
Scott McWhorter, Savannah River National Laboratory
Natalia Melcer, DOE Office of Science, Basic Energy Sciences
Raul Miranda, DOE Office of Science, Basic Energy Sciences
Mike Molner, National Institute of Standards and Technology
Karl Mueller, Pacific Northwest National Laboratory
Johanna Nelson-Weker, SLAC National Accelerator Laboratory
Garrett Nilsen, DOE Office of Energy Efficiency and Renewable Energy
Robert O'Brien, Idaho National Laboratory
Elliot Padgett, DOE Office of Energy Efficiency and Renewable Energy
Michael Pechan, DOE Office of Science, Basic Energy Sciences
Andre Pereira, DOE Office of Electricity

Kent Peters, DOE Office of Science, Biological and Environmental Research
Cindy Powell, Pacific Northwest National Laboratory
Sudarsan Rachuri, DOE Advanced Manufacturing Office
Tom Russell, DOE Office of Science, Basic Energy Sciences
Viviane Schwartz, DOE Office of Science, Basic Energy Sciences
Andy Schwartz, DOE Office of Science, Basic Energy Sciences
Michael Sennett, DOE Office of Science, Basic Energy Sciences
Igor Slowing, Ames Laboratory
Robert Stone, National Science Foundation
Xin Sun, Oak Ridge National Laboratory
Dave Teter, Los Alamos National Laboratory
Thiyaga Thiyagarajan, DOE Office of Science, Basic Energy Sciences
Michael Ulsh, National Renewable Energy Laboratory
Brian Valentine, DOE Advanced Manufacturing Office
Tony van Buuren, Lawrence Livermore National Laboratory
John Vetrano, DOE Office of Science, Basic Energy Sciences
Brenda Wyatt, Oak Ridge National Laboratory
Jane Zhu, DOE Office of Science, Basic Energy Sciences

Appendix B. Workshop Agenda

Basic Energy Sciences Basic Research Needs Workshop for Transformative Manufacturing

Hilton Washington DC

Rockville Hotel & Executive Meeting Center | March 9—11, 2020

Chair: Cynthia Jenks, Argonne National Laboratory

Co-chairs: Ho Nyung Lee, Oak Ridge National Laboratory
Jennifer Lewis, Harvard University

Monday, March 9, 2020

7:00 - 8:00 AM Registration/Breakfast

Opening Plenary Session

8:00 - 8:15 AM **Welcome and Workshop Charge**
Linda Horton, Basic Energy Sciences

8:15 - 8:30 AM **Chair Welcome and Workshop Structure**
Cynthia Jenks, Argonne National Laboratory

8:30 – 8:45 AM **Introduction to Plenaries**
Jennifer Lewis, Harvard University

8:45 – 10:00AM **Panel Discussion: AMO and Industrial Perspectives on Manufacturing Challenges**
Valri Lightner, Acting Director, Advanced Manufacturing Office (Moderator)
Leo Christodoulou, Boeing
Oleg Gang, Columbia University
William Grieco, AIChE
John Randall, Zyvek

10:00 – 10:30 AM **Break**

10:30 – 11:00 AM **Transformative Manufacturing and the DOE Office of Science, Basic Energy Sciences User Facilities—An Opportunity for Progress**
Simon Bare, SLAC National Accelerator Laboratory

11:00 - 11:30 AM **Manufacturing with Micron-Scale Devices: Scale-up in the Real World**
David Weitz, Harvard University

11:30 – Noon **Every Atom in the Right Place: Towards Mastery of Processing-Structure in Metals**
Christopher Schuh, Massachusetts Institute of Technology

Noon – 12:15 PM **Panel Introductions and Virtual Access for Panel Sessions**
Ho Nyung Lee, Oak Ridge National Laboratory
Katie Runkles, Basic Energy Sciences

12:15 - 1:00 PM **Working Lunch in Panel Breakout Rooms**

1:00 - 5:30 PM Parallel Panel Sessions

Panel 1: Precision Synthesis Science

Cherie Kagan, University of Pennsylvania
Paul Nealey, University of Chicago/Argonne National Laboratory

Panel 2: Processing and Scale Up Science

Paul Braun, University of Illinois Urbana-Champaign
John Holladay, Pacific Northwest National Laboratory

Panel 3: System Integration Science

Yan Gao, GE Research
David Sholl, Georgia Tech

Panel 4: Sustainable Manufacturing

Brett Helms, Lawrence Berkeley National Laboratory
John Sutherland, Purdue University

Panel 5: Digital Manufacturing

Julia Greer, Caltech
Chris Spadaccini, Lawrence Livermore National Laboratory

Crosscutting Themes

Elizabeth Holm, Carnegie Mellon University
Anthony Rollett, Carnegie Mellon University
Cathy Tway, Johnson Matthey

3:00 - 4:00 PM Refreshments available

5:30 – 7:00 PM Break for dinner (on own)

7:00 – 10:00 PM Parallel panel discussions (continued); Crosscutting Theme group meets in plenary session room

Tuesday, March 9, 2020

7:00 - 8:00 AM Breakfast

8:00 - 10:00 AM Parallel panel sessions for discussion/preparation of preliminary report
(Note: Panel reports MUST be on SharePoint)

10:00 - 10:30 AM Break

10:30 – 10:45 AM Report from Panel 1 **Precision Synthesis Science**

10:45 - 11:00 AM Report from Panel 2 **Processing and Scale Up Science**

11:00 - 11:15 AM Report from Panel 3 **System Integration Science**

11:15 – 11:30 AM Report from Panel 4 **Sustainable Manufacturing**

11:30 – 11:45 AM Report from Panel 5 **Digital Manufacturing**

11:45 – Noon **Report from Crosscutting Themes**

12:00 – 1:45 PM Working lunch (workshop chairs, panel leads, and DOE meet to
consolidate PRDs into a subset for afternoon discussion)

1:45 – 2:00 PM Reconvene in **Plenary Session—PRD Breakout Descriptions**

2:00 - 5:30 PM Reconfigured groups focus on PRD discussions
(Note: PRD 1 will be in Panel 1's room, etc.)

3:00 - 4:00 PM Refreshments available

5:30 -7:00 PM Break for dinner

7:00 – 10:00PM PRD discussions (continued) and Preparation for final PRD reports

Wednesday, March 10, 2020

7:00 - 8:00 AM Breakfast

8:00 – 9:45 AM **PRD Discussions** continue

9:45 – 10:00 AM Break

10:00 – 10:20 AM Report on PRD 1 **Achieve precision scalable synthesis and
processing of atomic-scale building blocks for
components and systems**

10:20 – 10:40 AM Report on PRD 2 **Integrate multiscale models and tools to enable
adaptive control of manufacturing processes**

10:40 – 11:00 AM Report on PRD 3 **Unravel the fundamentals of manufacturing
processes through innovations in operando
characterization**

11:00 – 11:20 AM Report on PRD 4 **Direct atom and energy flow to realize sustainable manufacturing**

11:20 – 11:40 AM Report on PRD 5 **Co-design materials, processes, and products to revolutionize manufacturing**

11:40 – 12:00 noon Discussion and closing remarks

12:00 noon Workshop adjourned

Noon – 5:00 PM Working lunch/writing (only chairs, panel leads and designated writers)

5:30 PM Adjourn

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