Low Temperature Sorption-Enhanced Hydrogen Production from Natural Gas using Variable Volume, Batch-Membrane Reactors

David Anderson
Research Advisor: Professor Andrei G. Fedorov
Multiscale Integrated Thermofluidics Laboratory

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Although abundant petroleum and coal have enabled the production of affordable primary power and transportation fuels over the past century, the recent emergence of natural gas as a significant fossil fuel resource has dramatically changed the energy landscape. With its favorable hydrogen-to-carbon ratio coupled with recently developed techniques to tap vast reserves that were previously not economically viable, natural gas has been touted by many as a bridge fuel to a low-carbon energy future. Hydrogen production from natural gas feedstock via the steam-methane reforming (SMR) process is a mature industrial technology; yet miniaturizing this process for distributed production has proven problematic. Owing to the strongly endothermic nature of the process, reaction temperatures in excess of 900°C are traditionally employed in large-scale SMR reactors to overcome the thermodynamic limitations of the process. For a distributed reactor system, a maximum operating temperature below 500°C is desired to improve safety and reduce material cost and preheating energy requirements.

Combined reaction-separation approaches are a promising means to overcome the SMR process scale-down limitations. To this end, this PhD research focuses on the advancement of the CO₂/H₂ Active Membrane Piston (CHAMP) variable volume, batch-membrane reactor concept to incorporate selective CO₂ adsorption and enable distributed H₂ production from natural gas feedstock. The CHAMP reactor, operating in a cycle similar to the Internal Combustion (IC) engine, has proven capable of dynamically maintaining optimal reaction conditions for transport of reactants to the catalyst, reaction kinetics at the catalyst, and H₂ membrane separation. While previous CHAMP studies used methanol feedstock and only H₂ membrane separation, the proposed work focuses on developing the CHAMP reactor concept further by (1) incorporating CO₂ adsorption to complement the H₂ membrane separation, and (2) reforming methane as opposed to methanol feedstock. This enhanced reactor concept incorporates sorbent regeneration steps in its operating cycle, thereby preventing eventual saturation of the sorbent and enabling methane conversion enhancement for repeated cyclic operation.