TARGETING CARBON CAPTURE IN FUTURE POWER GENERATION SYSTEMS

The U.S. Department of Energy's vision for future clean coal power generation systems assumes the availability of cost effective techniques to separate carbon dioxide (CO₂) from mixtures of gases. Once captured or separated, the CO₂ might be used to achieve enhanced recovery of oil from underground oil reservoirs or it might be sequestered in an underground geologic formation or at the bottom of the ocean.

Carbon dioxide capture processes are needed for a range of power generation systems. For example, a cost effective CO₂ separation method would make it possible to produce relatively pure and low cost hydrogen for use in fuel cells and other applications, it would make it possible to gasify coal for use in a combined cycle power plant while capturing the CO₂ produced in the gasifier, and it would make it possible to burn coal in a boiler and capture the CO₂ from the flue gas.

Process and materials research being carried out by Drs. Hugo S. Caram and Shivaji Sircar of the Chemical Engineering Department of Lehigh University on a novel CO₂ separation technology shows promise for meeting these various needs. One of their processes is referred to as the "Thermal Swing Sorption Enhanced Reaction (TSSER)" concept.

Sircar explains, "One of the important applications of the TSSER concept involves a power generation system which contains an oxygen and steam fired coal gasifier. This type of gasifier converts coal into a mixture of carbon monoxide (CO), carbon dioxide (CO₂), water vapor (H₂O), and hydrogen (H₂) gases. The gas mixture from the gasifier would be fed into a TSSER system where the CO and H₂O would be catalytically converted to H₂ and CO₂ by a process referred to as the water gas shift reaction. The CO₂ would simultaneously be adsorbed from the reaction zone by a special chemisorbent material, resulting in a relatively pure product stream of CO₂-free H₂ gas. Removal of CO₂ from the reaction zone will drive the equilibrium controlled reaction to completion. This will result in extremely high conversion of CO and H₂O to H₂ in the sorber-reactor. The catalyst and the chemisorbent will be contained as an admixture in the same reactor vessel and the reaction temperature will be in the range of 300 – 400°C. Once the sorbent material has become saturated with CO₂, the chemisorbed CO₂ will be desorbed by counter-currently purging the reactor with superheated steam at ~500 – 550°C and at feed gas pressure. The effluent steam will then be condensed, resulting in a flow of mostly CO₂ at feed gas pressure, suitable for sequestration (after necessary compression). At the conclusion of the purge process, the gasifier effluent would be reintroduced into the reactor, and the cyclic process would be resumed. To permit continuous operation, the TSSER system would need to contain two reactors so that one is in the sorption mode
while the other one is undergoing regeneration."

Sircar adds, "There are many competing process concepts for separating CO₂ from mixtures with H₂. These include physical adsorption of CO₂ on a zeolite or an activated carbon and physicochemical absorption of CO₂ by a liquid solvent. A variety of Pressure Swing Adsorption (PSA) and Gas Absorption schemes are commercially used. While these methods can produce high purity H₂, many of these processes do not recover CO₂. PSA processes have also been designed to produce separate streams of high purity H₂ and CO₂, but the CO₂ product is produced at near atmospheric pressure. Physical adsorbents also require the H₂O to be removed from the feed gas prior to CO₂ adsorption, which is a disadvantage.

The process we've developed simultaneously converts CO and H₂O to CO₂ and H₂ and also separates the H₂ and CO₂ with high recovery of both products in a single unit operation. A novel chemisorbent, which selectively and reversibly adsorbs CO₂ at an elevated temperature of 300 - 400°C in presence of excess steam, is used for this purpose. We use the temperature swing mode instead of a pressure swing for sorbent regeneration. This can be highly beneficial because it avoids or substantially lowers the need to recompress the CO₂ stream, which results in energy and cost savings.

We are also developing several variations of the basic TSSER concept for other applications such as steam reforming of natural gas (CH₄) to directly produce fuel-cell grade H₂ and a CO₂ by-product. The reforming reaction, in this case, can be carried out at a substantially reduced temperature of ~ 400°C instead of the conventional ~850°C without sacrificing conversion of CH₄ to H₂ and this saves energy.

Another application of the novel chemisorbent will be to capture and recover low concentration CO₂ from a hot and wet flue gas without precooling and pre-drying the gas."

Caram continues, "With financial support from the Pennsylvania Infrastructure Technology Alliance (PITA) and a donation from the Air Products and Chemicals, Inc., we have constructed a single-column laboratory scale TSSER system to study the production of H₂ and CO₂ by steam reforming of natural gas. We are currently in the process of testing CO₂ chemisorbents, by measuring the CO₂ adsorption and desorption characteristics under various conditions by determining the sorption column dynamics for various steps of the TSSER concept and by determining the thermal stability of the chemisorbent. We are also simulating various steps of the cyclic process. Our project team includes Michael Beaver, a graduate student from Chemical Engineering, Jennifer Purcell and Jonathan McMullen, undergraduate students from Chemical Engineering, and Dr. Alex Verdooren, a Postdoctoral Researcher.

We've also received funding from DOE for a project scheduled to start up in January 2006, to investigate the application of this technology to generate H₂ and CO₂ streams from syngas produced by a coal-fired gasifier. That project will focus on the design and operation of a laboratory scale system to characterize the performance of a TSSER system for the gasifier application. It will also develop a mathematical process design model needed for scale up and optimization studies."
**RESEARCHERS’ PROFILES**

- **Dr. Hugo Caram** is a Professor of Chemical Engineering at Lehigh. His research is in the areas of mass transfer, chemical reactor design and chemical thermodynamics.

- **Dr. Shivaji Sircar** has a Ph.D. in Chemical Engineering, with expertise in separation processes and materials. He is presently a Professor of Practice at Lehigh University.

- **Dr. John DuPont** is an Associate Professor of Materials Science and Engineering at Lehigh. His research interests include welding metallurgy and processes.

- **Dr. David Williams**, Professor of Materials Science and Engineering and Vice Provost for Research at Lehigh, is a specialist in electron microscopy.

- **Mr. Ronald Mizia** is an Engineering Fellow at the Idaho National Engineering and Environmental Laboratory.

- **Dr. Charles Robino and Dr. Joseph Michael** are Distinguished Members of the Technical Staff at Sandia National Laboratories.