Understanding Degradation Mechanisms of Aminopolymers Used in Direct Air Capture

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PI: Simon H. Pang
Lawrence Livermore National Laboratory

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Program Overview

- **Overall Project Objective**
  - Identify fundamental mechanisms and elucidate key chemical and structural parameters that impact degradation of aminopolymer-based DAC adsorbent materials

- **Timeline and Budget**
  - Project start date: 10/01/20
  - Project end date: 09/30/23
  - DOE-BES funding: $4.5M
**Technology Background**

Oxidative degradation of PEI causes **irreversible CO₂ capacity loss**: there is a **critical gap in the fundamental science** about the degradation mechanism and strategies that improve stability are not well understood.

**Impact**: Determining key degradation mechanisms can promote **novel, rationally motivated improvement strategies** for polymer modification, and better predictions of adsorbent lifetime will **reduce uncertainty** about direct air capture cost and economic viability.
Our approach uses a highly integrated combination of spectroscopic characterization, kinetic analysis, and advanced quantum simulations to generate fundamental knowledge about the mechanisms and kinetics of aminopolymer-based DAC adsorbent degradation.

A fully successful project will (i) determine key reaction mechanisms for oxidative degradation and (ii) develop models that allow better prediction of short- and long-term degradation behavior.
Progress and Current Status of Project

Model material studies

- Multi-analytical spectroscopic approach (NMR, FTIR, mass spectrometry) to investigate oxidation product distribution and formation kinetics
- Developing methods to sample the reaction space of $O_2$ interaction with model compounds; compute reaction free energies, activation barriers, and NMR spectra

Polymer mobility in model supports

- Fluorescence measurements of PEI under pore confinement indicate dramatic change in mobility (right)
- Examining polymer-support interactions with SSNMR to identify chemical environments for degraded materials
Opportunities for Collaboration

- Integration of project components
  - Synthesis and characterization with advanced quantum simulation and multiscale modeling
  - Characterization methods across team members coupled to simulation of spectroscopic fingerprints
  - Kinetic analysis connecting short-term accelerated behavior with long-term consequences

- Collaboration with industry provides context and model validation
  - Global Thermostat a leading practitioner of amine-based DAC
  - Project will develop models for predicting adsorbent lifetime, to be validated against quasi-accelerated aging experiments

- Looking forward...
  - Development and scale-up of next generation adsorbent materials and other degradation mitigation strategies