

Understanding & Controlling Accelerated and Gradual Evolution of Materials for Energy (UNCAGE-ME)
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Lead Institution: Georgia Institute of Technology
Class: 2014 – 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 H₂ and CO₂.*

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 objective 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 are leveraging 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 focus will be achieved by combining new syntheses, *in situ/operando* characterization, molecular modeling, and machine learning approaches applied to catalysts, sorbents, and membranes use in clean energy technologies such as H₂ generation and CO₂ capture and conversion.

We are leveraging 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 H₂ and CO₂ processing. These include: (i) direct air capture of CO₂; (ii) green generation of H₂ via water splitting / electrolysis; and (iii) electrochemical conversion of clean feedstocks (such as H₂ and CO₂) into useful products. 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) Synthesize a series of new adsorbents, (electro)catalysts, or membranes that meet or exceed established benchmarks in terms of cost, performance, and stability.
- 2) Create a library of the impact of emerging contaminants on adsorbent, (electro)catalyst, and membrane structure and activity.
- 3) Develop computational models to guide the rational design of new materials and predict their stability in realistic contaminant environments.

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 CO₂ and H₂ under conditions relevant to complex environments. Our experimental findings will be fused with complementary data

analytics and multi-scale modeling of emerging contaminant interactions with functional solids for targeted materials design. An overview of the various research tracks for Phase III is shown in Figure 1.

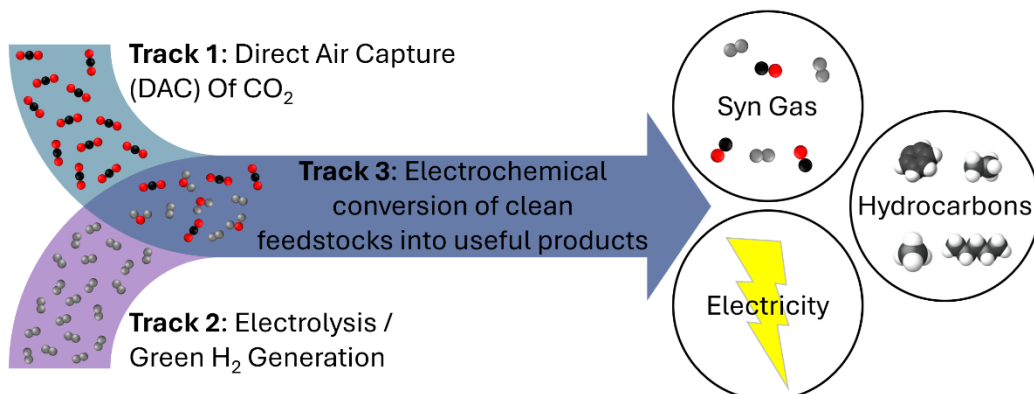


Figure 1. Schematic overview depicting the interrelated clean energy technology Tracks which are the focus of UNCAGE-ME.

Teams comprised of senior personnel are grouped into Research Thrusts based on expertise, and members from each Thrust will address basic scientific questions associated with the technologies described in each of the three Tracks. The three thrusts are: (i) New Materials Synthesis; (ii) Realistic Contaminant Effects; and (iii) Computation & Informatics. The New Materials Synthesis Thrust focuses on designing, synthesizing, and testing new functional materials with tailored properties for H₂ generation and CO₂ capture, as well as their electrochemical conversion into other useful products. The Realistic Contaminant Effects Thrust will focus on advanced *in situ/operando* structural and performance analysis of the new materials for H₂/CO₂ processing in realistic process streams. 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 Computation & 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 new materials discovery.

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