

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 several research organizations. 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 . A monolithic catalyst reactor (MCR) containing honeycomb catalysts is used to study conversion of H_2S into liquid elemental sulfur by passing simulated H_2S -containing coal gases through honeycomb catalysts 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 for removal of H_2S from coal gases and minimum formation of COS by impregnating catalytic metals into monolithic γ -alumina wash-coated catalyst supports, to develop a catalytic regeneration method for deactivated monolithic catalysts, to measure kinetics of both direct oxidation of H_2S to elemental sulfur with SO_2 as an oxidizer and formation of COS in the presence of a simulated coal gas mixture containing H_2S , H_2 , CO, CO_2 , and moisture, using a monolithic catalyst reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants. This heterogeneous catalytic reaction has gaseous reactants such as H_2S and SO_2 . However, this heterogeneous catalytic reaction has heterogeneous products such as liquid elemental sulfur and steam.

Experiments on conversion of hydrogen sulfide to elemental sulfur using a monolithic catalyst reactor were carried out for the space time range of 90 – 560 seconds at 140 - 155°C and 118 - 124 psia to evaluate effects of space time, reaction temperature, moisture concentration, and catalyst age on conversion of hydrogen sulfide to elemental sulfur and formation of COS. Simulated coal gas mixtures consist of 3,600 – 4,000-ppmv hydrogen sulfide, 1,800 – 2,000 ppmv sulfur dioxide, 24 – 25 v% hydrogen, 37 – 41 v% CO, 10 – 11.2 v% CO_2 , 0 – 10 vol % moisture, and nitrogen as remainder.

Volumetric feed rates of a simulated coal gas mixture to the monolithic catalyst reactor are 30 -180 SCCM. Each reaction experimental run proceeds after a blank experimental run, which is carried out in the absence of moisture and a monolithic catalyst. The molar ratio of H₂S to SO₂ in the monolithic catalyst reactor is approximately maintained at 2 for all the reaction experiment runs.

ACCOMPLISHMENTS TO DATE

- 0 Conversion of H₂S to elemental sulfur and formation of COS for both the reaction runs and the blank runs increase with space time. However, formation of COS for the reaction runs is higher than that for the blank runs over the space time range of 90 – 560 s. The difference of COS formation between the reaction runs and the blank runs increases with space time. Formation of COS for the reaction runs increases with conversion of H₂S to elemental sulfur over the space time range of 90 – 560 s. In other words, formation of COS for the reaction runs increases with formation of elemental sulfur over the space time range of 90 – 560 s.
- 0 Conversion of H₂S to elemental sulfur does not follow the Arrhenius' equation. Reaction temperature affects conversion of H₂S to elemental sulfur. Conversion of H₂S to elemental sulfur decreases with increased reaction temperature over the temperature range of 140 - 155°C. However, formation of COS increases with increased reaction temperature.
- 0 Conversions of H₂S to elemental sulfur and formation of COS in a monolithic catalyst reactor decreases with increased concentration of moisture.
- 0 Conversion of H₂S to elemental sulfur decreases linearly with increased catalyst age, whereas formation of COS increases with increased catalyst age. Formation of COS for the blank runs is higher than that for the reaction runs, and formation of COS for the reaction runs decreases generally with increased conversion of H₂S at various catalyst ages with space times held constant. In other words, formation of COS for the reaction runs decreases with decreased catalyst age, and increased formation of elemental sulfur at various catalyst ages with space times held constant.

FUTURE WORK

Monolithic catalysts for removal of H₂S from coal gases and minimum formation of COS will be formulated by impregnating catalytic metals into monolithic γ -alumina wash-coated catalyst supports.

PUBLICATIONS AND PRESENTATIONS

1. Oxidation of H₂S in Coal Gases to Liquid Elemental Sulfur with Monolithic Catalysts, Presented at AIChE Annual Meeting, Austin, TX, November 7 – 12, 2004.
2. Deactivation of Catalysts in Removal of Hydrogen Sulfide in Coal Gases as Liquid Sulfur, presented at AIChE Annual Meeting, Cincinnati, OH, November 2, 2005.
3. Removal of Hydrogen Sulfide in Coal Gases using a Monolithic Catalyst Reactor, presented at AIChE Annual Meeting, Cincinnati, OH, November 1, 2005.

AWARDS RECEIVED AS A RESULT OF SUPPORTED RESEARCH

None is awarded.

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