Understanding & Controlling Accelerated and Gradual Evolution of Materials for Energy (UNCAGE-ME)
EFRC Director: Ryan P. Lively
Lead Institution: Georgia Institute of Technology
Class: 2022 – 2026

***Mission Statement**: To develop a deep knowledge base in the characterization, prediction, and control of materials evolution in the presence of realistic contaminants, processes, and mixtures to accelerate materials discovery for sustainable production and utilization of H_2 and CO_2*

The overall objective of UNCAGE-ME from 2014-2022 has been to develop fundamental structure-property relations describing how acid gases interact with and induce evolution of adsorbents and catalysts. This was achieved via an interdisciplinary effort that combined novel synthesis, advanced in situ/operando characterization, machine learning techniques and molecular modeling. The creation of structure-property relationships accelerated materials discovery for acid gas separations, conversion, and utilization via these integrated design tools. The research accomplishments of the Center over this 8-year period provided detailed descriptions of the impact of acid gas exposure on metal-oxides, metal-organic frameworks, carbons, supported amines, porous organic cages, and other materials.

In Phase III, we will leverage our Center model and learnings from our acid gas campaigns to accelerate progress in new directions. In line with our Phase III Mission Statement our new focus is on the evolution of materials relevant to clean energy technologies when exposed to realistic contaminants, processes, and complex mixtures. This will be achieved by combining new syntheses, in situ/operando characterization, molecular modeling, and machine learning approaches applied to catalyst, sorbent, and membrane use in clean energy technologies such as H_2 generation and CO_2 capture and conversion.

We will leverage our learnings from Phase I-II to address fundamental aspects of novel materials synthesis, materials stability, and structural evolution that underpin a suite of clean energy technologies relevant to H_2 and CO_2 processing. These include: (i) generation of H_2 via polyelectrolyte membranes (PEMs) and solid oxide electrolysis cells (SOECs); (ii) direct air capture of CO_2; and (iii) electrochemical reduction of CO_2. The name and mission of UNCAGE-ME have been updated to reflect these new focus areas and serve to motivate the Center's research portfolio. These activities are driven by a set of four-year research goals:

1) Elucidate the overarching relationships for process-induced structure and property evolution of functional materials with a focus on separations media and (electro)catalysts.
2) Leverage and advance computational and machine learning techniques to enable fundamental molecular and electronic level predictions of materials interacting with complex mixtures of targeted gases and contaminants.
3) Demonstrate accelerated materials discovery for clean energy technologies via process-materials coupled research.

Materials-focused research for separations and catalysis often considers the materials as being passive in their process environment. In contrast, evolution of materials under working conditions due to exposure to contaminants and non-steady-state conditions under realistic conditions is the norm rather than an exception. In Phase III, we will synthesize novel forms of matter with tailored functional properties and combine in situ/operando molecular spectroscopic studies of both the surface functionalities and bulk structures of materials relevant to the catalytic formation and separations of CO_2 and H_2 under conditions relevant to complex environments. Our experimental findings will be fused with complementary data analytics and multi-scale computational and theoretical modeling of emerging contaminant interactions with functional solids for targeted materials design.

An overview of the various research thrusts and cross-cutting tracks for Phase III is shown in **Figure 1**. Teams comprised of members from each Thrust will address basic scientific questions associated with the technologies described in each track. We have built teams with a broad range of skillsets comprised of senior personnel from each of the three **Research Thrusts** and these teams will be challenged to address fundamental scientific questions related to two **Clean Energy Technology Tracks** (CO_2 Capture & Conversion and H_2 Conversion).

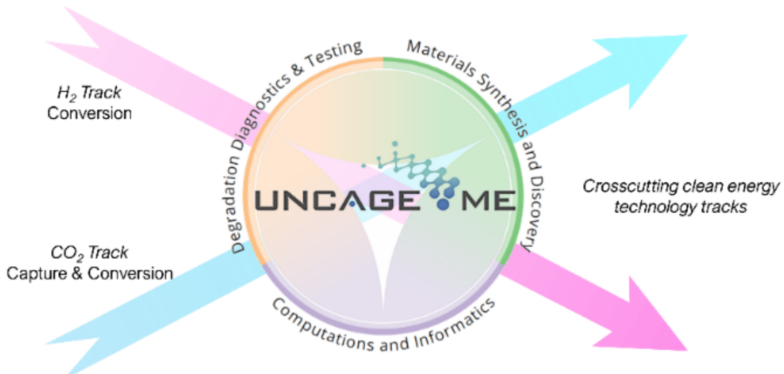

Figure 1: Schematic overview depicting the application of UNCAGE-ME research thrusts (circular emblem) to cross-cutting clean energy technology tracks (arrows). Teams of researchers from each thrust will address specific scientific questions associated with each track.

The Research Thrusts comprise groupings of senior personnel based on expertise. The three thrusts are: (i) Materials Synthesis & Discovery; (ii) Degradation, Diagnostics, & Testing; and (iii) Computation & Informatics. The Materials Thrust focuses on designing, synthesizing, and testing model and new functional materials with tailored properties for clean energy technologies in realistic process streams relevant to H_2 generation, separation, and conversion as well as CO_2 removal from the air and conversion. The Degradation Thrust will focus on advanced in situ/operando structural and performance analysis of model and new materials for H_2/CO_2 processing under relevant conditions. We will leverage existing infrastructure that was purpose-built for UNCAGE-ME Phases I-II as well as some new methods (e.g., Modulation Excitation Spectroscopy (MES), Near Atmospheric Pressure (NAP)-XPS, etc.) for some of the measurements conducted in this thrust. The Informatics Thrust will use advanced computational and data-driven approaches coupled with the experimental molecular-level insights from the other two Thrusts to determine underlying causes of material evolution, predicting this evolution under process-relevant conditions, and ultimately accelerating materials discovery and advancement into technology.

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