

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

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OBJECTIVES

The objectives of this research are to formulate monolithic catalysts for removal of H₂S 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 H₂S to elemental sulfur with SO₂ as an oxidizer and formation of COS in the presence of a simulated coal gas mixture containing H₂S, H₂, CO, CO₂, 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 H₂S and SO₂. 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 46 – 570 seconds at 120 - 150°C and 40 - 210 psia to evaluate effects of space time, reaction temperature, and pressure 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, 23 – 27 v% hydrogen, 36 – 41 v% CO, 10 – 12 v% CO₂, 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

Conversion of H₂S to elemental sulfur does not follow the Arrhenius' equation. Conversion of H₂S to elemental sulfur increases with increased reaction temperature over the temperature range of 120 -140°C, whereas conversion of H₂S to elemental sulfur decreases with increased reaction temperature over the temperature range of 140 -150°C. Catalytic formation of COS levels off and is lowest over the temperature range of 120 -130°C, increases with increased reaction temperature over the temperature range of 130 -140°C, and decreases with increased reaction temperature over the temperature range of 140 -150°C.

Formation of COS for both the reaction runs and the blank runs increases with space time over the space time range of 90 – 190 s and decreases with increased space time over the space time range of 190 – 550 s. Catalytic formation of COS for the reaction runs is slightly lower than thermal formation of COS for the blank runs over the space time range of 140 – 280 s. Catalytic formation of COS for the reaction runs is much lower than thermal formation of COS for the blank runs at the space time 90 s, whereas catalytic formation of COS

for the reaction runs is much higher than thermal formation of COS for the blank runs at the space time 550 s. Conversion of H₂S to elemental sulfur increases with space time. Catalytic formation of COS for the reaction runs appears to be independent of conversion of H₂S to elemental sulfur over the space time range of 90 – 570 s.

Conversion of H₂S, catalytic formation for reaction runs of COS, and thermal formation of COS for blank runs also increases with increased total pressure. Catalytic formation of COS is not significantly different from thermal formation of COS at 140°C and 40 – 210 psia. Catalytic formation of COS is independent of H₂S conversion over the H₂S conversion range of 0.68 – 0.83.

Conversion of H₂S to elemental sulfur does not decrease significantly with increased age of 140°C-nitrogen regenerated catalyst up to 3900 hr catalyst age. Catalytic formation of COS decreases with increased age of regenerated catalyst. Thermal formation of COS for the blank runs is lower than catalytic formation of COS for the reaction runs.

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. A reaction model on conversion of H₂S to elemental sulfur and formation of COS will be developed with experimental data

PUBLICATIONS AND PRESENTATIONS

Oxidation of H₂S in Coal Gases to Liquid Elemental Sulfur with Monolithic Catalysts, Presented at AIChE Annual Meeting, Austin, TX, November 7 – 12, 2004.

Deactivation of Catalysts in Removal of Hydrogen Sulfide in Coal Gases as Liquid Sulfur, presented at AIChE Annual Meeting, Cincinnati, OH, November 2, 2005.

Removal of Hydrogen Sulfide in Coal Gases using a Monolithic Catalyst Reactor, presented at AIChE Annual Meeting, Cincinnati, OH, November 1, 2005.

Formation of Liquid Element Sulfur and Gaseous Carbonyl Sulfide by Reacting Hydrogen Sulfide in Coal Gases with Sulfur Dioxide, presented at 2006 AIChE National Meeting, San Francisco, November 14, 2006

Cleanup of Coal Gases by Removing Hydrogen Sulfide in the Form of Liquid Element Sulfur, presented at 2006 AIChE National Meeting, San Francisco, November 14, 2006

Oxidation of Hydrogen Sulfide in Coal Gases to Liquid Element Sulfur, Using a Monolithic Catalyst Reactor, presented at 2006 AIChE National Meeting, San Francisco, November 14, 2006

Conversion of Hydrogen Sulfide in Coal Gases to Liquid Elemental Sulfur with Monolithic Catalysts, 2006 DOE/NETL HBCU/OMI Contractors Review Conference, June 8, 2006

AWARDS RECEIVED AS A RESULT OF SUPPORTED RESEARCH

None is awarded.

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