ENERGY

Kinetics of Direct Oxidation of H_2S in Coal Gas to Elemental Sulfur

Annual Technical Progress Report for the Period
October 1, 2001 to September 30, 2002

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January 2003

Work Performed Under Contract No
DE-FG26-00NT40835

For
U.S. Department of Energy
National Energy Technology Laboratory
Pittsburgh, PA 15236-0940

By
Tuskegee University
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DISCLAIMER

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SUMMARY

The direct oxidation of H_2S to elemental sulfur in the presence of SO_2 is ideally suited for coal gas from commercial gasifiers with a quench system to remove essentially all the trace contaminants except H_2S. This direct oxidation process has the potential to produce a super clean coal gas more economically than both conventional amine-based processes and HGD/DSRP. The objective of this research is to support the near- and long-term DOE efforts to commercialize this direct oxidation technology. The objectives of this research are to measure kinetics of direct oxidation of H_2S to elemental sulfur in the presence of a simulated coal gas mixture containing SO_2, H_2, and moisture, using 60-μm C-500-04 alumina catalyst particles and a PFA differential fixed-bed micro reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants.

To achieve the above-mentioned objectives, experiments on conversion of hydrogen sulfide into elemental sulfur were carried out for the space time range of 0.01 – 0.047 seconds at 125 - 155^oC to evaluate effects of reaction temperatures, moisture concentrations, reaction pressures on conversion of hydrogen sulfide into elemental sulfur. Simulated coal gas mixtures consist of 61 – 89 v% hydrogen, 2,300 - 9,200-ppmv hydrogen sulfide, 1,600 - 4,900 ppmv sulfur dioxide, and 2.6 – 13.7 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the reactor are 100 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). The temperature of the reactor is controlled in an oven at 125 - 155^oC. The pressure of the reactor is maintained at 28 - 127 psia.

The following results were obtained based on experimental data generated from the differential reactor system, and their interpretations,

1. Concentration of moisture and concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the moisture range of 2.5 – 13.6 v% moisture at 140^oC and 120 – 123 psia.

2. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the temperature range of 135 - 145^oC at 5-v% moisture and 112 – 123 psia. However, reaction rates of H_2S with SO2 appear to decrease slightly with increased reaction temperatures over the temperature range of 135 - 145^oC at 5-v% moisture and 112 – 123 psia.

3. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the pressure range of 28 – 123 psia at 5-v% moisture and 140^oC. However, reaction rates of H_2S with SO_2 increase significantly with increased reaction pressures over the pressure range of 28 – 123 psia at 5-v% moisture and 140^oC.

INTRODUCTION

Coal is our most abundant energy resource. It is strategically important to our nation to increase coal use as an energy source in an environmentally acceptable manner. Coal gasification, a primary step in advanced coal utilization processes, produces a coal gas containing hydrogen (H_2) and carbon monoxide (CO) as the fuel components. Raw coal gas however, also contains a number of major and trace contaminants including hydrogen sulfide (H_2S), carbonyl sulfide (COS), ammonia (NH_3), hydrogen chloride (HCl), alkali, heavy metals, and particulate. Thus, this gas must be cleaned before further use. H_2S is a major coal gas contaminant that can range from 1000 to 10,000 ppm, depending on the sulfur content of the coal. Removal of H_2S from coal gas and sulfur recovery are key steps in the development of Department of Energy's (DOE's) advanced Vision 21 plants combining a power plant and a refinery based on coal and natural gas to co-produce electricity and clean transportation-grade liquid fuels. These Vision 21 plants will require highly clean coal gas with H_2S below 1 ppm and negligible amounts of other contaminants such as COS, HCl, NH_3, alkali, heavy metals, and particulate.

The conventional method of removing H_2S and sulfur recovery involves a number of steps including amine scrubbing at low temperature followed by amine regeneration using steam to produce a concentrated H_2S-containing gas. This concentrated H_2S-containing gas is then combusted to produce a gas with a H_2S to sulfur dioxide (SO_2) ratio of 2 to 1 in a Claus furnace. This is followed by up to three (3) stages of Claus reaction at temperatures of around 250-280°C over an alumina catalyst to recover elemental sulfur:

$$2\ H_2S\ (g) + SO_2\ (g) \leftrightarrow 3S\ (\ell) + 2\ H_2O\ (g)$$

The Claus reaction is exothermic and equilibrium limited. To circumvent equilibrium limitations, the reaction is conducted in up to three (3) reaction stages with interstage cooling/ sulfur condensation followed by interstage re-heating. However, even with three (3) stages, the reaction is not complete due to thermodynamic limitations at 250°C. The Claus tail gas contains sulfur that must be further treated in an expensive tail gas treatment plant (e.g., SCOT) before discharge. Thus, overall H_2S removal and sulfur recovery using this conventional sequence are extremely cumbersome, equipment intensive, and expensive.

A second generation approach for sulfur removal/recovery developed under DOE's sponsorship involves three steps:

(i) hot-gas desulfurization (HGD) using regenerable zinc oxide-based sorbents

$ZnO + H_2S \leftrightarrow ZnS + H_2O$ (HGD)

(ii) sorbent regeneration using air to produce SO_2

$ZnS + 3/2\, O_2 \leftrightarrow SO_2 + ZnO$ (regeneration)

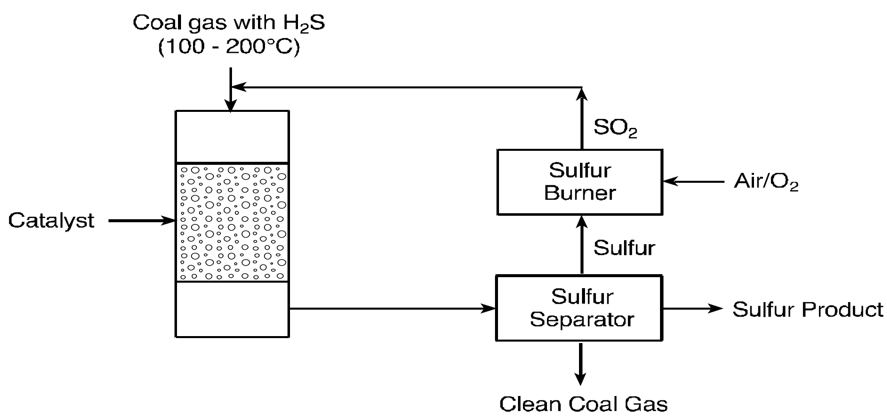
(iii) catalytic reduction of SO_2 using a small portion of the coal gas, to elemental sulfur by the Direct Sulfur Recovery Process (DSRP):

$SO_2 + 2H_2$ (or 2CO) $\leftrightarrow S + 2H_2O$ (or $2CO_2$) (DSRP)

This approach integrates well with a coal gasifier in an integrated gasification (IGCC) system because the raw coal gas does not have to be cooled all the way down to near room temperature as is the case with the conventional amine/Claus/tail-gas treatment method. However, the overall process scheme requires solid sorbent handling/circulation, and three separate reactors. Also, there is a small energy penalty associated with the use of coal gas to reduce SO_2 by DSRP. Furthermore, since trace contaminants e.g. NH_3 and HCl are not removed by the zinc-based sorbents. This approach is primarily targeted towards the development of advanced IGCC plants that produce electricity only (but do not coproduce both electricity and clean transportation grade fuels).

There is an immediate as well as long-term need for the development of clean processes that produce highly clean coal gas for next generation Vision 21 plants producing both electricity and transportation-grade liquid fuels. To this end, our subcontractor, Research Triangle Institute (RTI) is developing a novel process in which the H_2S in coal gas is directly oxidized to elemental sulfur over a selective catalyst using sulfur dioxide (SO_2) produced by burning a portion of the sulfur produced as shown in Figure 1.

Figure 1: RTI proposed direct oxidation process



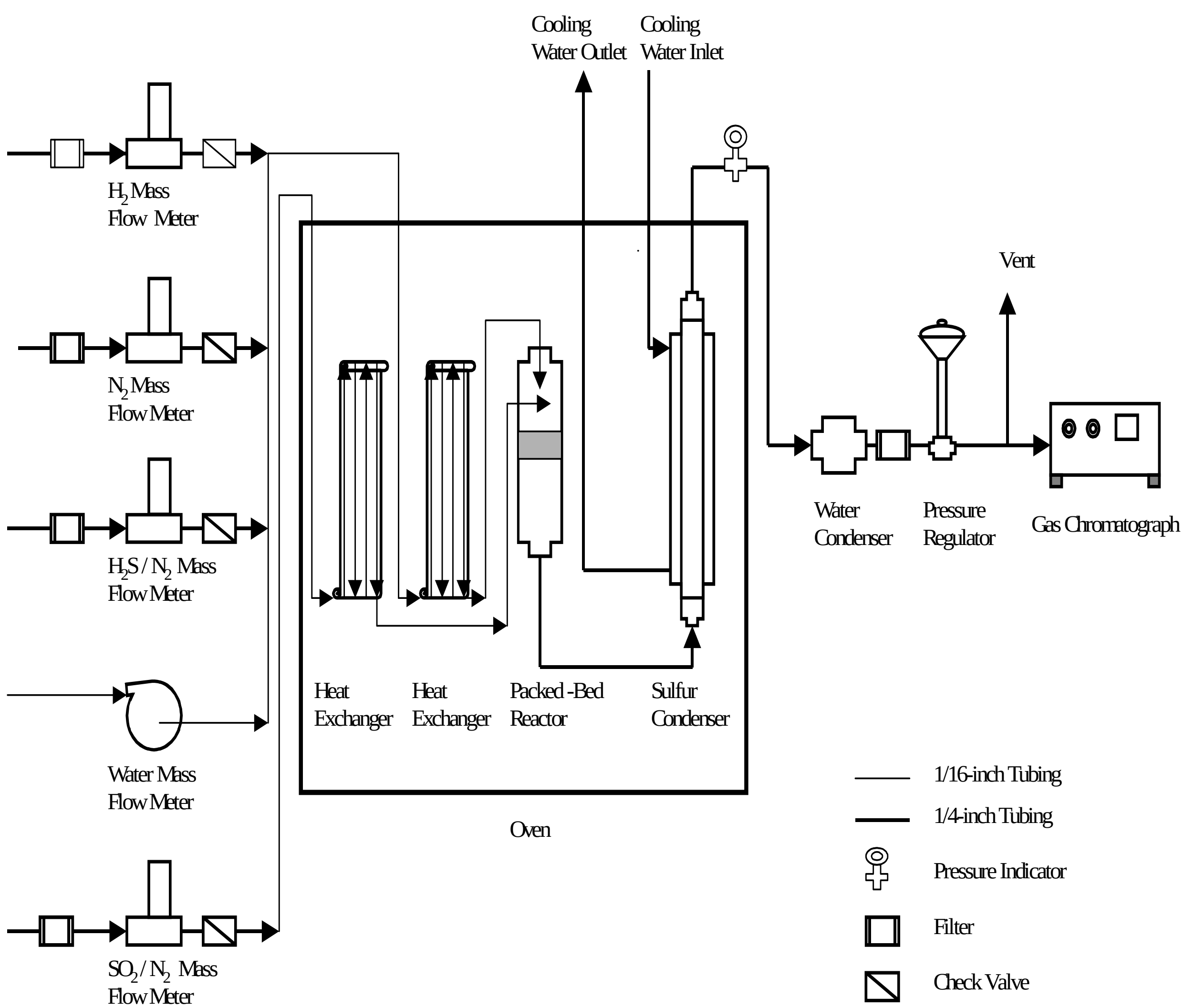
The direct oxidation process shown in Figure 1 is ideally suited for coal gas from a commercial gasifier with a quench system. During quench, the trace contaminants (except sulfur) are essentially completely removed and H_2S (with some COS) remains as the only contaminant. The gas contains all of the major coal gas components including H_2, CO, CO_2 and H_2O.

The objectives of this research are to measure kinetics of direct oxidation of H_2S to elemental sulfur in the presence of a simulated coal gas mixture containing SO_2, H_2, and moisture, using 60-μm C-500-04 alumina catalyst particles and a PFA differential fixed-bed micro reactor, and to develop kinetic rate equations and model the direct oxidation process to assist in the design of large-scale plants. Experiments on conversion of hydrogen sulfide into elemental sulfur were carried out for the space time range of 0.01 – 0.047 seconds at 125 - 155^{o}C and 28 - 127 psia to evaluate effects of reaction temperatures, moisture concentrations, reaction pressures on conversion of hydrogen sulfide into elemental sulfur. Simulated coal gas mixtures consist of 61 – 89 v% hydrogen, 2,300 - 9,200-ppmv hydrogen sulfide, 1,600 - 4,900 ppmv sulfur dioxide, and 2.6 – 13.7 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the reactor are 100 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM).

EXPERIMENTAL SETUPS

A differential fixed-bed micro reactor was fabricated with a ¼-inch PFA tee. The volume of a C-500-04 alumina catalyst packed in the reactor is 0.012 cm^3. The C-500-04 alumina catalyst in the form of 160-μm spherical particles was examined. A simulated coal gas mixture containing H_2S and SO_2 was reacted with the aid of the catalyst in the differential fixed-bed micro reactor at 125 - 155°C. Conversion of hydrogen sulfide into elemental sulfur was analyzed with the gas chromatograph. The range of space (residence) time of the reaction gas mixture in the reactor was 0.01 – 0.047 s under the reaction conditions. Space times are obtained by dividing the volume of a catalyst bed with a volumetric flow rate at reaction conditions

Figure 2. Schematic Diagram of the packed-bed reactor assembly.



A differential reactor assembly mainly consists of four mass flow meters for gases, one differential reactor, two preheaters, one high pressure liquid pump for water, one four-way switch valve, one oven, five filters for gases, four check valves, and one water collection bottle (see Figure 1). The differential reactor is fabricated with one ¼-inch PFA tee. The preheaters are made of 1/16-inch PFA tubing. The reactor was loaded with C-500-04 alumina catalyst. The reactor, loaded with the catalyst particles, was placed inside the oven to be heated at a desired temperature. Nitrogen was introduced into the catalyst-loaded reactor during preheating the reactor. When the temperature of the reactor was raised at the desired temperature, one simulated coal gas mixture stream containing H_2S and another feed stream containing SO_2 were introduced into the reactor, by switching nitrogen with the simulated coal gas mixture. The typical reaction conditions are shown in Table 1. The properties of the catalyst are shown in Table 2. The experimental data are shown in Tables 3 through 8, where the experimental data with asterisk (*) were used for constructing the Figures 3 through 14.

Table 1. Experimental conditions for the reaction of hydrogen sulfide with sulfur dioxide.

Volume of the catalyst-packed bed, cm^3:	0.012
Temperature, ^oC:	125 - 155
Reaction Pressure, psia	28 - 127
Space Time under the reaction conditions, s:	0.01 – 0.047
Mean Particle Size, μm	160
Amount of Catalyst, g	0.01
Gas Flow Rate, cc/min at room temperature and 1 atm (SCCM)	98 - 110
Hydrogen, vol %	61 - 89
Moisture, vol %:	2.6 – 13.7
Concentration of H_2S, ppmv	2,300 – 9,200
Concentration of SO_2, ppmv	1,600 – 4,900
Nitrogen, vol %	Remainder

Table 2. Properties of the C-500-04 alumina catalyst from the Research Triangle Institute (RTI).

BET Area, m^2/g	227
Bulk Density, g/cm^3	0.8346
Pore Volume, cm^3/g	0.6211
Mean Particle Size, μm	160
Composition	Alumina

Table 3. Conversion of 4,400 -6,300 ppmv hydrogen sulfide with 3,300 - 4,800 ppmv sulfur dioxide in the presence of 61 - 89 v-% hydrogen, 5-v % moisture, and 0.1-g catalyst at 125 - 155 °C, 112 - 124 psia, and 0.040 - 0.046 s space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
114	155	119.7	0.0103	99.7	0.042	70.211	0.499	0.378	5.015	23.897	0.399
115	155	119.7	0.0103	100	0.042	70.000	0.498	0.377	5.000	24.126	0.433
116	155	119.7	0.0102	100	0.042	70.000	0.498	0.377	5.000	24.126	0.502
117	155	119.7	0.0105	100	0.043	70.000	0.498	0.377	5.000	24.126	0.356
118	155	119.7	0.0105	100	0.043	70.000	0.498	0.377	5.000	24.126	0.535
113	155	119.7	0.0103	99.7	0.042	70.211	0.499	0.378	5.015	23.897	0.301
119	155	118.7	0.0104	99.7	0.042	70.211	0.499	0.378	5.015	23.897	0.303*
199	155	120.7	0.0101	100	0.042	70.000	0.498	0.377	5.000	24.126	0.262*
51	150	112.7	0.0105	95.5	0.043	73.298	0.521	0.394	5.236	20.550	0.341
58	150	114.7	0.0103	99.5	0.041	70.352	0.501	0.378	5.025	23.744	0.387
64	150	117.7	0.0104	99	0.043	70.707	0.503	0.380	5.051	23.359	0.458
65	150	118.7	0.0102	100.5	0.042	69.652	0.496	0.375	4.975	24.503	0.474
73	150	120.7	0.0103	100.7	0.043	69.513	0.495	0.374	4.965	24.653	0.492
103	150	123.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.461
121	150	122.7	0.0104	99	0.044	70.707	0.503	0.380	5.051	23.359	0.496
122	150	121.7	0.0104	100.5	0.043	69.652	0.496	0.375	4.975	24.503	0.490
200	150	122.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.243
46	150	113.7	0.0103	99	0.041	70.707	0.503	0.380	5.051	23.359	0.417*
56	150	114.7	0.0104	99.5	0.041	70.352	0.501	0.378	5.025	23.744	0.438*
112	150	119.7	0.0102	100	0.042	70.000	0.498	0.377	5.000	24.126	0.443*
120	150	118.7	0.0101	100	0.041	70.000	0.498	0.377	5.000	24.126	0.416*
226	150	115.7	0.0101	100	0.040	70.000	0.498	0.377	5.000	24.126	0.434*
45	145	113.7	0.0103	97.7	0.042	71.648	0.510	0.385	5.118	22.339	0.175
52	145	117.7	0.0102	95.5	0.044	73.298	0.521	0.394	5.236	20.550	0.334
57	145	114.7	0.0103	101	0.041	69.307	0.493	0.373	4.950	24.877	0.309
66	145	116.7	0.0103	99	0.042	70.707	0.503	0.380	5.051	23.359	0.459
74	145	119.7	0.0103	96	0.045	72.917	0.519	0.392	5.208	20.964	0.483
109	145	120.7	0.0103	100	0.043	70.000	0.498	0.377	5.000	24.126	0.402
110	145	119.7	0.0103	100	0.043	70.000	0.498	0.377	5.000	24.126	0.397
129	145	119.7	0.0104	100	0.043	70.000	0.498	0.377	5.000	24.126	0.493
50	145	115.7	0.0104	98	0.043	71.429	0.508	0.384	5.102	22.577	0.289*
201	145	121.7	0.0103	100	0.044	70.000	0.498	0.377	5.000	24.126	0.234*
227	145	121.7	0.0101	100	0.043	70.000	0.498	0.377	5.000	24.126	0.258*
49	140	115.7	0.0103	102.5	0.041	68.293	0.486	0.367	4.878	25.976	0.341
59	140	118.7	0.0102	100.7	0.042	69.513	0.495	0.374	4.965	24.653	0.361
67	140	116.7	0.0104	99.5	0.043	70.352	0.501	0.378	5.025	23.744	0.415
76	140	118.7	0.0104	99.5	0.044	70.352	0.501	0.378	5.025	23.744	0.437
101	140	117.7	0.0104	100	0.043	70.000	0.498	0.377	5.000	24.126	0.457
102	140	121.7	0.0105	100	0.045	70.000	0.498	0.377	5.000	24.126	0.448
123	140	121.7	0.0102	99.7	0.044	70.211	0.499	0.378	5.015	23.897	0.467
124	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.461
40	140	111.7	0.0104	100.5	0.041	69.652	0.496	0.375	4.975	24.503	0.312*

*: data selected for figures

Table 3. Continued – 1

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
53	140	117.7	0.0104	101	0.043	69.307	0.493	0.373	4.950	24.877	0.318*
193	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.301*
198	140	121.7	0.0101	100	0.043	70.000	0.498	0.377	5.000	24.126	0.309*
222	140	118.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.286*
41	135	110.7	0.0103	101	0.040	69.307	0.493	0.373	4.950	24.877	0.406
54	135	116.7	0.0104	101.7	0.043	68.830	0.490	0.370	4.916	25.394	0.390
60	135	112.7	0.0104	91	0.046	76.923	0.547	0.414	5.495	16.621	0.413
61	135	118.7	0.0103	100.5	0.043	69.652	0.496	0.375	4.975	24.503	0.424
68	135	117.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.508
69	135	117.7	0.0103	79	0.055	88.608	0.630	0.477	6.329	3.956	0.442
70	135	119.7	0.0104	102	0.044	68.627	0.488	0.369	4.902	25.613	0.485
72	135	118.7	0.0103	99.5	0.044	70.352	0.501	0.378	5.025	23.744	0.421
75	135	116.7	0.0102	99	0.043	70.707	0.503	0.380	5.051	23.359	0.507
111	135	119.7	0.0102	100	0.044	70.000	0.498	0.377	5.000	24.126	0.373
128	135	120.7	0.0104	100	0.045	70.000	0.498	0.377	5.000	24.126	0.447
108	135	122.7	0.0104	100	0.046	70.000	0.498	0.377	5.000	24.126	0.353*
202	135	121.7	0.01	100	0.043	70.000	0.498	0.377	5.000	24.126	0.326*
43	130	110.7	0.0104	99	0.042	70.707	0.503	0.380	5.051	23.359	0.402
47	130	111.7	0.0104	113.9	0.037	61.463	0.437	0.331	4.390	33.379	0.218
55	130	119.7	0.0104	103.2	0.044	67.829	0.483	0.365	4.845	26.478	0.496
62	130	111.7	0.0103	92	0.045	76.087	0.541	0.409	5.435	17.528	0.459
63	130	111.7	0.01	100	0.040	70.000	0.498	0.377	5.000	24.126	0.447
71	130	118.7	0.0104	100	0.045	70.000	0.498	0.377	5.000	24.126	0.461
104	130	120.7	0.0103	100	0.045	70.000	0.498	0.377	5.000	24.126	0.487
105	130	118.7	0.0104	99.7	0.045	70.211	0.499	0.378	5.015	23.897	0.410
125	130	120.7	0.0104	99.5	0.046	70.352	0.501	0.378	5.025	23.744	0.481
126	130	120.7	0.0105	99.7	0.046	70.211	0.499	0.378	5.015	23.897	0.469
48	130	117.7	0.0101	98.7	0.044	70.922	0.505	0.381	5.066	23.126	0.343*
203	130	120.7	0.0101	100	0.044	70.000	0.498	0.377	5.000	24.126	0.333*
42	125	112.7	0.0102	101	0.042	69.307	0.493	0.373	4.950	24.877	0.364
44	125	110.7	0.0102	98	0.042	71.429	0.508	0.384	5.102	22.577	0.502
106	125	118.7	0.0103	99.7	0.045	70.211	0.499	0.378	5.015	23.897	0.340
127	125	120.7	0.0104	100.5	0.046	69.652	0.496	0.375	4.975	24.503	0.432
107	125	122.7	0.0102	99.7	0.046	70.211	0.499	0.378	5.015	23.897	0.285*
204	125	121.7	0.0103	100	0.046	70.000	0.498	0.377	5.000	24.126	0.265*

*: data selected for figures

Table 4. Conversion of 4,400 – 5,200 ppmv hydrogen sulfide with 3,400 – 3,900 ppmv sulfur dioxide in the presence of 63 - 73 v-% hydrogen, 2.6 – 14.2 v-% moisture, and 0.1-g catalyst at 140 °C, 112 - 123 psia, and 0.039 - 0.046 s space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
78	140	121.7	0.0104	111.7	0.040	62.668	0.446	0.337	13.429	23.120	0.409
81	140	120.7	0.0103	106	0.041	66.038	0.470	0.355	14.151	18.986	0.506
132	140	120.7	0.0101	110	0.039	63.636	0.453	0.342	13.636	21.932	0.485
133	140	122.7	0.0102	110	0.040	63.636	0.453	0.342	13.636	21.932	0.436
194	140	121.7	0.0104	110	0.041	63.636	0.453	0.342	13.636	21.932	0.322*
77	140	115.7	0.0104	104	0.041	67.308	0.479	0.362	9.615	22.236	0.464
82	140	119.7	0.0101	104	0.041	67.308	0.479	0.362	9.615	22.236	0.506
131	140	120.7	0.0105	105	0.043	66.667	0.474	0.359	9.524	22.977	0.397
195	140	121.7	0.0103	105	0.042	66.667	0.474	0.359	9.524	22.977	0.317*
134	140	119.7	0.0104	102.5	0.043	68.293	0.486	0.367	7.317	23.537	0.443
196	140	122.2	0.0105	102.5	0.044	68.293	0.486	0.367	7.317	23.537	0.254*
49	140	115.7	0.0103	102.5	0.041	68.293	0.486	0.367	4.878	25.976	0.341
59	140	118.7	0.0102	100.7	0.042	69.513	0.495	0.374	4.965	24.653	0.361
67	140	116.7	0.0104	99.5	0.043	70.352	0.501	0.378	5.025	23.744	0.415
76	140	118.7	0.0104	99.5	0.044	70.352	0.501	0.378	5.025	23.744	0.437
101	140	117.7	0.0104	100	0.043	70.000	0.498	0.377	5.000	24.126	0.457
102	140	121.7	0.0105	100	0.045	70.000	0.498	0.377	5.000	24.126	0.448
123	140	121.7	0.0102	99.7	0.044	70.211	0.499	0.378	5.015	23.897	0.467
124	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.461
40	140	111.7	0.0104	100.5	0.041	69.652	0.496	0.375	4.975	24.503	0.312*
53	140	117.7	0.0104	101	0.043	69.307	0.493	0.373	4.950	24.877	0.318*
193	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.301*
198	140	121.7	0.0101	100	0.043	70.000	0.498	0.377	5.000	24.126	0.309*
222	140	118.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.286*
83	140	122.7	0.0103	96.5	0.046	72.539	0.516	0.390	2.591	23.964	0.467
130	140	120.7	0.0103	97.5	0.045	71.795	0.511	0.386	2.564	24.744	0.471
80	140	120.7	0.0104	97.2	0.046	72.016	0.512	0.387	2.572	24.512	0.339*
197	140	122.7	0.0103	97.5	0.046	71.795	0.511	0.386	2.564	24.744	0.325*

*: data selected for figures

Table 5. Conversion of 4,900 – 5,300 ppmv hydrogen sulfide with 3,700 – 4,900 ppmv sulfur dioxide in the presence of 68 - 74 v-% hydrogen, 2.6 – 13.7 moisture, and 0.1-g catalyst at 140 ^oC, 28 - 122 psia, and 0.010 - 0.045 s space time.

Run Number	Temperature, ^oC	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
49	140	115.7	0.0103	102.5	0.041	68.293	0.486	0.367	4.878	25.976	0.341
59	140	118.7	0.0102	100.7	0.042	69.513	0.495	0.374	4.965	24.653	0.361
67	140	116.7	0.0104	99.5	0.043	70.352	0.501	0.378	5.025	23.744	0.415
76	140	118.7	0.0104	99.5	0.044	70.352	0.501	0.378	5.025	23.744	0.437
101	140	117.7	0.0104	100	0.043	70.000	0.498	0.377	5.000	24.126	0.457
102	140	121.7	0.0105	100	0.045	70.000	0.498	0.377	5.000	24.126	0.448
123	140	121.7	0.0102	99.7	0.044	70.211	0.499	0.378	5.015	23.897	0.467
124	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.461
40	140	111.7	0.0104	100.5	0.041	69.652	0.496	0.375	4.975	24.503	0.312*
53	140	117.7	0.0104	101	0.043	69.307	0.493	0.373	4.950	24.877	0.318*
193	140	119.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.301*
198	140	121.7	0.0101	100	0.043	70.000	0.498	0.377	5.000	24.126	0.309*
222	140	118.7	0.0104	100	0.044	70.000	0.498	0.377	5.000	24.126	0.286*
97	140	87.7	0.0105	100.6	0.032	69.583	0.495	0.374	4.970	24.578	0.300
98	140	88.7	0.0102	100.6	0.032	69.583	0.495	0.374	4.970	24.578	0.286
135	140	91.7	0.0103	100	0.033	70.000	0.498	0.377	5.000	24.126	0.342
205	140	91.7	0.0101	100	0.033	70.000	0.498	0.377	5.000	24.126	0.208*
223	140	92.2	0.0101	100	0.033	70.000	0.498	0.377	5.000	24.126	0.232*
84	140	67.7	0.0102	102	0.024	68.627	0.488	0.369	4.902	25.613	0.238
85	140	65.7	0.0102	102	0.023	68.627	0.488	0.369	4.902	25.613	0.223
87	140	66.7	0.0102	94	0.026	74.468	0.530	0.401	5.319	19.282	0.106
88	140	67.7	0.0103	101	0.024	69.307	0.493	0.373	4.950	24.877	0.234
86	140	67.7	0.0102	99	0.025	70.707	0.503	0.380	5.051	23.359	0.183*
89	140	67.7	0.0102	99	0.025	70.707	0.503	0.380	5.051	23.359	0.207*
136	140	69.7	0.0103	100	0.025	70.000	0.498	0.377	5.000	24.126	0.182*
206	140	69.7	0.0105	100	0.026	70.000	0.498	0.377	5.000	24.126	0.180*
224	140	71.7	0.0104	100	0.026	70.000	0.498	0.377	5.000	24.126	0.192*
94	140	37.7	0.0103	99.7	0.014	70.211	0.499	0.378	5.015	23.897	0.150
95	140	36.7	0.0104	98	0.014	71.429	0.508	0.384	5.102	22.577	0.166
137	140	41.7	0.0103	100	0.015	70.000	0.498	0.377	5.000	24.126	0.151
207	140	41.7	0.0103	100	0.015	70.000	0.498	0.377	5.000	24.126	0.133*
225	140	39.7	0.0102	100	0.014	70.000	0.498	0.377	5.000	24.126	0.107*
90	140	28.7	0.0102	99	0.010	70.707	0.503	0.380	5.051	23.359	0.104*
91	140	27.7	0.0105	99	0.010	70.707	0.503	0.380	5.051	23.359	0.124*

*: data selected for figures

Table 6. Conversion of 3,200 – 3,600 ppmv hydrogen sulfide with 4,100 – 4,600 ppmv sulfur dioxide in the presence of 64 - 72 v-% hydrogen, 2.6 – 13.7 v-% moisture, and 0.1-g catalyst at 125 - 155 ^oC, 43 - 123 psia, and 0.016 - 0.046 s space time.

Run Number	Temperature, ^oC	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
168	155	120.7	0.0104	100	0.043	70.000	0.349	0.452	5.000	24.200	0.525*
165	150	119.7	0.0102	100	0.042	70.000	0.349	0.452	5.000	24.200	0.402*
164	140	121.7	0.0103	100	0.044	70.000	0.349	0.452	5.000	24.200	0.333*
166	130	120.7	0.0104	100	0.045	70.000	0.349	0.452	5.000	24.200	0.428*
167	125	120.7	0.0104	100	0.046	70.000	0.349	0.452	5.000	24.200	0.428*
178	140	92.7	0.0103	100	0.034	70.000	0.349	0.452	5.000	24.200	0.049
164	140	121.7	0.0103	100	0.044	70.000	0.349	0.452	5.000	24.200	0.333*
181	140	89.7	0.0102	100	0.032	70.000	0.349	0.452	5.000	24.200	0.215*
179	140	71.7	0.0102	100	0.026	70.000	0.349	0.452	5.000	24.200	0.193*
180	140	42.7	0.0103	100	0.016	70.000	0.349	0.452	5.000	24.200	0.136*
182	140	121.7	0.0103	110	0.040	63.636	0.317	0.411	13.636	22.000	0.415*
183	140	119.7	0.0101	105	0.041	66.667	0.332	0.430	9.524	23.047	0.351*
184	140	122.7	0.0104	102.5	0.044	68.293	0.340	0.441	7.317	23.609	0.425*
164	140	121.7	0.0103	100	0.044	70.000	0.349	0.452	5.000	24.200	0.333*
185	140	122.7	0.0101	97.5	0.045	71.795	0.358	0.463	2.564	24.820	0.347*

*: data selected for figures

Table 7. Conversion of 6,800 – 7,600 ppmv hydrogen sulfide with 2,300 – 2,600 ppmv sulfur dioxide in the presence of 64 - 72 v-% hydrogen, 2.6 – 13.7 v-% moisture, and 0.1-g catalyst at 125 - 155 ^oC, 38 - 125 psia, and 0.014 - 0.046 s space time.

Run Number	Temperature, ^oC	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
212	155	124.7	0.0105	100	0.045	70.000	0.747	0.251	5.000	24.002	0.047*
213	150	121.7	0.0104	100	0.044	70.000	0.747	0.251	5.000	24.002	0.142*
150	145	119.7	0.0103	100	0.043	70.000	0.747	0.251	5.000	24.002	0.324
214	145	122.7	0.0103	100	0.044	70.000	0.747	0.251	5.000	24.002	0.163*
142	140	121.7	0.0103	100	0.044	70.000	0.747	0.251	5.000	24.002	0.427
146	140	122.7	0.0104	100	0.045	70.000	0.747	0.251	5.000	24.002	0.378
149	140	120.7	0.0104	100	0.044	70.000	0.747	0.251	5.000	24.002	0.311
211	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.142
141	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.219*
218	140	122.7	0.0103	100	0.045	70.000	0.747	0.251	5.000	24.002	0.242*
151	135	119.7	0.0102	100	0.044	70.000	0.747	0.251	5.000	24.002	0.124
215	135	119.7	0.0105	100	0.045	70.000	0.747	0.251	5.000	24.002	0.230*
152	130	119.7	0.0105	100	0.045	70.000	0.747	0.251	5.000	24.002	0.336
216	130	119.7	0.0103	100	0.045	70.000	0.747	0.251	5.000	24.002	0.243*
153	125	121.7	0.0103	100	0.046	70.000	0.747	0.251	5.000	24.002	0.293*
217	125	121.7	0.0104	100	0.046	70.000	0.747	0.251	5.000	24.002	0.328*

Table 7. Continued – 1

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
142	140	121.7	0.0103	100	0.044	70.000	0.747	0.251	5.000	24.002	0.427
146	140	122.7	0.0104	100	0.045	70.000	0.747	0.251	5.000	24.002	0.378
149	140	120.7	0.0104	100	0.044	70.000	0.747	0.251	5.000	24.002	0.311
211	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.142
141	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.219*
218	140	122.7	0.0103	100	0.045	70.000	0.747	0.251	5.000	24.002	0.242*
100	140	88.7	0.0104	102	0.032	68.627	0.732	0.246	4.902	25.492	0.256
99	140	88.7	0.0104	100	0.033	70.000	0.747	0.251	5.000	24.002	0.232
140	140	91.7	0.0103	100	0.033	70.000	0.747	0.251	5.000	24.002	0.216
143	140	89.7	0.0105	100	0.033	70.000	0.747	0.251	5.000	24.002	0.315
210	140	90.7	0.0103	100	0.033	70.000	0.747	0.251	5.000	24.002	0.108*
139	140	72.7	0.0105	100	0.027	70.000	0.747	0.251	5.000	24.002	0.223
144	140	71.7	0.0105	100	0.027	70.000	0.747	0.251	5.000	24.002	0.190
209	140	70.7	0.0103	100	0.026	70.000	0.747	0.251	5.000	24.002	0.062*
93	140	38.7	0.0104	99.7	0.014	70.211	0.749	0.252	5.015	23.773	0.106*
96	140	37.7	0.0105	99	0.014	70.707	0.755	0.254	5.051	23.234	0.118*
138	140	41.7	0.0103	100	0.015	70.000	0.747	0.251	5.000	24.002	0.094*
145	140	41.7	0.0104	100	0.015	70.000	0.747	0.251	5.000	24.002	0.102*
208	140	42.7	0.0103	100	0.016	70.000	0.747	0.251	5.000	24.002	0.098*
187	140	120.7	0.0101	110	0.039	63.636	0.679	0.228	13.636	21.820	0.231*
190	140	120.7	0.0103	105	0.042	66.667	0.711	0.239	9.524	22.859	0.132
148	140	120.7	0.0104	105	0.042	66.667	0.711	0.239	9.524	22.859	0.202*
188	140	119.7	0.0102	105	0.041	66.667	0.711	0.239	9.524	22.859	0.187*
219	140	120.7	0.0102	105	0.041	66.667	0.711	0.239	9.524	22.859	0.189*
189	140	120.7	0.0101	102.5	0.042	68.293	0.729	0.245	7.317	23.417	0.140
191	140	120.7	0.0104	102.5	0.043	68.293	0.729	0.245	7.317	23.417	0.143
147	140	122.7	0.0101	102.5	0.043	68.293	0.729	0.245	7.317	23.417	0.267*
220	140	119.2	0.0103	102.5	0.042	68.293	0.729	0.245	7.317	23.417	0.268*
142	140	121.7	0.0103	100	0.044	70.000	0.747	0.251	5.000	24.002	0.427
146	140	122.7	0.0104	100	0.045	70.000	0.747	0.251	5.000	24.002	0.378
149	140	120.7	0.0104	100	0.044	70.000	0.747	0.251	5.000	24.002	0.311
211	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.142
141	140	119.7	0.0102	100	0.043	70.000	0.747	0.251	5.000	24.002	0.219*
218	140	122.7	0.0103	100	0.045	70.000	0.747	0.251	5.000	24.002	0.242*
192	140	119.7	0.0102	97.5	0.044	71.795	0.766	0.257	2.564	24.617	0.200*
221	140	121.7	0.0103	97.5	0.045	71.795	0.766	0.257	2.564	24.617	0.253*

*: data selected for figures

Table 8. Conversion of 8,200 – 9,200 ppmv hydrogen sulfide with 1,600 – 1,800 ppmv sulfur dioxide in the presence of 64 - 72 v-% hydrogen, 2.6 – 13.7 v-% moisture, and 0.1-g catalyst at 130 - 155 °C, 43 - 127 psia, and 0.015 - 0.047 s space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v% H_2	H_2S	SO_2	moisture	N_2	Conversion
174	140	121.7	0.0102	110	0.040	63.636	0.815	0.160	13.636	21.753	0.057*
175	140	121.7	0.0103	105	0.042	66.667	0.854	0.167	9.524	22.788	0.123*
176	140	120.7	0.0103	102.5	0.043	68.293	0.875	0.171	7.317	23.344	0.162*
154	140	126.7	0.0103	100	0.046	70.000	0.896	0.176	5.000	23.928	0.047
155	140	119.7	0.0102	100	0.043	70.000	0.896	0.176	5.000	23.928	0.086*
161	140	119.7	0.0103	100	0.044	70.000	0.896	0.176	5.000	23.928	0.258*
177	140	122.7	0.0105	97.5	0.047	71.795	0.919	0.180	2.564	24.541	0.136*
186	140	120.7	0.0101	97.5	0.044	71.795	0.919	0.180	2.564	24.541	0.132*
154	140	126.7	0.0103	100	0.046	70.000	0.896	0.176	5.000	23.928	0.047
155	140	119.7	0.0102	100	0.043	70.000	0.896	0.176	5.000	23.928	0.086*
161	140	119.7	0.0103	100	0.044	70.000	0.896	0.176	5.000	23.928	0.258*
171	140	91.7	0.0103	100	0.033	70.000	0.896	0.176	5.000	23.928	0.103*
172	140	70.7	0.0104	100	0.026	70.000	0.896	0.176	5.000	23.928	0.096*
173	140	42.7	0.0102	100	0.015	70.000	0.896	0.176	5.000	23.928	0.044*
169	155	121.7	0.0104	100	0.043	70.000	0.896	0.176	5.000	23.928	0.179*
162	150	120.7	0.0105	100	0.044	70.000	0.896	0.176	5.000	23.928	0.147*
163	145	120.7	0.0103	100	0.043	70.000	0.896	0.176	5.000	23.928	0.061
159	145	118.7	0.0102	100	0.042	70.000	0.896	0.176	5.000	23.928	0.209*
160	145	121.7	0.0102	100	0.043	70.000	0.896	0.176	5.000	23.928	0.132*
154	140	126.7	0.0103	100	0.046	70.000	0.896	0.176	5.000	23.928	0.047
155	140	119.7	0.0102	100	0.043	70.000	0.896	0.176	5.000	23.928	0.086
161	140	119.7	0.0103	100	0.044	70.000	0.896	0.176	5.000	23.928	0.258
156	130	121.7	0.0105	100	0.046	70.000	0.896	0.176	5.000	23.928	0.205*
157	130	121.7	0.0103	100	0.045	70.000	0.896	0.176	5.000	23.928	0.189*
158	135	119.7	0.0104	100	0.044	70.000	0.896	0.176	5.000	23.928	0.271*
170	135	121.7	0.0104	100	0.045	70.000	0.896	0.176	5.000	23.928	0.134*

*: data selected for figures

CALCULATIONS

Gaseous samples having a volume of 4-cm^3 volume, obtained from the outlet stream of the differential reactor, are injected into a gas chromatograph to analyze gas chromatography (GC) areas of gaseous samples. Conversions of H_2S are obtained by dividing the GC area of H_2S from a reaction run with that from its blank run.

$$x = \frac{(A_B - A_R)}{A_B} \tag{1}$$

where x: conversion of H_2S.

A_B: GC area of H_2S of the 4-cc gaseous sample of a blank run.

A_R: GC area of H_2S of the 4-cc gaseous sample of a reaction run.

$$2H_2S\ (g) + SO_2\ (g) \leftrightarrow 3S\ (\ell) + 2H_2S\ (g) \tag{2}$$

Surface reaction rates of conversion of H_2S into elemental sulfur in a differential reactor are obtained with amounts of the alumina catalyst loaded in the differential reactor, molar feed rates of H_2S to the differential reactor and conversion of H_2S, as shown in the following equation.

$$-r_A^{'} = \frac{F_{Ao}x}{W} \tag{3}$$

where $-r_A$': surface reaction rates

F_{Ao}: molar flow rates of H_2S in a feed stream to a reactor

x: conversion of H_2S into elemental sulfur

RESULTS AND DISCUSSION

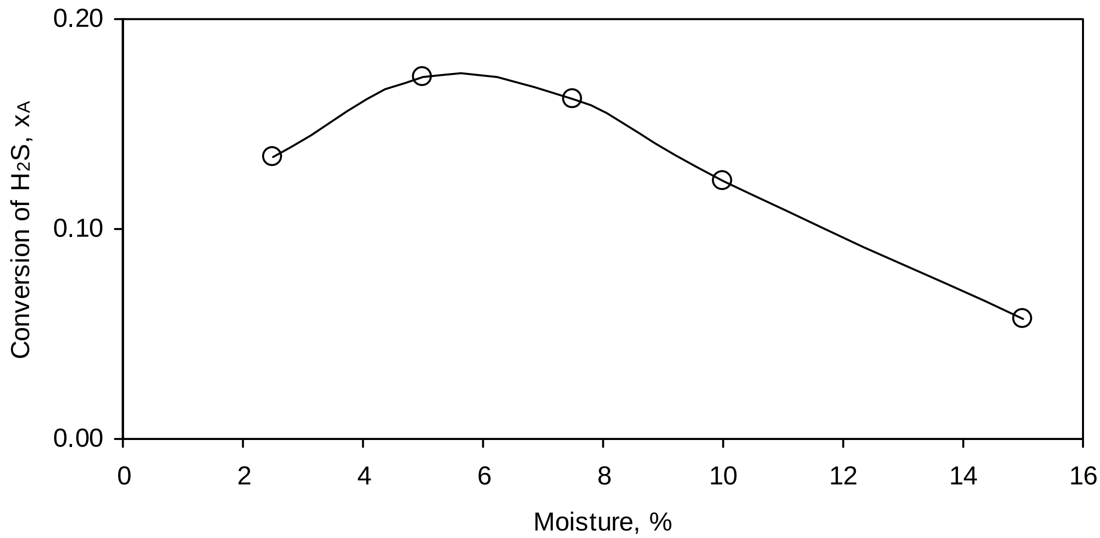
Experiments on conversion of hydrogen sulfide into element sulfur were carried out for the space time range of 0.01 – 0.047 seconds at 125 - 155°C (see Table 3) to evaluate effects of catalyst amounts, moisture concentrations, reaction pressures on conversion of hydrogen sulfide into elemental sulfur. Simulated coal gas mixtures consist of 61 – 89 v% hydrogen, 2,300 - 9,200-ppmv hydrogen sulfide, 1,600 - 4,900 ppmv sulfur dioxide, and 2.6 – 13.7 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the reactor are 100 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). The temperature of the reactor is controlled in an oven at 125 - 155°C. The pressure of the reactor is maintained at 28 - 127 psia.

Effects of Moisture on Conversion of H_2S into Elemental Sulfur

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.040 – 0.047 seconds (see Table 8) to evaluate effects of moisture concentrations on conversion of hydrogen sulfide into elemental sulfur at 140°C and 120 – 123 psia. Gas mixtures fed to the reactor contain 64 – 72 v% hydrogen, 8,200 - 9,200-ppmv H_2S, 1,600- 1,800 ppmv SO_2, 2.5 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.06 – 0.17.

Concentrations of moisture in the presence of 8,200 – 9200 ppmv H_2S, 1,600 - 1,800 ppmv SO_2, and 64 – 72 v-% hydrogen appear to affect significantly conversion of H_2S into elemental sulfur in the moisture range of 5 – 13.6 v% in a simulated coal gas mixture at 140 ^oC and 120 – 123 psia. Conversion of H_2S decreases with increased concentration of moisture over the moisture range of 5 – 13.6 v%, as shown in Figure 3.

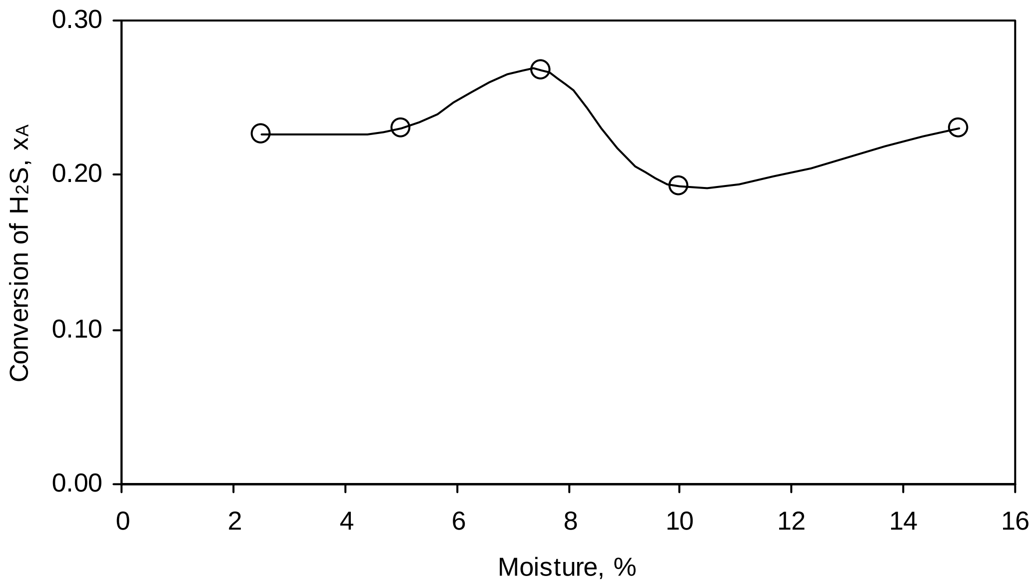
Figure 3. Effects of moisture on conversion of H_2S with 0.01-g catalyst and a 98 - 110 SCCM feed stream containing 9,000-ppmv H_2S, 1,800-ppmv SO_2, and 70-v % H_2 at 120 psia and 140^oC.



Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.040 – 0.047 seconds (see Table 7) to evaluate effects of moisture concentrations on conversion of hydrogen sulfide into elemental sulfur at 140^oC and 119 – 123 psia. Gas mixtures fed to the reactor contain 64 – 77 v% hydrogen, 6,800 - 7,700 ppmv H_2S, 2,300 – 2,600 ppmv SO_2, 2.6 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.19 – 0.27. Concentrations of moisture in the presence of 6,800 - 7,700 ppmv H_2S and 2,300 – 2,600 ppmv SO_2 appear to affect slightly reaction rates for the conversion of H_2S into elemental sulfur in the moisture range of 2.6 – 13.6 v% in a simulated coal gas mixture at 119 – 123 psia (see Figure 4). However, conversion of H_2S is highest at 7.3-v % moisture.

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.040 – 0.045 seconds (see Table 6) to evaluate effects of moisture concentrations on conversion of hydrogen sulfide into elemental sulfur at 140^oC and 120 – 123 psia. Gas mixtures fed to the reactor contain 64 – 72 v% hydrogen, 3,200 - 3,600-ppmv H_2S, 4,100 - 4,600 ppmv SO_2, 2.5 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.33 – 0.43. Concentrations of moisture in the presence of 3,200 - 3,600 ppmv H_2S and 4,100 - 4,600 ppmv SO_2 appear to affect slightly conversion of H_2S into elemental sulfur in the moisture range of 2.5 – 13.6 v% in a simulated coal gas mixture at 120 – 123 psia (see Figure 5). Conversion of H_2S slightly increases with moisture concentration. However, conversion of H_2S is highest at 7.3-v % moisture.

Figure 4. Effects of moisture on conversion of H_2S with 0.01-g catalyst and a 98 - 110 SCCM feed stream containing 7,500-ppmv H_2S, 2,500-ppmv SO_2, and 70-v % H_2 at 120 psia and 140°C.



Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.041 – 0.046 seconds (see Table 4) to evaluate effects of moisture concentrations on conversion of hydrogen sulfide into elemental sulfur at 140°C and 112 – 123 psia. Gas mixtures fed to the reactor contain 64 – 72 v% hydrogen, 4,500 - 5,100 ppmv H_2S, 3,400 – 3,900 ppmv SO_2, and 2.6 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.25 – 0.34. Concentrations of moisture in the presence of 4,500 - 5,100 ppmv H_2S and 3,400 – 3,900 ppmv SO_2 appear to affect slightly conversion of H_2S into elemental sulfur in the moisture range of 2.6 – 13.6 v% in a simulated coal gas mixture at 112 – 123 psia (see Figure 6). Conversion of H_2S is not affected significantly by moisture concentration. However, conversion of H_2S is lowest at 7.3-v % moisture.

Figure 5. Effects of moisture on conversion of H_2S with 0.01-g catalyst and a 98 - 110 SCCM feed stream containing 3,500-ppmv H_2S, 4,500-ppmv SO_2, and 70-v % H_2 at 120 psia and 140^oC.

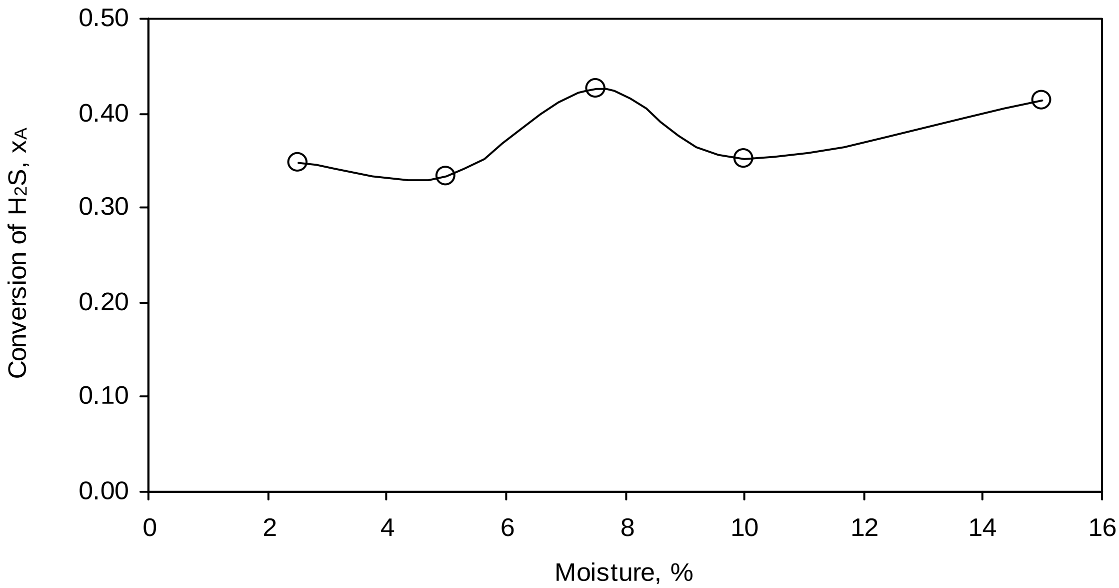
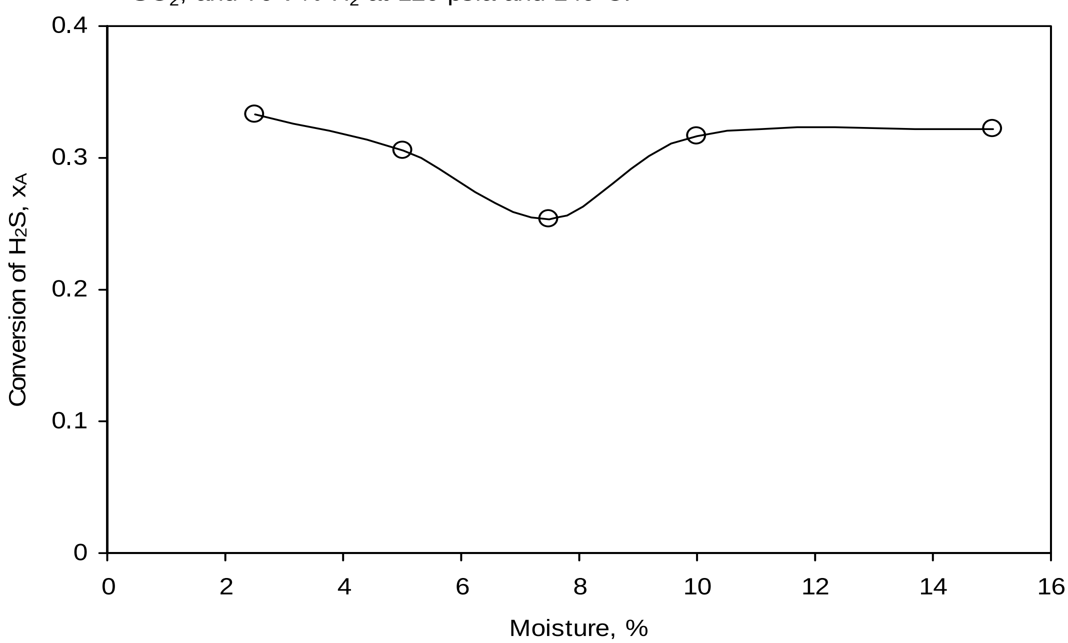


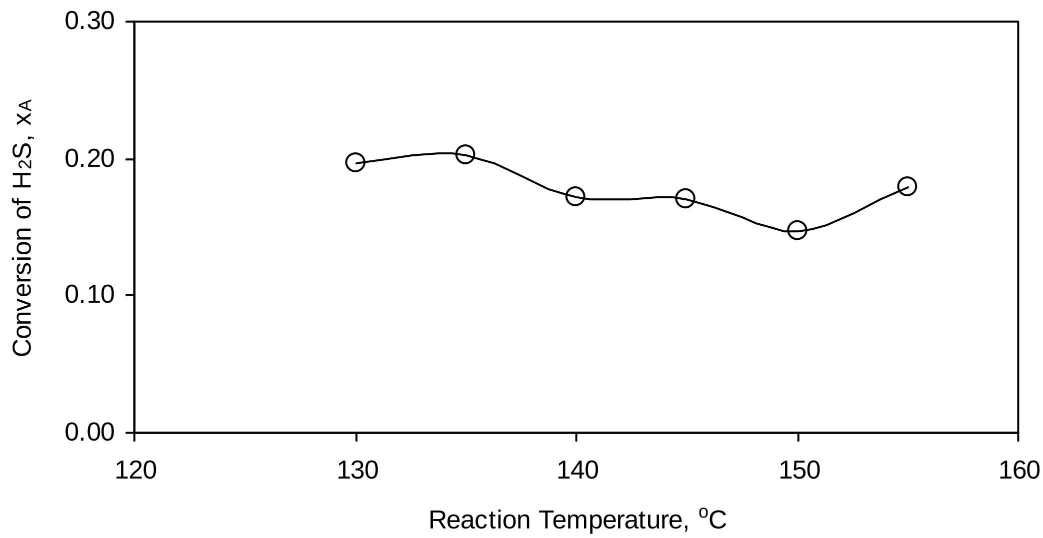
Figure 6. Effects of moisture on conversion of H_2S with 0.01-g catalyst and a 98 - 110 SCCM feed stream containing 5,000-ppmv H_2S, 3,800-ppmv SO_2, and 70-v % H_2 at 120 psia and 140^oC.



Effects of Reaction Temperature on Conversion of H_2S into Elemental Sulfur

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.043 – 0.046 seconds (see Table 8) to evaluate effects of reaction temperature on conversion of hydrogen sulfide into elemental sulfur at 130 - 155°C and 119 – 122 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 9,000-ppmv H_2S, 1,800-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the reactor are 100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.16 – 0.20. Reaction temperatures affect slightly conversion of H_2S into elemental sulfur. Conversion of H_2S into elemental sulfur decreases with increased reaction temperature over the temperature range of 130 – 150°C (see Figure 7).

Figure 7. Effects of reaction temperature on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 9,000-ppmv H_2S, 1,800-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 120 psia.



Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.043 – 0.046 seconds (see Table 7) to evaluate effects of reaction temperature on conversion of hydrogen sulfide into elemental sulfur at 125 - 155°C and 120 – 125 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 7,500-ppmv H_2S, 2,500-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the reactor are 100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.05 – 0.29. Reaction temperatures affect significantly conversion of H_2S into elemental sulfur. Conversion of H_2S into elemental sulfur decreases with increased reaction temperature (see Figure 8), as opposed to the Arrhenius' equation.

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.042 – 0.046 seconds (see Table 6) to evaluate effects of reaction temperature on conversion of hydrogen sulfide into elemental sulfur at 125 - 155°C and 120 – 122 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500-ppmv

H_2S, 4,500-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the reactor are 100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.33 – 0.53. Reaction temperatures affect slightly conversion of H_2S into elemental sulfur. However, conversion of H_2S into elemental sulfur is lowest at 140°C (see Figure 9).

Figure 8. Effects of reaction temperature on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 7,500-ppmv H_2S, 2,500-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 120 psia.

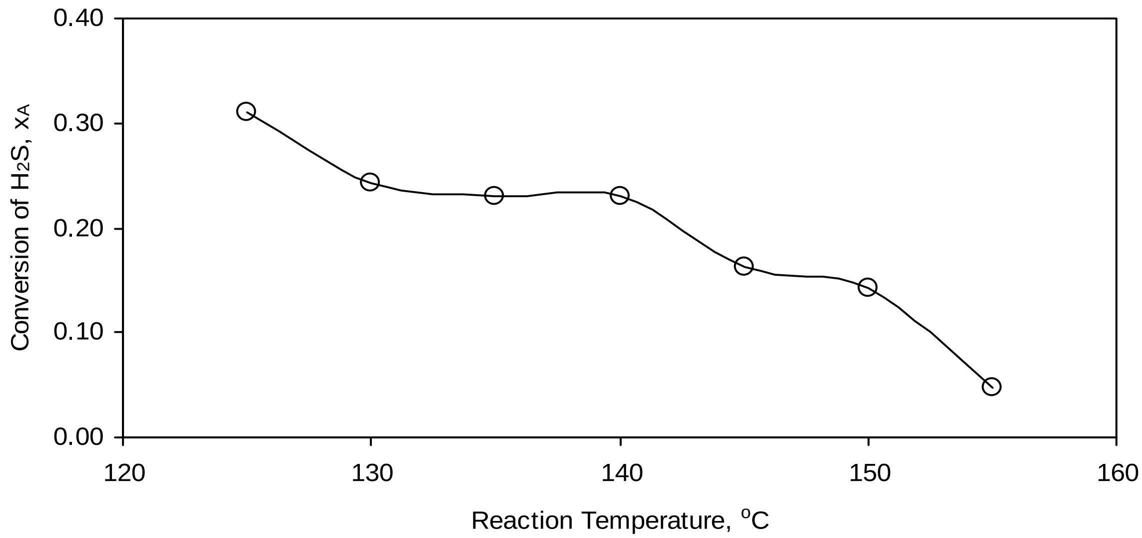


Figure 9. Effects of reaction temperature on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 3,500-ppmv H_2S, 4,500-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 120 psia.

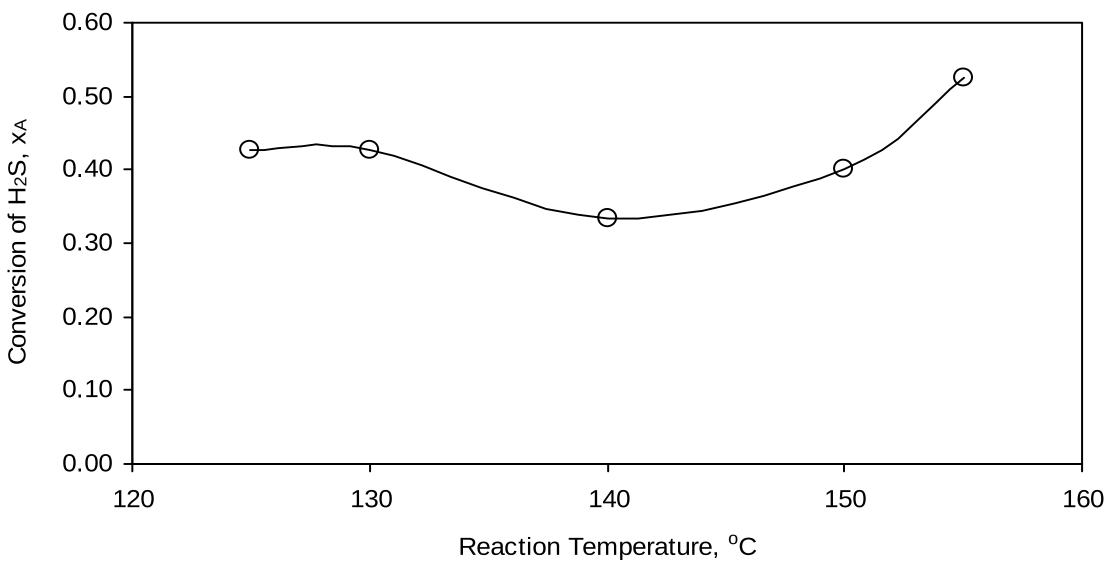
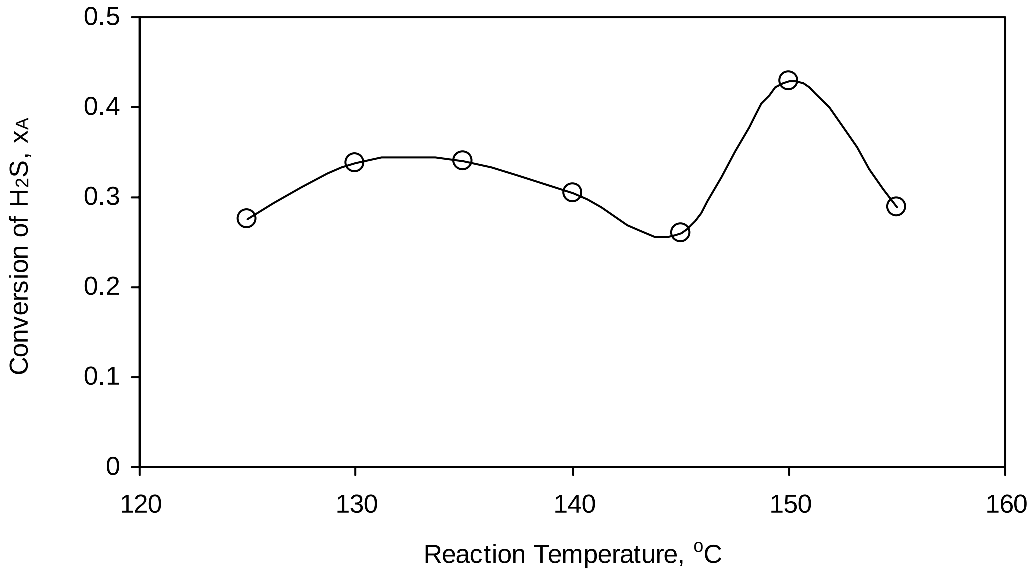


Figure 10. Effects of reaction temperature on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 5,000-ppmv H_2S, 3,800-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 120 psia.

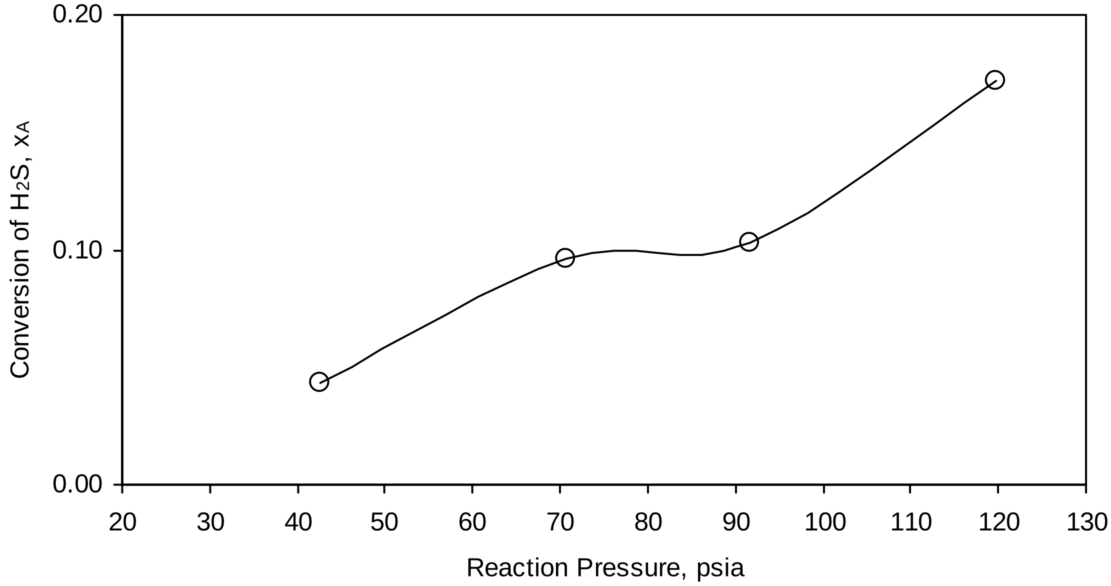


Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.041 – 0.046 seconds (see Table 3) to evaluate effects of reaction temperature on conversion of hydrogen sulfide into elemental sulfur at 125 - 155^{o}C and 112 – 123 psia. Gas mixtures fed to the reactor contain 69 – 71 v% hydrogen, 4,900 - 5,100-ppmv H_2S, 3,700 - 3,800 ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 99 - 101 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.27 – 0.44. Reaction temperatures affect slightly conversion of H_2S into elemental sulfur. However, conversion of H_2S into elemental sulfur is highest at 150^{o}C (see Figure 10).

Effects of Pressure on Conversion of H_2S into Elemental Sulfur

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.015 – 0.044 seconds (see Table 8) to evaluate effects of reaction pressures on conversion of hydrogen sulfide into elemental sulfur at 140^{o}C and 43 -123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 9,000-ppmv H_2S, 1,800-ppmv SO_2, and 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.04 – 0.17. Reaction pressures affect significantly conversion of H_2S into elemental sulfur in the pressure range of 43 -123 psia. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 43 -123 psia (see Figure 11).

Figure 11. Effects of reaction pressure on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 9,000-ppmv H_2S, 1,800-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 140°C.



Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.014 – 0.045 seconds (see Table 7) to evaluate effects of reaction pressures on conversion of hydrogen sulfide into elemental sulfur at 140°C and 38 -123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 7,500 - 7,600 ppmv H_2S, 2,500-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 99 -100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.06 – 0.24. Reaction pressures affect significantly conversion of H_2S into elemental sulfur in the pressure range of 71 -123 psia. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 71 -123 psia (see Figure 12). However, conversion of H_2S is lowest at 71 psia.

Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.015 – 0.044 seconds (see Table 6) to evaluate effects of reaction pressures on conversion of hydrogen sulfide into elemental sulfur at 140°C and 43 -122 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500-ppmv H_2S, 4,500-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 100 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.14 – 0.33. Reaction pressures affect significantly conversion of H_2S into elemental sulfur in the pressure range of 43 -122 psia. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 43 -122 psia (see Figure 13).

Figure 12. Effects of reaction pressure on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 7,500-ppmv-ppm H_2S, 2,500-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 140°C.

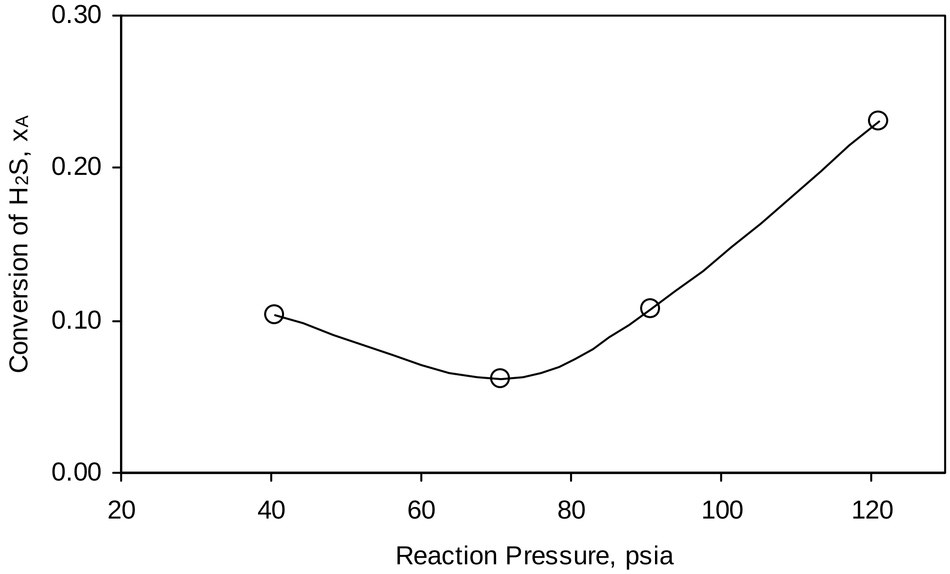
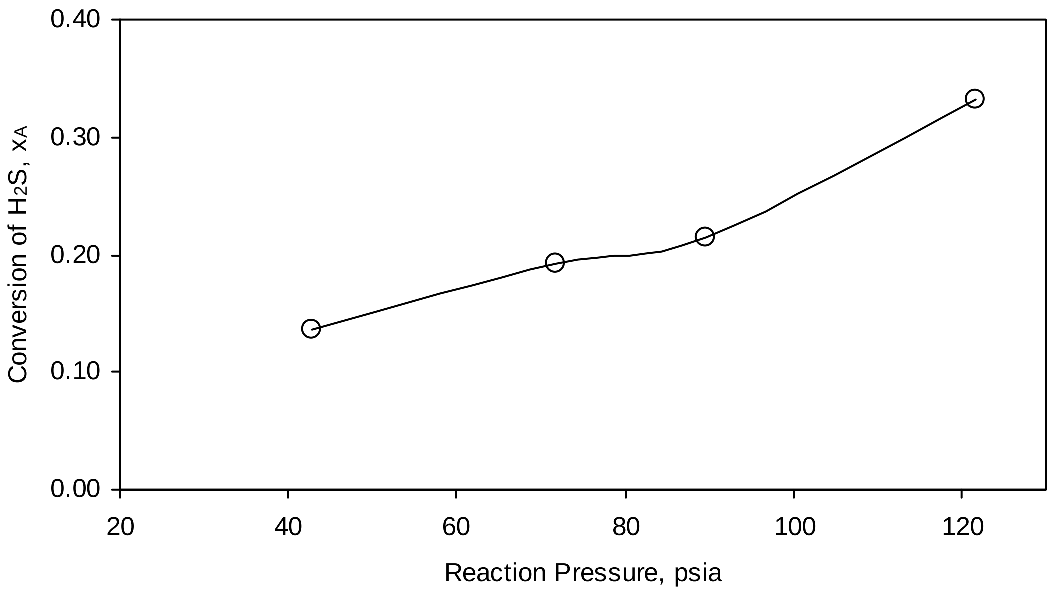
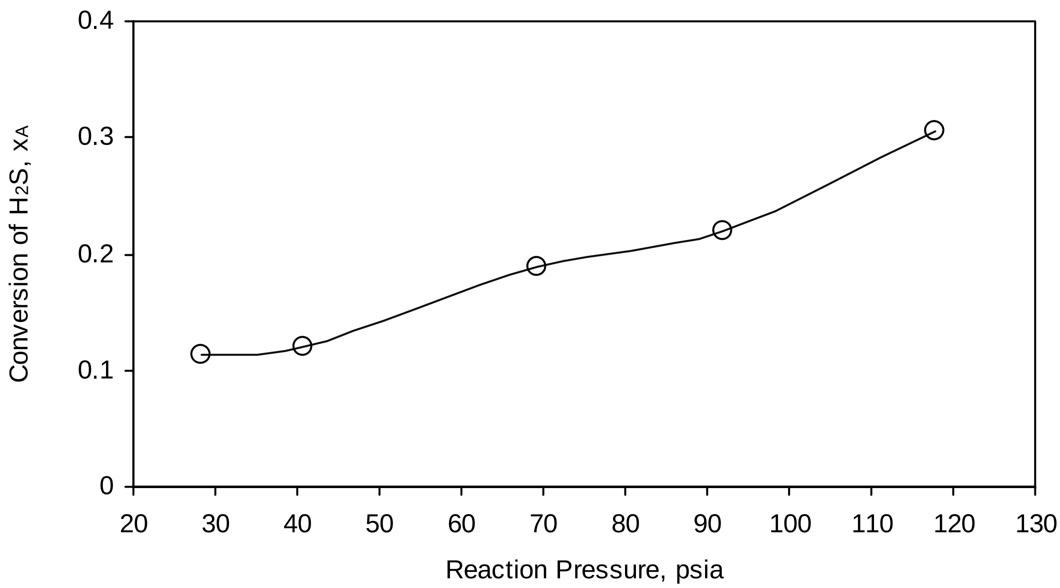


Figure 13. Effects of reaction pressure on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 3,500-ppmv H_2S, 4,500-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 140°C.



Experiments on conversion of hydrogen sulfide into element sulfur with 0.01-g catalyst were carried out for the space time range of 0.010 – 0.044 seconds (see Table 5) to evaluate effects of reaction pressures on conversion of hydrogen sulfide into elemental sulfur at 140^{o}C and 28 -122 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 5,000-ppmv H_2S, 3,800-ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 99 - 101 cm^3/min at room temperature and atmospheric pressure (SCCM). Conversions of H_2S into elemental sulfur are 0.10 – 0.32. Reaction pressures affect significantly conversion of H_2S into elemental sulfur in the pressure range of 28 -122 psia. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 28 -122 psia (see Figure 14).

Figure 14. Effects of reaction pressure on conversion of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 5,000-ppmv H_2S, 3,800-ppmv SO_2, 5-v% moisture, and 70-v % H_2 at 140^{o}C.



Effects of H_2S Concentrations on Reaction Rates of H_2S with SO_2

Reaction rates of hydrogen sulfide with 0.01-g catalyst were calculated for the space time range of 0.040 – 0.047 seconds with Equation 2 to evaluate effects of moisture concentrations on reaction rates of hydrogen sulfide at 140^{o}C and 120 – 123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500 - 9,000-ppmv H_2S, 1,800- 4,500 ppmv SO_2, 2.5 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Reaction rates of H_2S with SO_2 are 0.35×10^{-5} – 1.17×10^{-5} g-mole/s/g-cat, as shown in Figure 15. Concentration of moisture and concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2

Reaction rates of hydrogen sulfide with 0.01-g catalyst were calculated for the space time range of 0.041 – 0.046 seconds with Equation 2 to evaluate effects of reaction temperatures on reaction rates of hydrogen sulfide at 125 - 155^oC and 112 – 123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500 - 9,000-ppmv H_2S, 1,800- 4,500 ppmv SO_2, 5- v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 112 - 125 cm^3/min at room temperature and atmospheric pressure (SCCM). Reaction rates of H_2S with SO_2 are $0.24x10^{-5}$ – 1.56×10^{-5} g-mole/s/g-cat, as shown in Figure 16. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2. However, reaction rates of H_2S with SO2 appear to decrease slightly with increased reaction temperatures over the temperature range of 135 - 145^oC.

Reaction rates of hydrogen sulfide with 0.01-g catalyst were calculated for the space time range of 0.010 – 0.045 seconds with Equation 2 to evaluate effects of reaction pressures on reaction rates of hydrogen sulfide at 140^oC and 28 – 123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500 - 9,000-ppmv H_2S, 1,800- 4,500 ppmv SO_2, 5-v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 99 – 101 cm^3/min at room temperature and atmospheric pressure (SCCM). Reaction rates of H_2S with SO_2 are $0.27x10^{-5}$ – 1.17×10^{-5} g-mole/s/g-cat, as shown in Figure 17. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2. However, reaction rates of H_2S with SO_2 appear to increase with increased reaction pressures over the pressure range of 28 – 123 psia.

Figure 15. Effects of moisture on reaction rate of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 70-v% H_2 and 2.5-13.6 v% moisture at 140^oC.

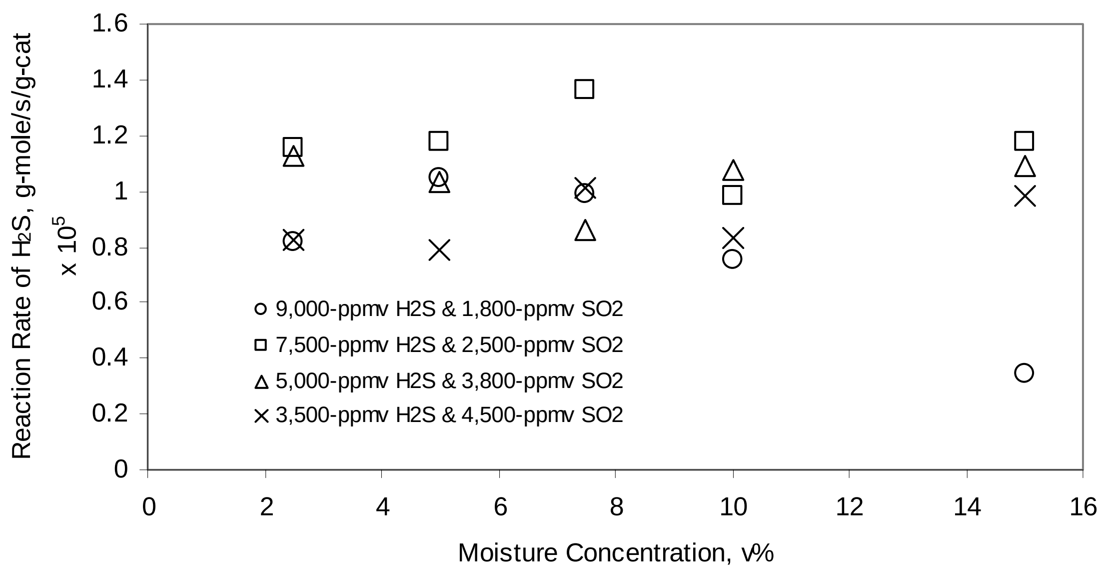


Figure 16. Effects of reaction temperature on reaction rate of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 70-v% H_2 and 5-v% moisture at 140^{o}C.

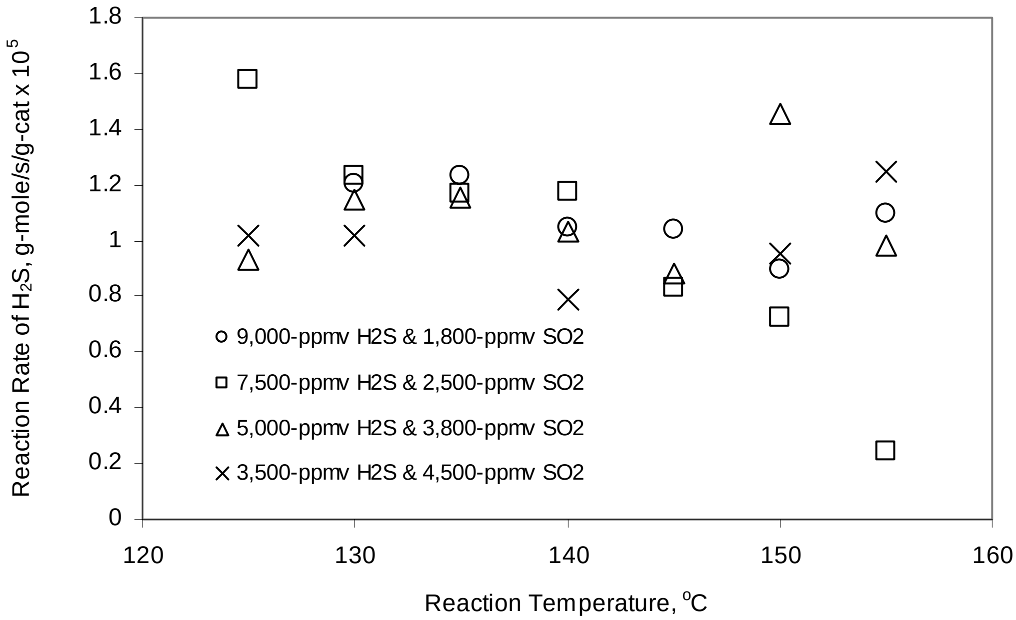
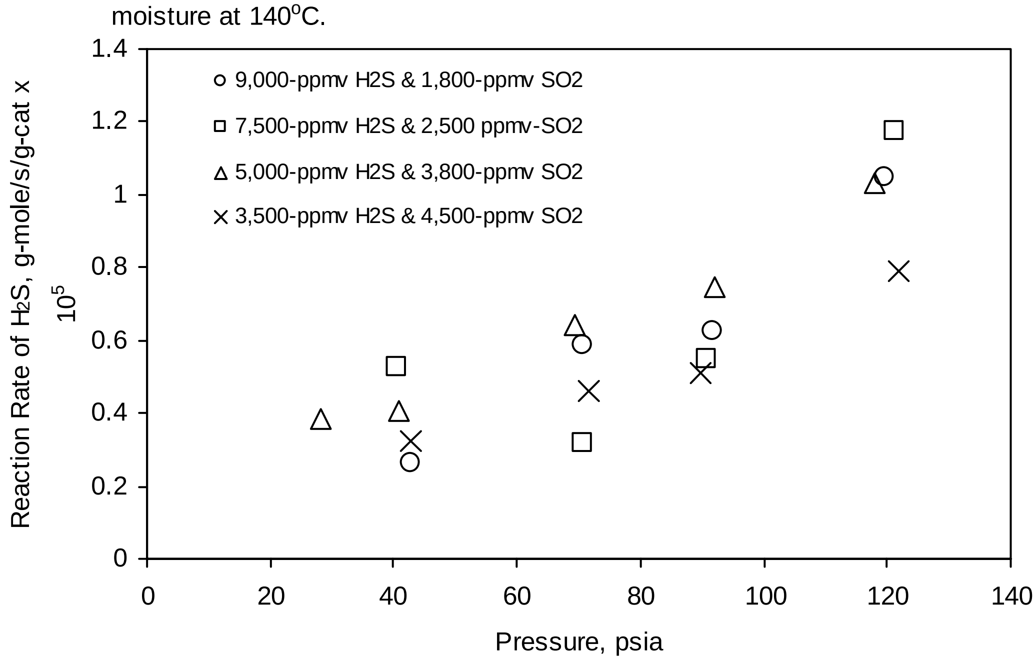


Figure 17. Effects of reaction pressure on reaction rate of H_2S with 0.01-g catalyst and a 100-SCCM feed stream containing 70-v% H_2 and 5-v% moisture at 140^{o}C.



CONCLUSIONS

The following conclusions were drawn based on experimental data generated from the differential reactor system, and their interpretations,

Concentration of moisture with 98 – 110 SCCM feed streams containing 8,200 – 9200 ppmv H_2S, 1,600 - 1,800 ppmv SO_2, and 64 – 72 v-% hydrogen appear to affect significantly conversion of H_2S into elemental sulfur in the moisture range of 5 – 13.6 v% at 140 ^oC and 120 psia. Conversion of H_2S decreases with increased concentration of moisture over the moisture range of 5 – 13.6 v%, as shown in Figure 3. Concentrations of moisture with 98 – 110 SCCM feed streams containing 64 – 77 v% hydrogen, 6,800 - 7,700 ppmv H_2S, and 2,300 – 2,600 ppmv SO_2 appear to affect slightly reaction rates for the conversion of H_2S into elemental sulfur in the moisture range of 2.6 – 13.6 v% at 119 – 123 psia and 140 ^oC. However, conversion of H_2S is highest at 7.3-v % moisture (see Figure 4).

Concentrations of moisture with 98 – 110 SCCM feed streams containing 64 – 77 v% hydrogen, 3,200 - 3,600 ppmv H_2S, and 4,100 - 4,600 ppmv SO_2 appear to affect slightly conversion of H_2S into elemental sulfur in the moisture range of 2.5 – 13.6 v% at 120 – 123 psia and 140 ^oC (see Figure 5). Conversion of H_2S slightly increases with moisture concentration. However, conversion of H_2S is highest at 7.3-v % moisture. Concentrations of moisture with 98 – 110 SCCM feed streams containing 64 – 72 v% hydrogen, 4,500 - 5,100 ppmv H_2S, and 3,400 – 3,900 ppmv SO_2 appear to affect slightly conversion of H_2S into elemental sulfur in the moisture range of 2.6 – 13.6 v% at 112 – 123 psia and 140 ^oC. Conversion of H_2S is not affected significantly by moisture concentration. However, conversion of H_2S is lowest at 7.3-v % moisture (see Figure 6).

Reaction temperatures with 100-SCCM feed streams containing 70-v% hydrogen, 9,000-ppmv H_2S, 1,800-ppmv SO_2, and 5-v% moisture affect slightly conversion of H_2S into elemental sulfur. Conversion of H_2S into elemental sulfur decreases with increased reaction temperature over the temperature range of 130 – 150^oC at 119 – 122 psia (see Figure 7). Reaction temperatures with 100-SCCM feed streams containing 70-v% hydrogen, 7,500-ppmv H_2S, 2,500-ppmv SO_2, and 5-v% moisture affect significantly conversion of H_2S into elemental sulfur. Conversion of H_2S into elemental sulfur decreases with increased reaction temperature over the temperature range of 125 – 155^oC at 120 – 125 psia, as opposed to the Arrhenius' equation (see Figure 8).

Reaction temperatures with 100 SCCM feed streams containing 70-v% hydrogen, 3,500-ppmv H_2S, 4,500-ppmv SO_2, and 5-v% moisture affect slightly conversion of H_2S into elemental sulfur over the temperature range of 125 – 155^oC at 120 – 122 psia. However, conversion of H_2S into elemental sulfur is lowest at 140^oC (see Figure 9). Reaction temperatures with 99 - 101 SCCM feed streams containing 69 – 71 v% hydrogen, 4,900 - 5,100-ppmv H_2S, 3,700- 3,800 ppmv SO_2, and 5-v% moisture affect slightly conversion of H_2S into elemental sulfur over the temperature range of 125 – 155^oC at 112 – 123 psia. However, conversion of H_2S into elemental sulfur is highest at 150^oC (see Figure 10).

Reaction pressures with 100 SCCM feed streams containing 70-v% hydrogen, 9,000-ppmv H_2S, 1,800-ppmv SO_2, and 5-v% moisture affect significantly conversion of H_2S into elemental sulfur in the pressure range of 43 -123 psia. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 43 -123 psia at 140^oC (see Figure 11). Reaction pressures with 99 -100 SCCM feed streams containing 70-v% hydrogen, 7,500 - 7,600 ppmv H_2S, 2,500-ppmv SO_2, and 5-v% moisture affect significantly conversion of H_2S into elemental sulfur in the pressure range of 71 -123 psia at 140^oC. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 71 -123 psia. However, conversion of H_2S is lowest at 71 psia (see Figure 12).

Reaction pressures with 100 SCCM feed streams containing 70-v% hydrogen, 3,500-ppmv H_2S, 4,500-ppmv SO_2, and 5 v% moisture affect significantly conversion of H_2S into elemental sulfur in the pressure range of 43 -122 psia at 140^oC. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 43 -122 psia (see Figure 13). Reaction pressures with 99 - 101 SCCM feed streams containing 70-v% hydrogen, 5,000-ppmv H_2S, 3,800-ppmv SO_2, and 5-v% moisture affect significantly conversion of H_2S into elemental sulfur in the pressure range of 28 -122 psia at 140^oC. Conversion of H_2S into elemental sulfur increases with reaction pressure in the pressure range of 28 -122 psia (see Figure 14).

Reaction rates of hydrogen sulfide with 0.01-g catalyst were calculated with experimental data to study effects of moisture concentrations, reaction temperature, and reaction pressure on reaction rates of hydrogen sulfide at 125-155^oC and 28 – 123 psia. Gas mixtures fed to the reactor contain 70-v% hydrogen, 3,500 - 9,000-ppmv H_2S, 1,800- 4,500 ppmv SO_2, and 2.5 – 13.6 v% moisture, and nitrogen as remainder. Volumetric feed rates of a gas mixture to the reactor are 98 - 110 cm^3/min at room temperature and atmospheric pressure (SCCM). Concentration of moisture and concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the moisture range of 2.5 – 13.6 v% moisture at 140^oC and 120 – 123 psia, as shown in Figure 15. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the temperature range of 135 - 145^oC at 5-v% moisture and 112 – 123 psia. However, reaction rates of H_2S with SO2 appear to decrease slightly with increased reaction temperatures over the temperature range of 135 - 145^oC at 5-v% moisture and 112 – 123 psia, as shown in Figure 16. Concentrations of both H_2S and SO_2 appear to affect slightly reaction rates of H_2S with SO_2 over the pressure range of 28 – 123 psia at 5-v% moisture and 140^oC. However, reaction rates of H_2S with SO_2 appear to increase with increased reaction pressures over the pressure range of 28 – 123 psia at 5-v% moisture and 140^oC, as shown in Figure 17.

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PUBLICATIONS AND PRESENTATIONS

Kyung C. Kwon, Santosh K. Gangwal, Janelle C. Houston, and Erica D. Jackson, Kinetics of Direct Oxidation of Hydrogen Sulfide in Coal Gas to Elemental Sulfur, DOE Annual Contractors' Review Meeting, Pittsburgh Marriott City Center, Pittsburgh, PA, June 4 – 5, 2002

Kyung C. Kwon, Santosh K. Gangwal, Suresh C. Jain, YoonKook Park, Janelle C. Houston and Erica D. Jackson, Conversion of Hydrogen Sulfide in Coal Gas to Elemental Sulfur, AIChE Annual Meeting, Indiana Convention Center, Indianapolis, Indiana, November 3 – 8, 2002.

STUDENTS ASSIGNED FOR THIS PROJECT

R. El-Amin-Foster, Max L. Weathersby, Janelle C. Houston, and Erica D. Jackson