TITLE: NEW ADSORPTION CYCLES FOR CARBON DIOXIDE CAPTURE AND CONCENTRATION

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ABSTRACT

Objectives

Since a viable separations technology has yet to be identified for the cost-effective capture of CO_2 from flue gas, and based on the results obtained during the IC Phase I project, "Radically New Adsorption Cycles for Carbon Dioxide Sequestration," the objective of this 3 yr continuation project is to study new pressure swing adsorption (PSA) cycles 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 PSA cycles. Some key questions that are being 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 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 12 mo period of performance four tasks were carried out, with key results obtained for each one. The first task was the modification of the rigorous PSA process simulator for studying various HR PSA cycle configurations, but now with an unequal step times. The key results were that the new version of the code is running and unequal step time cycles are being studied. The second task was the development of an ideal HR PSA process simulator based on equilibrium theory. This type of model predicts the ultimate separation possible for a given set of conditions, while providing fundamental insight to the process performance. The key result so far was that the equilibrium theory model verified that the inclusion of a recovery or feed plus recycle step does not have any effect on the process performance. This finding is contrary to current understanding, but in agreement with results from the rigorous PSA process simulator. The third task was the design and construction of a fixed bed apparatus to study the adsorption breakthrough and desorption elution behavior of commercial K-promoted HTlc on a scale of hundreds of grams instead of milligrams. The key results so far are that the system is operational and preliminary results have been obtained that are being analyzed. The fourth task was the extension of the non-equilibrium kinetic model that describes the reversible adsorption and desorption behavior of CO_2 in K-promoted HTlc to include not only temperature dependence but also pressure dependence. The key results of this task are that the model is ready for inclusion in the PSA process simulator. A study is under way that compares the performance obtained from a given

HR PSA cycle when using the different models that describe the uptake and release of CO_2 in K-Promoted HTlc.

Future Work

This three-year grant began August 2005 and will end on July 31, 2008. This project began as a continuation project 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 through the end of the award will be to increase the feed throughput by configuring unequal step time PSA cycles. To this end, a novel cycle sequencing strategy has been developed that easily discloses how to design unequal step time PSA cycles. The simulation of these 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 is now being studied; TGA and fixed bed testing is under way. Of the three key questions posed to answer during this Phase II project, i.e., 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, questions 1 and 2 are essential complete and question 3 will be addressed during the final six months of the project.

2. LIST OF JOURNAL ARTICLES, PRESENTATIONS AND STUDENTS SUPPORTED

Journal Articles Published, in Press or Under Review

- 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," *Adsorption*, 14 399-413 (2008).
- H. Du, A. D. Ebner and J. A. Ritter, "New Model that Describes the Temperature Dependence of the Adsorption and Desorption Behavior of CO_2 in a K-Promoted HTlc," *Ind. Eng. Chem. Res.*, submitted (2008).
- A. Mehrotra, S. P. Reynolds, A. D. Ebner and J. A. Ritter, "Heavy Reflux PSA Cycles for CO_2 Recovery from Flue Gas. Part II. Interpretation of Trends," *Ind. Eng. Chem. Res*, submitted (2008).

Presentations

- A. D. Ebner, H. Du and J. A. Ritter, "Temperature Dependent Non-Equilibrium Kinetic Model that Describes the Reversible Adsorption and Desorption Behavior of CO_2 in a K-Promoted HTlc," AIChE 2007 Annual Meeting, Salt Lake City, UT, November 2007.
- A. Mehrotra, A. D. Ebner and J. A. Ritter, "Extreme Configurations in Heavy Reflux PSA Cycles," AIChE 2007 Annual Meeting, Salt Lake City, UT, November 2007.
- S. P. Reynolds, A. Mehrotra, A. D. Ebner and J. A. Ritter, "Novel Heavy Reflux Cycles in Pressure Swing Adsorption Processes," Fundamentals of Adsorption FOA9, Giardini Naxos, Italy, May 2007, contributed.

Students Supported Under this Grant

- Steven P. Reynolds, PhD candidate, Department of Chemical Engineering, USC. Steven, while being supported through this grant, and a MeadWestvaco Fellowship, worked on PSA code development. He successfully defended his PhD in May 2007 and is now working for Flour.
- Hai Du, PhD candidate, Department of Chemical Engineering, USC. Hai, while being supported through this grant and a MeadWestvaco Fellowship, is working on the synthesis, characterization and fixed bed testing of K-promoted HTlc for high temperature CO_2 capture and concentration. Results from his work are forthcoming, and he will remain on this project until its completion.
- Amal Mehrotra, PhD candidate, Department of Chemical Engineering, USC. Amal, while being supported through this grant and a MeadWestvaco Fellowship is developing HR PSA models based on equilibrium theory, carrying out simulations of novel PSA cycle configurations, and developing novel ways to sequence PSA cycles. Results from his work are forthcoming, and he will remain on this project until its completion.