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. Some key questions that are trying to be answered in Phase II 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. 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.

ACCOMPLISHMENTS TO DATE:

During this period of performance (12 months), the stripping reflux (SR) PSA code was used to continue to study various SR PSA cycle configurations with and without heavy and or light reflux and with or without recovery or feed plus recycle steps 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. Key findings included: the light reflux step was very important to the process performance, making the SR PSA cycle a dual reflux configuration; mass transfer effects were important, with a decrease in the mass transfer resistance allowing the performance for the first time to exceed a CO_2 purity of 90% at a CO_2 recovery over 98%; including a recovery or a feed plus recycle step did not have a significant effect on the process performance. 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; and that the five times greater mass transfer coefficients that led to the significant increase in the process performance stated above was validated through the development of a non-equilibrium kinetic model that describes the reversible adsorption and desorption behavior of CO_2 in a K-Promoted HTlc. These findings have been published or are under review .

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 the SR PSA cycle configuration on the process performance. Since high CO_2 purities and CO_2 recoveries can both be achieved now, the focus will be to increase the feed throughput by learning how to configure unequal step time cycles; the simulation of these kinds of novel SR PSA cycles is just underway. 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. Commercial K-promoted HTLC has finally been obtained; fixed bed testing with it will be under way soon.

LIST OF JOURNAL ARTICLES PUBLISHED, IN PRESS OR UNDER REVIEW

- 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.*, **45**, 4278-4294 (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.*, 45, 6387-6392 (2006).
- S. P. Reynolds, A. D. Ebner and J. A. Ritter, "Carbon Dioxide Capture from Flue Gas by PSA at High Temperature using a K-Promoted HTlc: Effects of Mass Transfer on the Process Performance," *Environmental Progress*, 25, 334-342 (2006).
- A. D. Ebner, S. P. Reynolds and J. A. Ritter, "Non-Equilibrium Kinetic Model that Describes the Reversible Adsorption and Desorption Behavior of CO_2 in a K-Promoted HTlc," *Ind. Eng. Chem. Res.*, 46, 1737-1744 (2007).
- S. P. Reynolds, A. Mehrotra, A. D. Ebner and J. A. Ritter, "Heavy Reflux PSA Cycles for CO_2 Recovery from Flue Gas. Part I. Performance Evaluation," *Ind. Eng. Chem. Res.*, submitted (2007).

LIST OF PRESENTATIONS:

- 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," AIChE 2006 Annual Meeting, San Francisco, CA, November 2006.
- S. P. Reynolds, A. D. Ebner and J. A. Ritter, "Novel Heavy reflux PSA Cycles for the Recovery of Carbon Dioxide at high temperature with K-Promoted HTlc," AIChE 2006 Annual Meeting, San Francisco, CA, November 2006.
- S. P. Reynolds, A. D. Ebner, and J. A. Ritter, "Non-Equilibrium Dynamic Adsorption and Desorption Isotherms of CO_2 on a K-Promoted HTlc," 4^{th} Pacific Basin Conference on Adsorption Science and Technology, Tianjin, China, May 2006, contributed.
- 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," 4^{th} Pacific Basin Conference on Adsorption Science and Technology, Tianjin, China, May 2006, contributed.

STUDENTS SUPPORTED UNDER THIS GRANT

- Steven P. Reynolds, PhD candidate, Department of Chemical Engineering, University of South Carolina. Steven, while being supported through this grant, and a MeadWestvaco Fellowship, worked on PSA code development, and has carried out thousands of simulations and corresponding analyses of nine different SR PSA cycle configurations. He will continue on this project until he graduates with his PhD in May 2007.
- Hai Du, PhD candidate, Department of Chemical Engineering, University of South Carolina. Hai, as a relatively 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.
- Amal Mehrotra, PhD candidate, Department of Chemical Engineering, University of South Carolina. Amal, as a new PhD student in the group that joined in August 2006 was brought in to take Steven's place. He is being supported through this grant and a MeadWestvaco Fellowship. Amal has been coming up to speed very quickly on PSA code development and has been carrying out simulations of novel PSA cycle configurations that include recovery and feed plus recycle steps. He will remain on this project until its completion.