

**ANNUAL PROJECT REPORT  
AS OF DECEMBER 1999**

1. PROJECT SPONSOR:

Department of Chemical Engg.  
The Ohio State University  
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2. PROJECT MANAGER:

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Professor and Chair  
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3. OCDO GRANT NO: OCRC/99-A4.1

4. PROJECT UPDATE ✓ OR  
FINAL REPORT

5. PROJECT TITLE: Kinetics and Mechanism of H<sub>2</sub>S/Sorbent Interaction at High Temperature and Pressure

6. PROJECT TERM: FROM: September 1, 1999

TO: August 31, 2000

7. PROJECT CO-SPONSORS:

<u>NAME</u>	<u>COST SHARE</u>
OCDO	\$ 74,766
Ohio State University	\$ 34,914
Total Project Cost:	\$ 109,680

**I. ABSTRACT**

8. OBJECTIVES:

The Ohio coal contains large amount of sulfur. When this coal is gasified in advanced coal-based power generation systems, such as IGCC, the sulfur in the coal forms hydrogen sulfide gas (H<sub>2</sub>S). The removal of H<sub>2</sub>S is necessary not only for the protection of the gas turbine hardware, but also to comply with the environmental regulations. Coal gas desulfurization to sufficiently low levels at higher pressures and temperatures of above 500°C is now recognized as crucial to efficient and economical coal utilization in advanced IGCC systems. The primary focus of this project is to investigate the reaction kinetics and mechanism of H<sub>2</sub>S/sorbent interaction at high pressures and temperatures representative of advanced integrated coal gasification systems.

9. WORK DONE AND CONCLUSIONS:

Experiments were conducted to obtain high-pressure and high temperature kinetic data for uncalcined limestone, fully-calcined dolomite and half-calcined dolomite sulfidation. The effects of pressure, temperature, partial pressure of H<sub>2</sub>S, initial sorbent surface area and pore volume were investigated. Increasing pressure had an adverse effect on sulfidation conversion of fully-calcined dolomite, which was attributed to a reduction in surface area and pore volume of the sorbent generated by in-situ calcination of dolomite at high pressures.

Uncalcined limestone and half-calcined dolomite sulfidation conversion faced an adverse effect of increasing pressure due to the stoichiometry of the reaction. The kinetic data of limestone/dolomite sulfidation revealed the basic kinetic parameters, such as, order of reaction and activation energy.

#### 10. PLANS FOR THE COMING YEAR:

Fourth year of the project would involve the study of sulfidation characteristics of metal oxide sorbents (ZnO, Fe<sub>2</sub>O<sub>3</sub>, ZnFe<sub>2</sub>O<sub>4</sub>) supported on a SiC matrix under fixed bed conditions. Various operating parameters such as pressure, temperature and gas-phase composition would be varied to determine the optimum sulfidation conditions and obtain relevant kinetic parameters. Experiments would effectively cover the entire range of operating temperatures and pressures, *i.e.*, 500-700°C and 15-20 atm. The effect of sorbent/support structural properties, such as, surface area, pore volume and pore size distribution would be investigated to understand H<sub>2</sub>S removal. As a commercially viable support has to last at least over 100 sulfidation/regeneration cycles, cyclical sulfidation/regeneration studies would be undertaken to study the viability of SiC as a support.

## II. HIGHLIGHTS/ACCOMPLISHMENTS

The calcium-based sorbents can be used to effectively remove bulk of coal gas H<sub>2</sub>S at high temperature and high-pressure in-gasifier conditions. This would result in cost reduction in downstream polishing units utilizing relatively expensive metal oxide sorbents. The removal of H<sub>2</sub>S occurs by reaction with CaO/CaCO<sub>3</sub> (either from limestone or dolomite) to form solid product CaS, which is removed with ash and stabilized to form CaSO<sub>4</sub> before disposal. Presence of inert MgO in dolomite gives it a more porous matrix after calcination, which enables complete utilization of the calcium in dolomite and gives better sulfidation conversions. The distinguishing feature of this study is that it gives for the first time the high pressure limestone/dolomite sulfidation kinetic data that most closely simulates the “in-gasifier” conditions of H<sub>2</sub>S removal. This high pressure kinetic data would help in designing an optimum sorbent for H<sub>2</sub>S removal.

## III. ARTICLES/PRESENTATIONS

1. “H<sub>2</sub>S Removal using Metal Oxide and Ca-based Sorbents at High Pressure and High Temperature: Kinetics and Mechanism,” Santhosh Misro, R. Agnihotri, S. Chauk, R. Jadhav, H. Gupta and L.-S. Fan, presented at AIChE Annual Meeting, Dallas, TX, Nov. 1999.
2. “Sulfidation of Dolomite at High Pressure and High Temperature: Effect of Initial Sorbent Structural Properties,” Santhosh K. Misro, R. Jadhav, H. Gupta, S. Chauk, R. Agnihotri and L.-S. Fan, *Proceedings of 16th Annual International Coal Conference: Coal - Energy and Environment*, Pittsburgh, PA, Oct. 1999.
3. “High-Pressure Reaction Kinetics of Hydrogen Sulfide and Uncalcined Limestone Powder,” Agnihotri, R., Chauk, S. S., Misro, S. K., Fan, L.-S., *Industrial & Engineering Chemistry Research*; 1999; 38(10); 3802-3811.