# New Adsorption Cycles for Carbon Dioxide Capture and Concentration

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**OBJECTIVES:**

Since a viable separations technology has yet to be identified for the cost-effective capture and concentration of $CO_2$ from coal gasification processes, and based on the very promising and extensive results obtained during the IC Phase I project, "Radically New Adsorption Cycles for Carbon Dioxide Sequestration," the objective of this three-year continuation project is to study the new pressure swing adsorption (PSA) cycles discovered for $CO_2$ capture and concentration at high temperature. The heavy reflux (HR) PSA concept and the use of a hydrotalcite like (HTlc) adsorbent that captures $CO_2$ reversibly at high temperatures simply by changing the pressure are two key features of these new PSA cycles. An expansive bench-scale experimental and theoretical program is proposed to complement and extend the extensive process simulation feasibility study that was carried out during Phase I. Because feasibility of the HR PSA-HTlc cycle concept was demonstrated only theoretically during Phase I, some key questions remain to be answered experimentally in Phase II to make this newly proposed high temperature cyclic adsorption process a viable, potentially economical, technology for recovering $CO_2$ from coal gasification processes. These questions, in the form of objectives, include determining 1) the type of HR PSA cycle configuration that should be used, 2) the definitive equilibrium and kinetic properties of the viable HTlc adsorbent, and 3) the economics of the resulting HR PSA-HTlc process for $CO_2$ capture and concentration at high temperature compared to other separations and capture technologies. In addition, ancillary objectives include developing and evaluating newly modified HTlc adsorbents for improved $CO_2$ cycle life capacity and kinetics, and determining the cycle life performance of viable HTlc adsorbents in the presence of $H_2O$ and/or $SO_2$. The major outcome of this three-year project will be the definitive analysis and viability of an HR PSA-HTlc process for $CO_2$ capture and concentration at high temperatures.

To carry out this ambitious and potentially definitive project on the use of high temperature PSA technology for $CO_2$ concentration and capture from coal gasification processes and possibly other stack and flue gas streams containing $CO_2$, a strong team comprised of academic (USC), government (EPA) and industrial (Air Products and Chemicals) experts on adsorption technology has been assembled. Three global tasks have been identified and will be carried out. They include single and multi-bed experimentation, PSA process simulator validation, and HTlc adsorbent equilibrium and kinetic property determination. These tasks are further broken down into nine sub-tasks that collectively define the three-year project in terms of personnel, goals and outcomes. One to two full-time PhD students will carry out the gist of this research with assistance from undergraduates, the PI, co-PI, and Drs. Hutson and Hufton from EPA and APCI, respectively.

**ACCOMPLISHMENTS TO DATE:**

During this period of performance (8 months), the stripping reflux PSA code was used to study various SR PSA cycle configurations with and without heavy and or light reflux for the high temperature PSA cycle utilizing a K-promoted HTlc to concentrate $CO_2$ at high temperature. Thousands of simulations have been carried out using mostly the SR PSA cycle concept. This work has been published recently in *Adsorption* and *I&ECR*. Additional manuscripts are either submitted (*Environmental Progress*) or in preparation. The overall conclusions from these works were that it is indeed possible to separate $CO_2$ from high temperature flue and stack gases at high purity, high recovery and reasonable throughputs using a K-promoted HTlc adsorbent and an SR PSA cycle with a heavy reflux step, but that mass transfer effects were also important. Also, through HTlc materials research and modeling it has been shown for the first time that the adsorption and desorption behavior of $CO_2$ on K-promoted HTlc is associated with a complex combination of completely reversible adsorption, diffusion and reaction phenomena.

**FUTURE WORK:**

This three-year grant began August 2005 as a continuation grant of a one-year exploratory grant that expired March 2004. The novel PSA cycle research is continuing to gain a better understanding of not only the mass transfer effects but also heat effects on the process performance. Also, the HTlc materials research and modeling is continuing to gain a mechanistic understanding and better estimation of the uptake and release rates of $CO_2$ in K-promoted HTlc.

**LIST OF JOURNAL ARTICLES PUBLISHED, IN PRESS OR UNDER REVIEW**

- S. P. Reynolds, A. D. Ebner and J. A. Ritter, New Pressure Swing Adsorption Cycles for Carbon Dioxide Sequestration," *Adsorption*, 11, 531-536 (2005).
- S. P. Reynolds, A. D. Ebner, and J. A. Ritter, "Stripping PSA Cycles for $CO_2$ Recovery from Flue Gas at High Temperature Using a Hydrotalcite-Like Adsorbent," *Ind. Eng. Chem. Res.*, in press on line (2006).
- A. D. Ebner, S. P. Reynolds and J. A. Ritter, "Understanding the Adsorption and Desorption Behavior of $CO_2$ on a K-Promoted HTlc through Non-Equilibrium Dynamic Isotherms," *Ind. Eng. Chem. Res.*, submitted (2006).
- A. D. Ebner, S. P. Reynolds and J. A. Ritter, "Non-Equilibrium Kinetic Model for the Reversible Adsorption of $CO_2$ on a K-Promoted HTlc," *Chem. Eng. Sci.*, submitted (2006).
- S. P. Reynolds, A. D. Ebner, and J. A. Ritter, Capture of $CO_2$ from Flue Gas by PSA using K-Promoted HTlc: Mass transfer Effects," Environmental Progress, submitted (2006).

**LIST OF PRESENTATIONS:**

- S. P. Reynolds, A. D. Ebner, and J. A. Ritter, "Concentration and Recovery of Carbon Dioxide at High Temperature with Heavy Reflux PSA Cycles," AIChE 2005 Annual Meeting, Cincinnati, OH, November 2005, contributed.
- S. P. Reynolds, A. D. Ebner, and J. A. Ritter, "Dynamic Adsorption and Desorption of $CO_2$ in K-Promoted Hydrotalcite," AIChE 2005 Annual Meeting, Cincinnati, OH, November 2005, contributed.

**STUDENTS SUPPORTED UNDER THIS GRANT**

- Steven P. Reynolds, PhD candidate, Department of Chemical Engineering, University of South Carolina. Steven, while being supported through an NSF K-12 Graduate Fellowship, this grant, and a MeadWestvaco Fellowship, worked on PSA code development, in particular on the SR and ER PSA codes and also on the design of a high temperature SR, ER or DR universal PSA system to be built in the near future with this funding. He carried out the simulations and corresponding analyses completed to date, and is continuing on this project to completion.
- Hai Du, PhD candidate, Department of Chemical Engineering, University of South Carolina. Hai, as a new PhD student in the group and while being supported through this grant and a MeadWestvaco Fellowship, recently began working on the synthesis and characterization of K-promoted HTlcs for high temperature $CO_2$ capture and concentration. Results from his work are forthcoming.
- A new PhD student is being sought to take Steven's place in the group. This new PhD student will tentatively join the group in the Fall of 2006.