Center for Closing the Carbon Cycle (4C)
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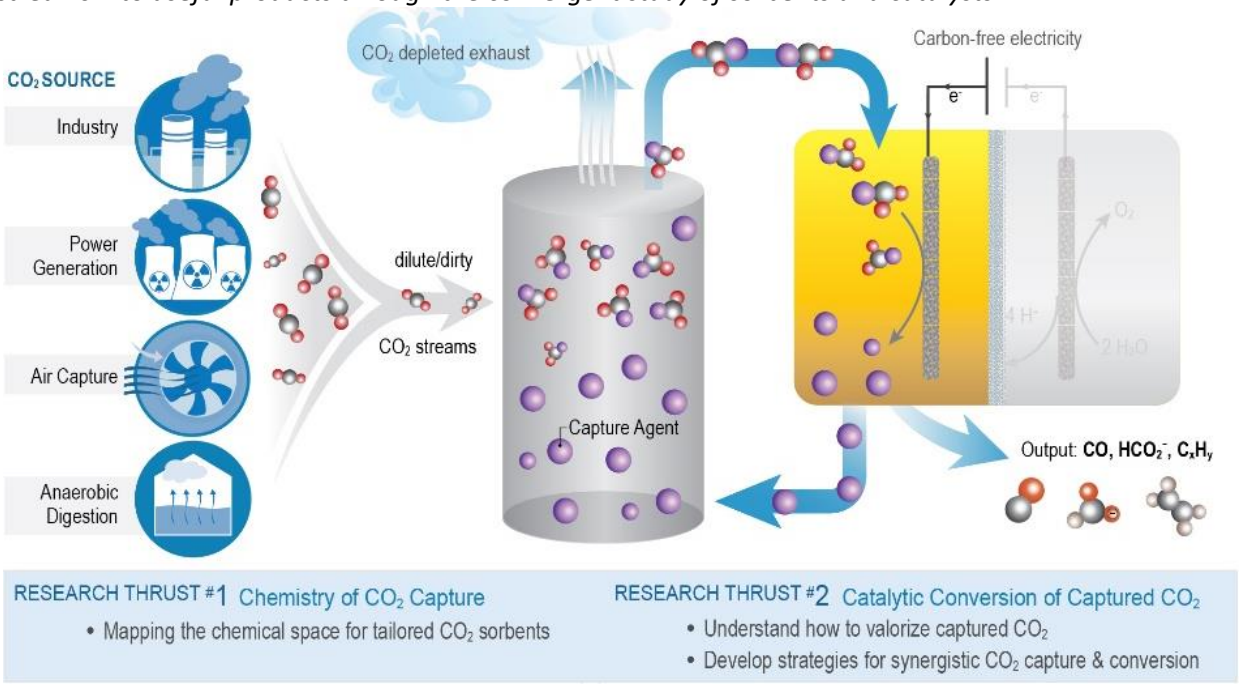
Lead Institution: University of California, Irvine
Class: 2022 – 2026

Mission Statement*: To advance synergistic capture and conversion of carbon dioxide (CO_2) from dilute streams into useful products through the convergent study of sorbents and catalysts.*



The **Center for Closing the Carbon Cycle (4C)** will advance the foundational science and define key integration parameters for synergistic CO_2 capture and conversion, or reactive capture of CO_2 (RCC). While there has been significant independent research in either CO_2 capture or pure CO_2 conversion, it is not well understood what knowledge is translatable to RCC. 4C combines expertise on capture and valorization in center-wide collaborations what will enable co-design of CO_2 sorbents with catalysts for conversion. 4C will establish guidelines for CO_2 capture from various dilute and dirty streams and define how captured CO_2 can most effectively be utilized, leading to selective, durable, and efficient pathways from CO_2 source-to-product.

Research Thrust #1 (RT #1) is focused on the chemistry of CO_2 capture. Libraries of CO_2 sorbents will be established using computational screening in parallel with high-throughput experimentation. These studies will establish structure-electronic relationships for different classes of soluble sorbents and functionalized solvents and their stability towards oxygen, water, and other common contaminants. The effect of electrolyte and other additives on sorbent properties will be quantified, and their microstructures will be spectroscopically and computationally interrogated. Neutron scattering will provide a link between macroscopic CO_2 binding and atomistic structure, and picosecond dynamic simulations will provide crucial insights to speciation and structural reorganization at atomic length scales. This information be used to develop improved models for CO_2 binding, solvation energies, and microenvironment effects on sorbent molecules and materials.

The research will advance our understanding of CO_2 sorption chemistry and expand the library of CO_2 sorbents. The knowledge base will valuable for understanding how to capture CO_2 from air or other point sources, which is increasingly important for carbon neutral and net negative technology outside of RCC.

Research Thrust #2 (RT #2) will focus on electrocatalytic valorization of captured CO_2. Our understanding of mechanisms and guiding concepts in electrochemical CO_2 Reduction (CO2R) has grown to include canonical reaction paths and thermodynamic descriptors for reactivity and selectivity. The discovery of new catalysts and electrolyte interactions for CO2R have illuminated effects of specific ions, dielectric constant, and pH on activation barriers, reaction selectivity, and product selectivity. 4C aims to understand how these principles translate to RCC, as captured CO_2 adducts are an intrinsically different substrate for catalysis with a broad range of molecular diversity, as described in RT #1. RCC allows tuning the physical and reactivity properties of the substrate, captured CO_2, to access a broader scope of chemical transformations compared to traditional CO_2 chemistry. Moreover, the dynamics between CO_2, the capture agent, supporting electrolyte, and solvent create new catalyst considerations.

4C will build on our understanding of electrochemical CO2R by establishing the concepts that underpin the reactivity of captured CO_2. By bridging knowledge between capture and conversion, 4C will enable the co-design efficient and selective homogeneous, heterogeneous, and hybrid catalysts for RCC to C-based commodities and fuels. A key theme is understanding the interface between substrate, solvent, and electrolyte with catalyst active sites, which is necessary to improve models and develop new theories regarding the design of catalyst microenvironments. These principles are broadly applicable, and will be transformative in how the community considers the role of interfaces and chemical microenvironments.

Summary. By studying CO_2 capture (RT #1) and conversion (RT #2) together, 4C will address the fundamental challenges associated with integration, including identifying and matching the relevant kinetics at multiple time scales to enable continuous operation. In addition to improving the overall efficiency and thus lowering the cost of CO_2-derived products, cooperative research between capture and conversion will lead to systems that more economically valorize CO_2 from dilute and dirty streams.

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