

## **Conversion of Hydrogen Sulfide in Coal Gases to Elemental Sulfur with Monolithic catalysts**

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### **OBJECTIVES**

Removal of hydrogen sulfide ( $H_2S$ ) from coal gas and sulfur recovery as elemental sulfur are key steps in the development of Department of Energy's (DOE's) advanced gasification plants that employ coal and natural gas, and produce electric power and clean transportation fuels. The conventional method of sulfur removal and recovery employing amine scrubbing, Claus, and tail-gas treatment involves a number of steps and is energy intensive. A novel process called Single-step Sulfur Recovery Process (SSRP) is under development at DOE/National Energy Technology Laboratory (NETL), Research Triangle Institute (RTI) and Tuskegee University. In this process, the  $H_2S$  in the coal gas is selectively oxidized in a single step to elemental sulfur using sulfur dioxide ( $SO_2$ ) or oxygen ( $O_2$ ) in the presence of an alumina-or-carbon-based catalyst via the Claus reaction. Burning a portion of the sulfur makes the required  $SO_2$ . Micro reactor studies have shown the potential of converting 99 % of the  $H_2S$  in coal gases to elemental sulfur with a COS slip as low as 40 ppmv. The commercial embodiment envisioned is a monolithic catalyst reactor (MCR) containing honeycomb catalysts through which coal gas is passed at 125 to 155°C. A special flow regime (Taylor flow) in a monolithic catalyst channel removes the sulfur formed at the catalyst surface thereby regenerating the catalyst in-situ. The process has the potential to convert selectively  $H_2S$  to elemental sulfur by wetting the interface between the solid catalyst surface and gas phase with a thin liquid sulfur film, which is developed with the aid of the special flow pattern (Taylor flow) in parallel catalyst channels. The process has also the potential to reduce undesired conversion of produced elemental sulfur to COS with the aid of the special flow pattern (Taylor flow) in parallel catalyst channels.

The objectives of this research are to formulate monolithic catalysts with ceramic honeycomb monolithic catalyst support, gamma-alumina or carbon wash coat, and catalytic metals, to measure reaction kinetics of both conversion of  $H_2S$  to elemental sulfur and formation of COS in the presence of major coal gas components with a fabricated monolithic catalyst reactor, and to develop a model of SSRP based on reaction kinetics from the fabricated monolithic catalyst reactor.

Exploratory experiments on conversion of hydrogen sulfide into liquid elemental sulfur in the

absence of carbon monoxide were carried out in a reactor equipped with a 400 cells/inch<sup>2</sup> (CPSI)  $\gamma$ -alumina-wash-coated CORDIERITE ceramic honeycomb monolithic catalyst for the space time range of 110 - 556 seconds at 125 - 155°C to evaluate effects of reaction temperature, H<sub>2</sub>S concentration, and reaction pressure on conversion of hydrogen sulfide into liquid elemental sulfur. Simulated coal gas mixtures consist of 50 - 78 v% hydrogen, 5,000 – 10,000-ppmv hydrogen sulfide, 2,500 - 5,000 ppmv sulfur dioxide, and 5 - 21 vol % moisture, and nitrogen as remainder. The volumetric feed rate of a simulated coal gas mixture to the honeycomb catalyst reactor is 50 cm<sup>3</sup>/min at room temperature and atmospheric pressure. The temperature of the reactor is controlled in an oven at 125 - 155°C. The pressure of the reactor is maintained at 25 - 184 psia. The molar ratio of H<sub>2</sub>S to SO<sub>2</sub> in the honeycomb catalyst reactor is maintained at around 2 for all the reaction experiment runs.

### **ACCOMPLISHMENTS TO DATE**

- o Conversion of H<sub>2</sub>S into elemental sulfur increases with reaction pressure in the pressure range of 20 - 80 psia, and decreases with reaction pressure in the pressure range of 80 -180 psia.
- o Conversion of H<sub>2</sub>S into elemental sulfur decreases with increased reaction temperatures over the reaction temperature range of 125 – 155°C.
- o Conversion of H<sub>2</sub>S into elemental sulfur decreases with increased moisture over the moisture concentration range of 5 – 21 v%.
- o Catalytic activities of the honeycomb catalyst decrease up to 20-hrs reaction duration in the conversion of H<sub>2</sub>S into elemental sulfur with a 50-SCCM feed stream containing 5,000-ppmv H<sub>2</sub>S, 2,500-ppmv SO<sub>2</sub>, 10-v% moisture, and 70-v% H<sub>2</sub> for 6 - 33 hrs at 140°C and 120 psia.
- o Conversion of H<sub>2</sub>S into elemental sulfur increases with increased initial concentration of H<sub>2</sub>S at 140°C and 120 psia.

### **FUTURE WORK**

A reactor equipped with a honeycomb monolithic catalyst will be operated in the presence of CO and H<sub>2</sub> as the components of a simulated coal gas to accomplish the above-mentioned objectives.

### **PUBLICATIONS AND PRESENTATIONS**

Oxidation of H<sub>2</sub>S in Coal Gases to Liquid Elemental Sulfur with Monolithic Catalysts, Kyung C. Kwon, Suresh C. Jain, Melanie N Ratcliffe, Monica I McCoy and Crystal B. Jones, Presented at AIChE 2004 Annual Meeting, Austin, TX, November 7 - 12.

### **AWARDS RECEIVED AS A RESULT OF SUPPORTED RESEARCH**

None is awarded.

### **STUDENTS SUPPORTED UNDER THIS GRANT**

Monica I McCoy, Crystal Jones, and Melanie N Ratcliffe