ENERGY

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

Annual Technical Progress Report for the Period
October 1, 2003 to September 30, 2004

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January 2005

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 the hot-gas desulfurization using regenerable metal oxide sorbents followed by Direct Sulfur Recovery Process. The objective of this research is to support the near- and long-term process development 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 160-μm C-500-04 alumina catalyst particles and a micro bubble 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.

To achieve the above-mentioned objectives, experiments on conversion of hydrogen sulfide into liquid elemental sulfur were carried out for the space time range of 0.059 – 0.87 seconds at 125 - 155°C to evaluate effects of reaction temperature, H_2S concentration, reaction pressure, and catalyst loading on conversion of hydrogen sulfide into liquid elemental sulfur. Simulated coal gas mixtures consist of 62 - 78 v% hydrogen, 3,000 – 7,000-ppmv hydrogen sulfide, 1,500 - 3,500 ppmv sulfur dioxide, and 10 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to a micro bubble reactor are 50 cm^3/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 40 - 170 psia. The molar ratio of H_2S to SO_2 in the bubble reactor is maintained at 2 for all the reaction experiment runs.

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 advanced gasification 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 gasification 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:

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 involves three steps:

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

$$ZnO + H_2S \leftrightarrow ZnS + H_2O \quad \text{(HGD)}$$

(ii) sorbent regeneration using air to produce SO_2

$$ZnS + 3/2\, O_2 \leftrightarrow SO_2 + ZnO \quad \text{(regeneration)}$$

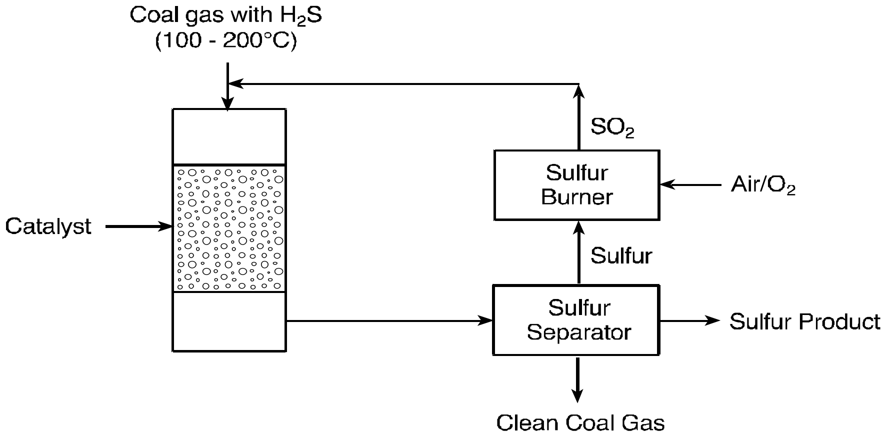
(iii) catalytic reduction of SO_2 using a small portion of the coal gas, to elemental sulfur by the Direct Sulfur Recovery Process (DSRP):

$$SO_2 + 2H_2 \text{ (or } 2CO) \leftrightarrow S + 2H_2O \text{ (or } 2CO_2) \quad \text{(DSRP)}$$

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 gasification plants producing both electricity and transportation-grade liquid fuels. To this end, 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 160-μm C-500-04 alumina catalyst particles and a fluidized-bed micro bubble reactor fabricated with perfluoroalkoxy (PFA), 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 to elemental sulfur were carried out for the space time range of 0.059 – 0.87 second at 125 - 155^{o}C and 40 - 170 psia to evaluate effects of reaction temperature, H_2S concentration, reaction pressure, and catalyst loading on conversion of hydrogen sulfide to elemental sulfur. Simulated coal gas mixtures consist of 62 – 78-v% hydrogen, 3,000 – 7,000-ppmv hydrogen sulfide, 1,500 - 3,500 ppmv sulfur dioxide, and 10 vol % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to the reactor are 50 cm^3/min at room temperature and atmospheric pressure. The amount range of catalyst in the reactor is 0.01 – 0.1 g. The molar ratio of H_2S to SO_2 in the bubble reactor is maintained at 2 for all the reaction experiment runs.

EXPERIMENTAL SETUPS

A fluidized-bed micro bubble reactor was fabricated with PFA (perfluoroalkoxy). The amount of a C-500-04 alumina catalyst in the reactor is 0.01 - 0.1 g. 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 fluidized-bed micro bubble reactor at 125 - 155^{o}C. Conversion of hydrogen sulfide to elemental sulfur was analyzed with a gas chromatograph. The range of space (residence) time of the reaction gas mixture in the reactor was 0.059 – 0.87 seconds under the reaction conditions. Space times are obtained by dividing the bulk volume of catalyst particles with the volumetric flow rate of a feed gaseous mixture at reaction conditions.

Figure 2. Schematic diagram of the experimental set-up

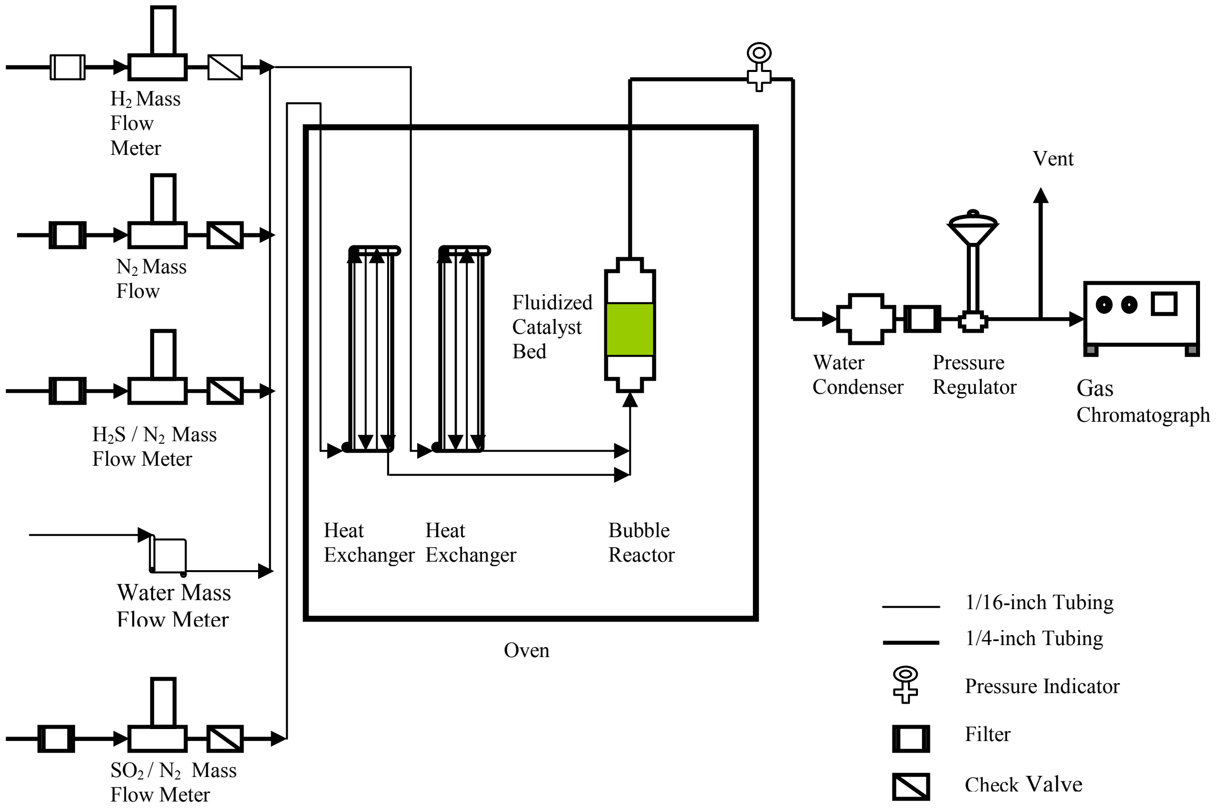


Table 1. Experimental conditions for the reaction of hydrogen sulfide with sulfur dioxide.

Amounts of catalyst particles in the reactor, g	0.01 – 0.1
Temperature, ^{o}C:	125 - 155
Reaction Pressure, psia	40 - 170
Space Time under the reaction conditions, second:	0.59 – 0.87
Mean Particle Size, μm	160
Gas Flow Rate, cc/min at room temperature and 1 atm (SCCM)	50
Hydrogen, vol %	62 - 78
Moisture, vol %:	10
Concentration of H_2S, ppmv	3,000 – 7,000
Concentration of SO_2, ppmv	1,500 – 3,500
Nitrogen, vol %	Remainder

A micro bubble reactor assembly mainly consists of four mass flow meters for gases, one micro bubble 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 condenser (see

Figure 2). The preheaters are made of 1/16-inch PFA tubing. The reactor, loaded with the C-500-04 alumina 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.

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 3,000 – 7,000 ppmv hydrogen sulfide with 1,500 – 3,500 ppmv sulfur dioxide in the presence of 62 - 78 v-% hydrogen, 10 v-% moisture, and 0.01 – 0.10 g catalyst at 125 - 155 °C, 40 - 170 psia, and 0.059 – 0.87 second space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v%					Conversion %
						H_2	H_2S	SO_2	moisture	N_2	
B5-1	140	120.2	0.0205	50	0.174	70	0.498	0.25	10	19.252	52.38
B5-2	140	120.7	0.0206	50	0.176	70	0.498	0.25	10	19.252	31.87
B5-3	140	117.7	0.0206	50	0.171	70	0.498	0.25	10	19.252	24.04
B5-4	140	121.03	0.0206	50	0.176	70	0.498	0.25	10	19.252	23.41
B5-5	140	119.7	0.0204	50	0.172	70	0.498	0.25	10	19.252	23.98
B5-6	140	122	0.0404	50	0.348	70	0.498	0.25	10	19.252	34.75
B5-7	140	121.3	0.0406	50	0.348	70	0.498	0.25	10	19.252	17.90
B5-8	140	118.9	0.0403	50	0.338	70	0.498	0.25	10	19.252	31.48
B5-9	140	117.87	0.0401	50	0.334	70	0.498	0.25	10	19.252	32.88
B5-10	140	119.5	0.0601	50	0.507	70	0.498	0.25	10	19.252	38.38
B5-11	140	118	0.0604	50	0.503	70	0.498	0.25	10	19.252	36.55
B5-12	140	119.2	0.0801	50	0.674	70	0.498	0.25	10	19.252	46.27
B5-13	140	122	0.0806	50	0.694	70	0.498	0.25	10	19.252	45.30
B5-14	140	114.45	0.0104	50	0.084	70	0.498	0.25	10	19.252	41.87
B5-15	140	120.08	0.0108	50	0.092	70	0.498	0.25	10	19.252	56.15
B5-16	140	119.03	0.0207	50	0.174	70	0.498	0.25	10	19.252	21.45
B5-17	140	168.99	0.0207	50	0.247	70	0.498	0.25	10	19.252	64.59
B5-18	140	169.7	0.0205	50	0.246	70	0.498	0.25	10	19.252	53.72
B5-19	140	169.2	0.0209	50	0.250	70	0.498	0.25	10	19.252	56.04
B5-20	140	145.5	0.0205	50	0.211	70	0.498	0.25	10	19.252	34.30
B5-21	140	144.87	0.0208	50	0.213	70	0.498	0.25	10	19.252	26.46
B5-22	140	144.45	0.0205	50	0.209	70	0.498	0.25	10	19.252	42.86
B5-23	140	143.12	0.0205	50	0.207	70	0.498	0.25	10	19.252	54.61

Table 3. Continued-1

						Feed Composition, v%					
Run Number	Temperature, ^oC	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	H_2	H_2S	SO_2	moisture	N_2	Conversion %
B5-24	140	144.41	0.0205	50	0.209	70	0.498	0.25	10	19.252	21.74
B5-25	140	145.7	0.0203	50	0.209	70	0.498	0.25	10	19.252	43.53
B5-26	140	95.84	0.0205	50	0.139	70	0.498	0.25	10	19.252	24.26
B5-27	140	95.7	0.0205	50	0.138	70	0.498	0.25	10	19.252	20.22
B5-28	140	96.7	0.02	50	0.137	70	0.498	0.25	10	19.252	24.57
B5-29	140	67.7	0.0202	50	0.097	70	0.498	0.25	10	19.252	20.78
B5-30	140	67.7	0.0201	50	0.096	70	0.498	0.25	10	19.252	28.36
B5-31	140	67.7	0.0208	50	0.099	70	0.498	0.25	10	19.252	17.50
B5-32	140	67.7	0.0207	50	0.099	70	0.498	0.25	10	19.252	18.09
B5-33	140	41.2	0.0205	50	0.060	70	0.498	0.25	10	19.252	9.10
B5-34	140	42	0.0205	50	0.061	70	0.498	0.25	10	19.252	14.47
B5-35	140	40.2	0.0207	50	0.059	70	0.498	0.25	10	19.252	12.46
B5-36	140	119.7	0.0205	50	0.173	70	0.5	0.25	10	19.25	27.58
B5-37	140	120.7	0.0207	50	0.176	70	0.5	0.25	10	19.25	28.90
B5-38	155	116.95	0.0202	50	0.161	70	0.5	0.25	10	19.25	21.42
B5-39	155	121.87	0.0205	50	0.170	70	0.5	0.25	10	19.25	23.45
B5-40	155	119.7	0.0108	50	0.088	70	0.5	0.25	10	19.25	43.60
B5-41	155	118.7	0.0102	50	0.082	70	0.5	0.25	10	19.25	24.58
B5-42	155	120.7	0.0206	50	0.169	70	0.5	0.25	10	19.25	23.44
B5-43	125	119.7	0.0201	50	0.176	70	0.5	0.25	10	19.25	19.90
B5-44	125	119.7	0.0206	50	0.181	70	0.5	0.25	10	19.25	35.31
B5-45	125	120.7	0.0205	50	0.181	70	0.5	0.25	10	19.25	15.78
B5-46	125	120.7	0.0204	50	0.180	70	0.5	0.25	10	19.25	17.94
B5-47	125	120.7	0.0207	50	0.183	70	0.5	0.25	10	19.25	16.65
B5-48	130	119.7	0.0205	50	0.178	70	0.5	0.25	10	19.25	23.65
B5-49	130	118.7	0.0206	50	0.177	70	0.5	0.25	10	19.25	33.92
B5-50	130	119.7	0.0203	50	0.176	70	0.5	0.25	10	19.25	28.14
B5-51	130	118.7	0.0202	50	0.173	70	0.5	0.25	10	19.25	18.12
B5-52	130	117.7	0.0207	50	0.176	70	0.5	0.25	10	19.25	18.76
B5-53	150	118.7	0.0205	50	0.168	70	0.5	0.25	10	19.25	19.50
B5-54	150	119.7	0.0204	50	0.168	70	0.5	0.25	10	19.25	21.29
B5-55	150	118.7	0.0204	50	0.167	70	0.5	0.25	10	19.25	23.69
B5-56	150	118.7	0.0202	50	0.165	70	0.5	0.25	10	19.25	22.37
B5-57	145	120.7	0.0205	50	0.173	70	0.5	0.25	10	19.25	31.83
B5-58	145	119.7	0.0207	50	0.173	70	0.5	0.25	10	19.25	30.46
B5-59	145	118.7	0.0207	50	0.171	70	0.5	0.25	10	19.25	32.12
B5-60	135	118.7	0.0202	50	0.171	70	0.5	0.25	10	19.25	31.10
B5-61	135	118.7	0.0206	50	0.175	70	0.5	0.25	10	19.25	27.23
B5-62	135	119.7	0.0206	50	0.176	70	0.5	0.25	10	19.25	23.54
B5-63	135	119.7	0.0208	50	0.178	70	0.5	0.25	10	19.25	24.89
B5-64	135	118.4	0.0205	50	0.173	70	0.5	0.25	10	19.25	10.19
B5-65	135	116.7	0.0208	50	0.173	70	0.5	0.25	10	19.25	13.81

Table 3. Continued-2

						Feed Composition, v%					
Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	H_2	H_2S	SO_2	moisture	N_2	Conversion %
B5-66	135	119.7	0.0207	50	0.177	70	0.5	0.25	10	19.25	13.96
B5-67	140	121.7	0.0204	50	0.175	70	0.5	0.25	10	19.25	31.94
B5-68	140	119.7	0.0207	50	0.175	70	0.5	0.25	10	19.25	4.98
B5-69	140	122.7	0.0204	50	0.177	70	0.5	0.25	10	19.25	20.45
B5-70	140	114.7	0.0209	50	0.169	70	0.5	0.25	10	19.25	24.49
B5-71	140	118.7	0.0202	50	0.169	70	0.5	0.25	10	19.25	24.74
B5-72	140	119.7	0.1001	50	0.846	70	0.5	0.25	10	19.25	47.34
B5-73	140	118.7	0.1004	50	0.841	70	0.5	0.25	10	19.25	54.50
B5-74	140	122.7	0.1004	50	0.870	70	0.5	0.25	10	19.25	44.49
B5-75	140	119.7	0.1004	50	0.848	70	0.5	0.25	10	19.25	45.12
B5-76	140	121.7	0.0801	50	0.688	70	0.5	0.25	10	19.25	41.45
B5-77	140	121.7	0.0802	50	0.689	70	0.5	0.25	10	19.25	38.91
B5-78	140	120.7	0.0801	50	0.682	70	0.5	0.25	10	19.25	32.42
B5-79	140	121.7	0.0804	50	0.691	70	0.5	0.25	10	19.25	33.62
B5-80	140	122.7	0.0802	50	0.695	70	0.5	0.25	10	19.25	35.18
B5-81	140	122.7	0.0604	50	0.523	70	0.5	0.25	10	19.25	22.25
B5-82	140	120.7	0.0601	50	0.512	70	0.5	0.25	10	19.25	21.52
B5-83	140	121.7	0.0607	50	0.521	70	0.5	0.25	10	19.25	32.77
B5-84	140	122.7	0.0601	50	0.521	70	0.5	0.25	10	19.25	38.02
B5-85	140	117.7	0.0604	50	0.502	70	0.5	0.25	10	19.25	22.01
B5-86	140	119.7	0.0604	50	0.510	70	0.5	0.25	10	19.25	33.36
B5-87	140	121.7	0.0404	50	0.347	70	0.5	0.25	10	19.25	19.33
B5-88	140	120.7	0.0401	50	0.342	70	0.5	0.25	10	19.25	23.72
B5-89	140	119.7	0.0407	50	0.344	70	0.5	0.25	10	19.25	21.55
B5-90	140	121.7	0.0402	50	0.345	70	0.5	0.25	10	19.25	26.93
B5-91	140	119.7	0.04	50	0.338	70	0.5	0.25	10	19.25	16.75
B5-92	140	119.7	0.0404	50	0.341	70	0.5	0.25	10	19.25	27.82
B5-93	140	122.7	0.0207	50	0.179	62	0.7	0.35	10	26.95	26.33
B5-94	140	122.7	0.0206	50	0.178	62	0.7	0.35	10	26.95	30.54
B5-95	140	122.7	0.0204	50	0.177	62	0.7	0.35	10	26.95	30.7
B5-96	140	121.7	0.0206	50	0.177	62	0.7	0.35	10	26.95	29.74
B5-97	140	121.7	0.0203	50	0.174	78	0.3	0.15	10	11.55	14.86
B5-98	140	117.7	0.0204	50	0.169	78	0.3	0.15	10	11.55	13.24
B5-99	140	121.7	0.0202	50	0.174	78	0.3	0.15	10	11.55	16.65
B5-100	140	118.7	0.0201	50	0.168	78	0.3	0.15	10	11.55	11.31
B5-101	140	120.7	0.0204	50	0.174	78	0.3	0.15	10	11.55	13.78

Table 4. Effects of reaction temperature on conversion of H_2S to elemental sulfur with 0.01 – 0.02-g catalyst and a 50-SCCM feed stream containing 5,000-ppmv H_2S and 2,500-ppmv SO_2, 10-v% moisture, and 70-v% H_2 at 125 – 155°C, 117 – 122 psia and 0.082 – 0.183 second space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v%					Conversion %
						H_2	H_2S	SO_2	moisture	N_2	
B5-43	125	119.7	0.0201	50	0.176	70	0.5	0.25	10	19.25	19.90
B5-44	125	119.7	0.0206	50	0.181	70	0.5	0.25	10	19.25	35.31
B5-45	125	120.7	0.0205	50	0.181	70	0.5	0.25	10	19.25	15.78
B5-46	125	120.7	0.0204	50	0.180	70	0.5	0.25	10	19.25	17.94
B5-47	125	120.7	0.0207	50	0.183	70	0.5	0.25	10	19.25	16.65
B5-48	130	119.7	0.0205	50	0.178	70	0.5	0.25	10	19.25	23.65
B5-49	130	118.7	0.0206	50	0.177	70	0.5	0.25	10	19.25	33.92
B5-50	130	119.7	0.0203	50	0.176	70	0.5	0.25	10	19.25	28.14
B5-51	130	118.7	0.0202	50	0.173	70	0.5	0.25	10	19.25	18.12
B5-52	130	117.7	0.0207	50	0.176	70	0.5	0.25	10	19.25	18.76
B5-60	135	118.7	0.0202	50	0.171	70	0.5	0.25	10	19.25	31.10
B5-61	135	118.7	0.0206	50	0.175	70	0.5	0.25	10	19.25	27.23
B5-62	135	119.7	0.0206	50	0.176	70	0.5	0.25	10	19.25	23.54
B5-63	135	119.7	0.0208	50	0.178	70	0.5	0.25	10	19.25	24.89
B5-64	135	118.4	0.0205	50	0.173	70	0.5	0.25	10	19.25	10.19
B5-65	135	116.7	0.0208	50	0.173	70	0.5	0.25	10	19.25	13.81
B5-66	135	119.7	0.0207	50	0.177	70	0.5	0.25	10	19.25	13.96
B5-3	140	117.7	0.0206	50	0.171	70	0.498	0.25	10	19.25	24.04
B5-4	140	121.03	0.0206	50	0.176	70	0.498	0.25	10	19.25	23.41
B5-5	140	119.7	0.0204	50	0.172	70	0.498	0.25	10	19.25	23.98
B5-16	140	119.03	0.0207	50	0.174	70	0.498	0.25	10	19.25	21.45
B5-67	140	121.7	0.0204	50	0.175	70	0.5	0.25	10	19.25	31.94
B5-71	140	118.7	0.0202	50	0.169	70	0.5	0.25	10	19.25	24.74
B5-57	145	120.7	0.0205	50	0.173	70	0.5	0.25	10	19.25	31.83
B5-58	145	119.7	0.0207	50	0.173	70	0.5	0.25	10	19.25	30.46
B5-59	145	118.7	0.0207	50	0.171	70	0.5	0.25	10	19.25	32.12
B5-53	150	118.7	0.0205	50	0.168	70	0.5	0.25	10	19.25	19.50
B5-54	150	119.7	0.0204	50	0.168	70	0.5	0.25	10	19.25	21.29
B5-55	150	118.7	0.0204	50	0.167	70	0.5	0.25	10	19.25	23.69
B5-56	150	118.7	0.0202	50	0.165	70	0.5	0.25	10	19.25	22.37
B5-38	155	116.95	0.0202	50	0.161	70	0.5	0.25	10	19.25	21.42
B5-39	155	121.87	0.0205	50	0.170	70	0.5	0.25	10	19.25	23.45
B5-40	155	119.7	0.0108	50	0.088	70	0.5	0.25	10	19.25	43.60
B5-41	155	118.7	0.0102	50	0.082	70	0.5	0.25	10	19.25	24.58
B5-42	155	120.7	0.0206	50	0.169	70	0.5	0.25	10	19.25	23.44

Table 5. Effects of reaction pressure on conversion of H_2S with 0.02-g alumina catalyst and a 50-SCCM feed stream containing 5,000-ppmv H_2S and 2,500-ppmv SO_2, 10-v% moisture, and 70-v% H_2 at 140°C, 40 – 170 psia, and 0.059 – 0.25 second space time.

						Feed Composition, v%					
Run Number	Temperature , °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	H_2	H_2S	SO_2	moisture	N_2	Conversion %
B5-33	140	41.2	0.0205	50	0.060	70	0.498	0.25	10	19.252	9.10
B5-34	140	42	0.0205	50	0.061	70	0.498	0.25	10	19.252	14.47
B5-35	140	40.2	0.0207	50	0.059	70	0.498	0.25	10	19.252	12.46
B5-29	140	67.7	0.0202	50	0.097	70	0.498	0.25	10	19.252	20.78
B5-30	140	67.7	0.0201	50	0.096	70	0.498	0.25	10	19.252	28.36
B5-31	140	67.7	0.0208	50	0.099	70	0.498	0.25	10	19.252	17.50
B5-26	140	95.84	0.0205	50	0.139	70	0.498	0.25	10	19.252	24.26
B5-27	140	95.7	0.0205	50	0.138	70	0.498	0.25	10	19.252	20.22
B5-28	140	96.7	0.02	50	0.137	70	0.498	0.25	10	19.252	24.57
B5-3	140	117.7	0.0206	50	0.171	70	0.498	0.25	10	19.252	24.04
B5-4	140	121.03	0.0206	50	0.176	70	0.498	0.25	10	19.252	23.41
B5-5	140	119.7	0.0204	50	0.172	70	0.498	0.25	10	19.252	23.98
B5-16	140	119.03	0.0207	50	0.174	70	0.498	0.25	10	19.252	21.45
B5-67	140	121.7	0.0204	50	0.175	70	0.5	0.25	10	19.25	31.94
B5-68	140	119.7	0.0207	50	0.175	70	0.5	0.25	10	19.25	4.98
B5-69	140	122.7	0.0204	50	0.177	70	0.5	0.25	10	19.25	20.45
B5-70	140	114.7	0.0209	50	0.169	70	0.5	0.25	10	19.25	24.49
B5-71	140	118.7	0.0202	50	0.169	70	0.5	0.25	10	19.25	24.74
B5-20	140	145.5	0.0205	50	0.211	70	0.498	0.25	10	19.252	34.30
B5-21	140	144.87	0.0208	50	0.213	70	0.498	0.25	10	19.252	26.46
B5-22	140	144.45	0.0205	50	0.209	70	0.498	0.25	10	19.252	42.86
B5-23	140	143.12	0.0205	50	0.207	70	0.498	0.25	10	19.252	54.61
B5-24	140	144.41	0.0205	50	0.209	70	0.498	0.25	10	19.252	21.74
B5-25	140	145.7	0.0203	50	0.209	70	0.498	0.25	10	19.252	43.53
B5-17	140	168.99	0.0207	50	0.247	70	0.498	0.25	10	19.252	64.59
B5-18	140	169.7	0.0205	50	0.246	70	0.498	0.25	10	19.252	53.72
B5-19	140	169.2	0.0209	50	0.250	70	0.498	0.25	10	19.252	56.04

Table 6. A reaction model developed with experimental data of conversion of 5,000 – 7,000 ppmv hydrogen sulfide with 2,500 – 3,500 ppmv sulfur dioxide in the presence of 62 - 70 v-% hydrogen, 10-v % moisture, and 0.02-g catalyst at 140 °C, 40 – 123 psia, and 0.059 - 0.178 second space time.

						Feed Composition, v%					
Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	H_2	H_2S	SO_2	moisture	N_2	Conversion %
B5-33	140	41.2	0.0205	50	0.060	70	0.498	0.25	10	19.252	9.10
B5-34	140	42	0.0205	50	0.061	70	0.498	0.25	10	19.252	14.47
B5-35	140	40.2	0.0207	50	0.059	70	0.498	0.25	10	19.252	12.46

Table 6. Continued-1

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v%					Conversion %
						H_2	H_2S	SO_2	moisture	N_2	
B5-31	140	67.7	0.0208	50	0.099	70	0.498	0.25	10	19.252	17.50
B5-32	140	67.7	0.0207	50	0.099	70	0.498	0.25	10	19.252	18.09
B5-26	140	95.84	0.0205	50	0.139	70	0.498	0.25	10	19.252	24.26
B5-28	140	96.7	0.02	50	0.137	70	0.498	0.25	10	19.252	24.57
B5-3	140	117.7	0.0206	50	0.171	70	0.498	0.25	10	19.252	24.04
B5-67	140	121.7	0.0204	50	0.175	70	0.5	0.25	10	19.25	31.94
B5-71	140	118.7	0.0202	50	0.169	70	0.5	0.25	10	19.25	24.74
B5-94	140	122.7	0.0206	50	0.178	62	0.7	0.35	10	26.95	30.54
B5-95	140	122.7	0.0204	50	0.177	62	0.7	0.35	10	26.95	30.7
B5-96	140	121.7	0.0206	50	0.177	62	0.7	0.35	10	26.95	29.74

Table 7. A comparison of experimental surface reaction rates with predicted surface reaction rates of H_2S for conversion of 5,000 - 7,000 ppmv hydrogen sulfide with 2,500 - 3,500 ppmv sulfur dioxide in the presence of 62 - 70 v-% hydrogen, 10-v % moisture, and 0.02-g catalyst at 140 °C, 40 – 123 psia, and 0.059 - 0.178 second space time.

Run Number	Pressure, psia			Conversion of H_2S	Molar Flow Rate H_2S, g-mole/s	Left-Side Value of Rearranged Reaction Model psia$^{1.5}$ (g-cat-s/g-mole)$^{0.5}$	Surface Reaction Rate, g-mole/s-g-cat	
	Total	H_2S	SO_2				Experimental	Predicted
B5-33	41.2	0.1865	0.0937	9.10	1.6966E-07	66	7.535E-07	9.355E-07
B5-34	42	0.1789	0.0899	14.47	1.6966E-07	49	1.198E-06	8.911E-07
B5-35	40.2	0.1752	0.0880	12.46	1.6966E-07	52	1.022E-06	8.695E-07
B5-31	67.7	0.2781	0.1397	17.50	1.6966E-07	87	1.427E-06	1.481E-06
B5-32	67.7	0.2762	0.1388	18.09	1.6966E-07	85	1.483E-06	1.469E-06
B5-26	95.84	0.3615	0.1817	24.26	1.6966E-07	109	2.007E-06	1.982E-06
B5-28	96.7	0.3632	0.1826	24.57	1.6966E-07	108	2.085E-06	1.992E-06
B5-3	117.7	0.4452	0.2238	24.04	1.6966E-07	150	1.980E-06	2.484E-06
B5-67	121.7	0.4141	0.2071	31.94	1.7034E-07	116	2.656E-06	2.290E-06
B5-71	118.7	0.4467	0.2233	24.74	1.7034E-07	146	2.078E-06	2.483E-06
B5-94	122.7	0.5966	0.2983	30.54	2.3847E-07	173	3.535E-06	3.360E-06
B5-95	122.7	0.5952	0.2976	30.7	2.3847E-07	171	3.589E-06	3.352E-06
B5-96	121.7	0.5985	0.2993	29.74	2.3847E-07	176	3.443E-06	3.372E-06

Table 8. Effects of catalyst loading and space time on both conversion of H_2S to elemental sulfur and reaction rate of H_2S with 0.02 – 0.1 g catalyst and a 50-SCCM feed stream containing 5,000-ppmv H_2S and 2,500-ppmv SO_2, 10-v% moisture, and 70-v% H_2 at 115 - 123 psia, 140°C, and 0.169 – 0.870 s space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Reaction Rate of H_2S, g-mole/s-g-cat	Space Time, s	Feed Composition, v%					Conversion %
						H_2	H_2S	SO_2	moisture	N_2	
B5-3	140	117.7	0.0206	1.9801E-06	0.171	70	0.498	0.25	10	19.252	24.04
B5-4	140	121.03	0.0206	1.9282E-06	0.176	70	0.498	0.25	10	19.252	23.41
B5-5	140	119.7	0.0204	1.9946E-06	0.172	70	0.498	0.25	10	19.252	23.98
B5-16	140	119.03	0.0207	1.7579E-06	0.174	70	0.498	0.25	10	19.252	21.45
B5-69	140	122.7	0.0204	1.7073E-06	0.177	70	0.5	0.25	10	19.25	20.45
B5-70	140	114.7	0.0209	1.9963E-06	0.169	70	0.5	0.25	10	19.25	24.49
B5-71	140	118.7	0.0202	2.0862E-06	0.169	70	0.5	0.25	10	19.25	24.74
B5-88	140	120.7	0.0401	1.0076E-06	0.342	70	0.5	0.25	10	19.25	23.72
B5-90	140	121.7	0.0402	1.1411E-06	0.345	70	0.5	0.25	10	19.25	26.93
B5-92	140	119.7	0.0404	1.1728E-06	0.341	70	0.5	0.25	10	19.25	27.82
B5-81	140	122.7	0.0604	6.2737E-07	0.523	70	0.5	0.25	10	19.25	22.25
B5-83	140	121.7	0.0607	9.1968E-07	0.521	70	0.5	0.25	10	19.25	32.77
B5-84	140	122.7	0.0601	1.0775E-06	0.521	70	0.5	0.25	10	19.25	38.02
B5-85	140	117.7	0.0604	6.2083E-07	0.502	70	0.5	0.25	10	19.25	22.01
B5-86	140	119.7	0.0604	9.4075E-07	0.510	70	0.5	0.25	10	19.25	33.36
B5-77	140	121.7	0.0802	8.2645E-07	0.689	70	0.5	0.25	10	19.25	38.91
B5-78	140	120.7	0.0801	6.8951E-07	0.682	70	0.5	0.25	10	19.25	32.42
B5-79	140	121.7	0.0804	7.1231E-07	0.691	70	0.5	0.25	10	19.25	33.62
B5-80	140	122.7	0.0802	7.4729E-07	0.695	70	0.5	0.25	10	19.25	35.18
B5-72	140	119.7	0.1001	8.0551E-07	0.846	70	0.5	0.25	10	19.25	47.34
B5-74	140	122.7	0.1004	7.5478E-07	0.870	70	0.5	0.25	10	19.25	44.49
B5-75	140	119.7	0.1004	7.6557E-07	0.848	70	0.5	0.25	10	19.25	45.12

Table 9. Effects of concentrations of H_2S on conversion of H_2S with 0.02-g alumina catalyst and a 50-SCCM feed stream containing 3,000 – 7,000-ppmv H_2S and 1,500 - 3,500-ppmv SO_2, 10-v% moisture, and 62 - 78-v% H_2 at 140°C, 115 – 123 psia, and 0.168 – 0.178 second space time.

Run Number	Temperature, °C	Pressure psia	Catalyst Amount, g	Total Feed scc/min	Space Time, s	Feed Composition, v%					Conversion %
						H_2	H_2S	SO_2	moisture	N_2	
B5-3	140	117.7	0.0206	50	0.171	70	0.498	0.25	10	19.252	24.04
B5-4	140	121.03	0.0206	50	0.176	70	0.498	0.25	10	19.252	23.41
B5-5	140	119.7	0.0204	50	0.172	70	0.498	0.25	10	19.252	23.98
B5-16	140	119.03	0.0207	50	0.174	70	0.498	0.25	10	19.252	21.45
B5-36	140	119.7	0.0205	50	0.173	70	0.5	0.25	10	19.25	27.58
B5-37	140	120.7	0.0207	50	0.176	70	0.5	0.25	10	19.25	28.90

Table 9. 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 %
B5-69	140	122.7	0.0204	50	0.177	70	0.5	0.25	10	19.25	20.45
B5-70	140	114.7	0.0209	50	0.169	70	0.5	0.25	10	19.25	24.49
B5-71	140	118.7	0.0202	50	0.169	70	0.5	0.25	10	19.25	24.74
B5-94	140	122.7	0.0206	50	0.178	62	0.7	0.35	10	26.95	30.54
B5-95	140	122.7	0.0204	50	0.177	62	0.7	0.35	10	26.95	30.7
B5-96	140	121.7	0.0206	50	0.177	62	0.7	0.35	10	26.95	29.74
B5-97	140	121.7	0.0203	50	0.174	78	0.3	0.15	10	11.55	14.86
B5-98	140	117.7	0.0204	50	0.169	78	0.3	0.15	10	11.55	13.24
B5-99	140	121.7	0.0202	50	0.174	78	0.3	0.15	10	11.55	16.65
B5-100	140	118.7	0.0201	50	0.168	78	0.3	0.15	10	11.55	11.31
B5-101	140	120.7	0.0204	50	0.174	78	0.3	0.15	10	11.55	13.78

CALCULATIONS

Gaseous samples having a 4-cm^3 volume, obtained from the outlet stream of a micro bubble reactor, are injected into a gas chromatograph to analyze gas chromatography (GC) areas of gaseous samples. Conversion of H_2S is obtained by dividing the GC area of H_2S from a reaction run with that from its blank run (see Equation 1).

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

where x_A: conversion of H_2S.

A_B: GC area of H_2S of the 4-cm^3 gaseous sample from a blank run.

A_R: GC area of H_2S of the 4-cm^3 gaseous sample from a reaction run.

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

Experimental surface reaction rate of conversion of H_2S to elemental sulfur in a micro bubble reactor is obtained with amount of the alumina catalyst loaded in the micro bubble reactor, molar feed rate of H_2S to the micro bubble reactor, and conversion of H_2S, as shown in the following equation.

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

where $-r_A'$: surface reaction rate

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

x_A: conversion of H_2S to elemental sulfur

W: amount of catalyst particles in a bubble reactor

Space time of gaseous reaction mixtures in the bubble reactor is calculated by dividing bulk volume of catalyst particles with volumetric flow rate of gaseous reaction mixtures fed under reaction conditions, as shown in the following equation.

$$\tau = \frac{V}{v_o} \qquad (4)$$

where τ: space time

V: bulk volume of catalyst particles

v_o: volumetric flow rate of gaseous reaction mixture fed under reaction conditions

RESULTS AND DISCUSSION

Experiments on conversion of hydrogen sulfide to elemental sulfur were carried out for the space time range of 0.059 – 0.87 second at 125 - 155°C to evaluate effects of catalyst amount, hydrogen sulfide concentration, and reaction pressure on conversion of hydrogen sulfide to elemental sulfur. Simulated coal gas mixtures consist of 62 - 78 v% hydrogen, 3,000 – 7,000 ppmv hydrogen sulfide, 1,500 - 3,500 ppmv sulfur dioxide, and 10-v % moisture, and nitrogen as remainder. Volumetric feed rates of a simulated coal gas mixture to a bubble reactor are 50 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 40 - 170 psia, loading 0.01 – 0.1 g catalyst particles in the micro bubble reactor. The molar ratio of H_2S to SO_2 in the bubble reactor is maintained at 2 for all the reaction experiment runs.

Effects of Concentration of H_2S on Conversion of H_2S to elemental sulfur

Experiments on conversion of hydrogen sulfide to elemental sulfur with 0.02-g catalyst were carried out for the space time range of 0.168 – 0.178 s to evaluate effects of H_2S concentration on conversion of hydrogen sulfide to elemental sulfur at 140°C and 115 – 123 psia. A gas mixture consisting of 62 - 78 v% hydrogen, 3,000 – 7,000-ppmv H_2S, 1,500 – 3,500 ppmv SO_2, 10 v% moisture, and nitrogen as remainder is fed to a micro bubble reactor. Volumetric feed rates of the gas mixture to the micro bubble reactor are 50 SCCM. Conversion of H_2S to elemental sulfur is 0.11 – 0.30.

Concentration of H_2S in the presence of 10-v% moisture and 62 - 78 v% H_2 affects conversion of H_2S to elemental sulfur in the H_2S concentration range of 3,000 - 7,000 ppmv in a simulated coal gas mixture at 115 – 123 psia (see Figure 3). Conversion of H_2S to elemental sulfur increases with increased concentrations of both H_2S and SO_2.

Figure 3. Effects of initial H_2S concentration on conversion of H_2S with a stoichiometric molar feed of H_2S and SO2 and a 50 cc/min feed stream containing 62-78 v-% H_2, 10-V% moisture, and 0.02-g catalyst at 140°C and 115 -123 psia.

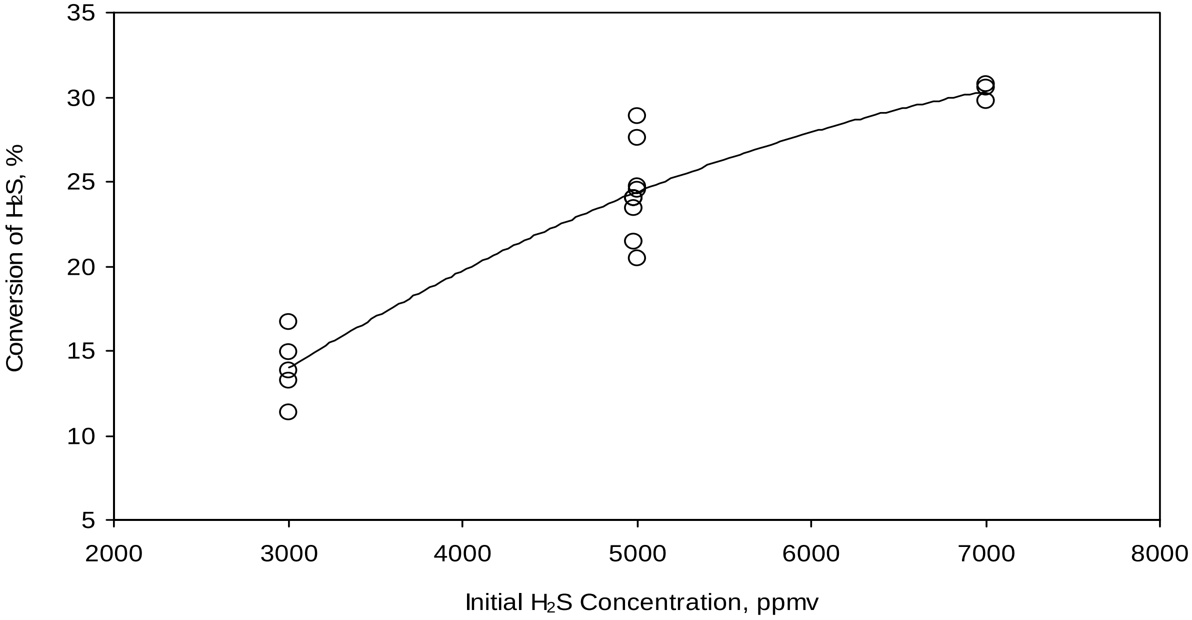


Figure 4. Effects of temperature on conversion of H_2S with a 50 cc/min feed stream containing 5000-ppm H_2S, 2500-ppm SO_2, 10-v% moisture, 70-v% H_2, and 0.02-g catalyst at 117 - 122 psia and 0.161 - 0.183 s space time .

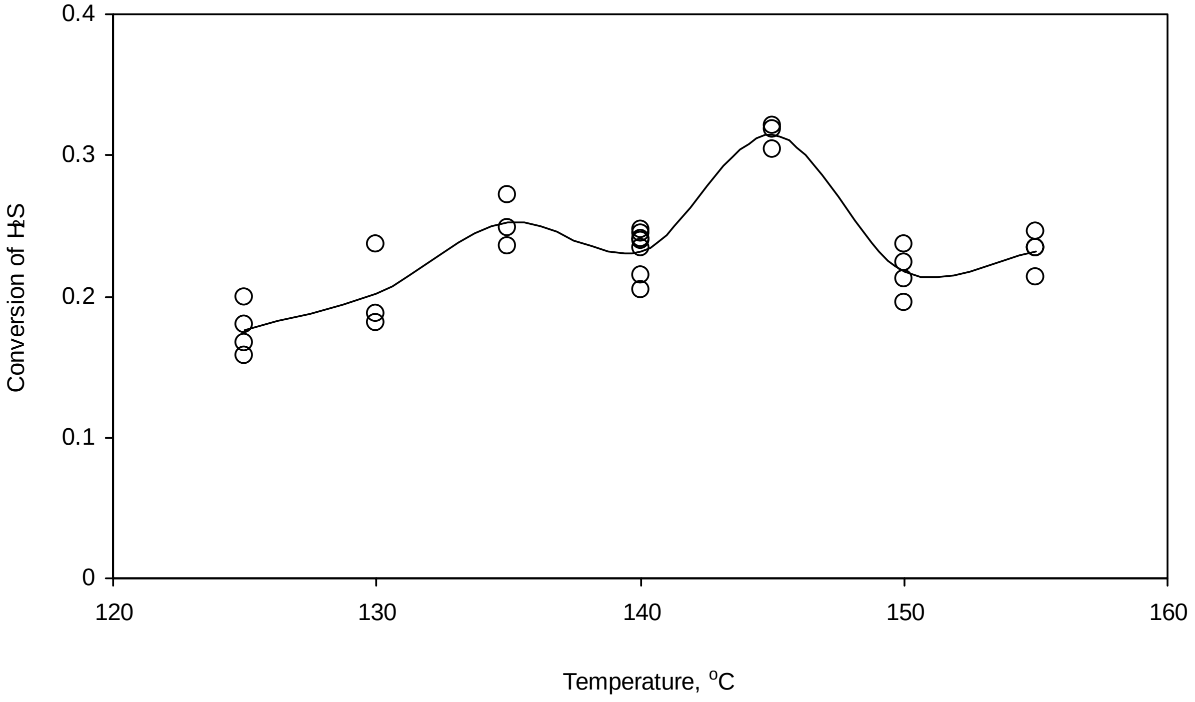
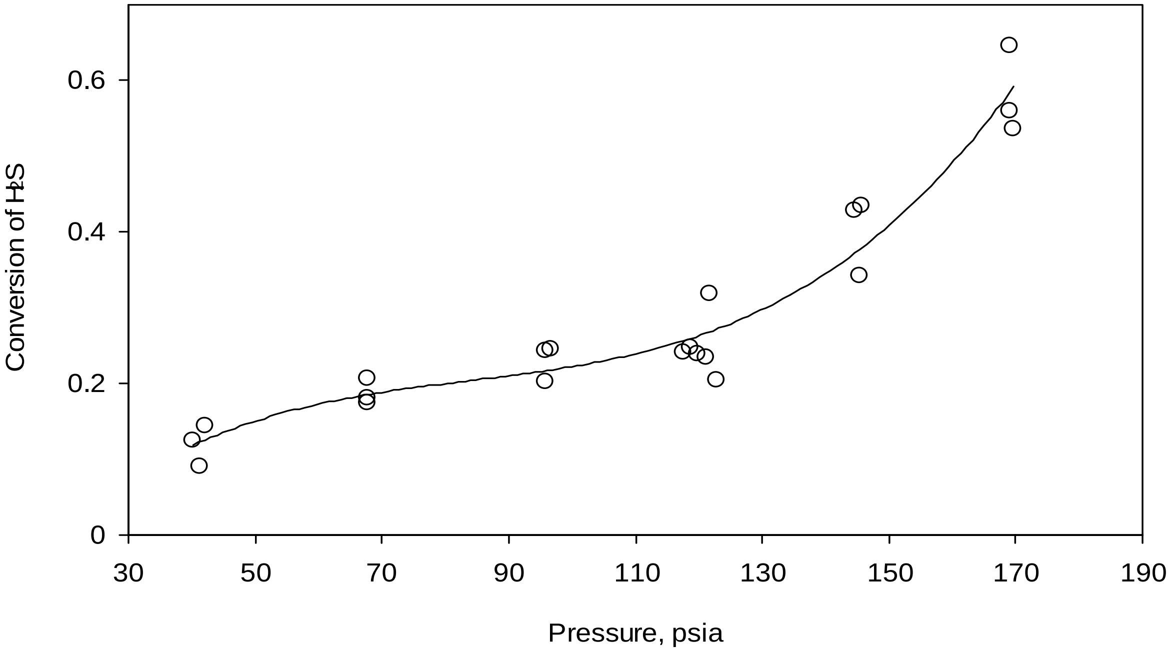


Figure 5. Effects of pressure on conversion of H_2S with a 50 cc/min feed stream containing 5000-ppm H_2S, 2500-ppm SO_2, 10-v% moisture, 70-v% H_2, and 0.02-g catalyst, at 140°C and 0.059 - 0.25 s space time.



Effects of Reaction Temperature on Conversion of H_2S to elemental sulfur

Experiments on conversion of hydrogen sulfide to elemental sulfur with 0.02-g catalyst were carried out for the space time range of 0.082 – 0.183 s to evaluate effects of reaction temperature on conversion of hydrogen sulfide to elemental sulfur at 125 - 155°C and 117 – 122 psia. Gas mixtures are fed to a micro bubble reactor containing 70-v% hydrogen, 5,000-ppmv H_2S, 2,500 ppmv SO_2, 10-v% moisture, and nitrogen as remainder. Volumetric feed rates of gas mixtures to the micro bubble reactor are 50 SCCM . Conversion of H_2S to elemental sulfur is 0.1 – 0.44. Conversion of H_2S to elemental sulfur does not follow the Arrhenius' equation. Reaction temperature affects conversion of H_2S to elemental sulfur. However, conversion of H_2S to elemental sulfur is highest at 145°C, while conversion of H_2S to elemental sulfur is lowest at 125°C over the reaction temperature range of 125 – 155°C (see Figure 4). Conversion of H_2S to elemental sulfur increases with increased reaction temperature over the temperature ranges of 125 – 135°C, 140 – 145°C, and 150 – 155°C, whereas conversion of H_2S to elemental sulfur decreases with increased reaction temperature over the temperature ranges of 135 – 140°C and 145 – 150°C

Effects of Pressure on Conversion of H_2S to elemental sulfur

Experiments on conversion of hydrogen sulfide to elemental sulfur with 0.02-g catalyst were carried out for the space time range of 0.059 – 0.25 s to evaluate effects of reaction pressure on conversion of hydrogen sulfide to elemental sulfur at 140°C and 40 -170 psia. A gas mixture fed to a micro bubble reactor contains 70-v% hydrogen, 5,000-ppmv H_2S, 2,500-ppmv SO_2, and 10-v% moisture, and nitrogen as remainder. Volumetric feed rates of the gas mixture to the micro bubble reactor are 50 SCCM. Conversion of H_2S to elemental sulfur is 0.05 – 0.65. Reaction pressure affects significantly conversion of H_2S to elemental sulfur in the pressure range of 40 -170 psia. Conversion of H_2S to elemental sulfur increases with reaction pressure over the pressure range of 40 -170 psia (see Figure 5). Conversion of H_2S to elemental sulfur increases moderately with increased total reaction pressure over the pressure range of 40 – 120 psia, whereas conversion of H_2S to elemental sulfur increases sharply with increased total reaction pressure over the pressure range of 120 – 170 psia.

Effects of H_2S and SO_2 Partial Pressures on Reaction Rates of H_2S

A reaction rate equation (see Equation 5) was developed with the following surface reaction mechanisms. Gaseous hydrogen sulfide is predominantly attached to active sites on the surface of catalyst particles, and then the H_2S attached to active sites on the surface of catalyst particles is reacted with gaseous SO_2 from a bulk gaseous reaction mixture to produce liquid elemental sulfur and water, as shown in Equation 2. Water on active sites, produced from the reaction of H_2S with SO_2, is mostly evaporated into the gaseous bulk reaction mixture.

$$-r_A' = \frac{kP_A^2 P_B}{(1+k_A P_A)^2} \tag{5}$$

where $-r_A'$: surface reaction rates of H_2S

k: surface reaction rate constant

k_A: equilibrium adsorption constant of H_2S on active sites of catalyst particles

P_A: partial pressure of H_2S

P_B: partial pressure of SO_2

Rearranging Equation 5 produces Equation 6. Experimental data in Table 6 obtained at 140°C, 40 – 123 psia, 0.059 - 0.178 s space time and 50 SCCM feed rate are applied to Equation 6 to determine the surface reaction rate constant k and the equilibrium adsorption constant k_A of H_2S.

$$\left(\frac{P_A^2 P_B}{-r_A'}\right)^{0.5} = \frac{1}{k^{0.5}} + \frac{k_A}{k^{0.5}} P_A \tag{6}$$

Partial pressures of H_2S and experimental surface reaction rates of H_2S determined with experimental data obtained in the reaction pressure range of 40 – 123 psia at 140°C, as shown in Tables 6 and 7, are applied to Equation 6. Left-side values of Equation 6 are plotted against partial pressures of H_2S, as shown in Figure 6. The value of a surface reaction rate constant k can be calculated from the intercept value of a linear regression line in Figure 6, whereas the value of an equilibrium adsorption constants k_A of H_2S on active sites of catalyst particles can be obtained from the slope value of the linear regression line. The value of the surface reaction rate constant k of H_2S and the value of the equilibrium adsorption constant k_A of H_2S, determined with the experimental data in Tables 6 and 7, are 0.037 g-mole/s-g-(psia)^3 and 55.64 psia^{-1}, respectively. The developed reaction model suggests that H_2S is strongly adsorbed onto active sites of catalyst particles in the preference over SO_2, and the reaction for conversion of H_2S to elemental sulfur is second order with respect to partial pressure of H_2S and first order with respect to partial pressure of SO_2. In other words, this reaction may be called element reaction.

Figure 6. Left-side values of the rearranged reaction model vs. partial pressures of H_2S.

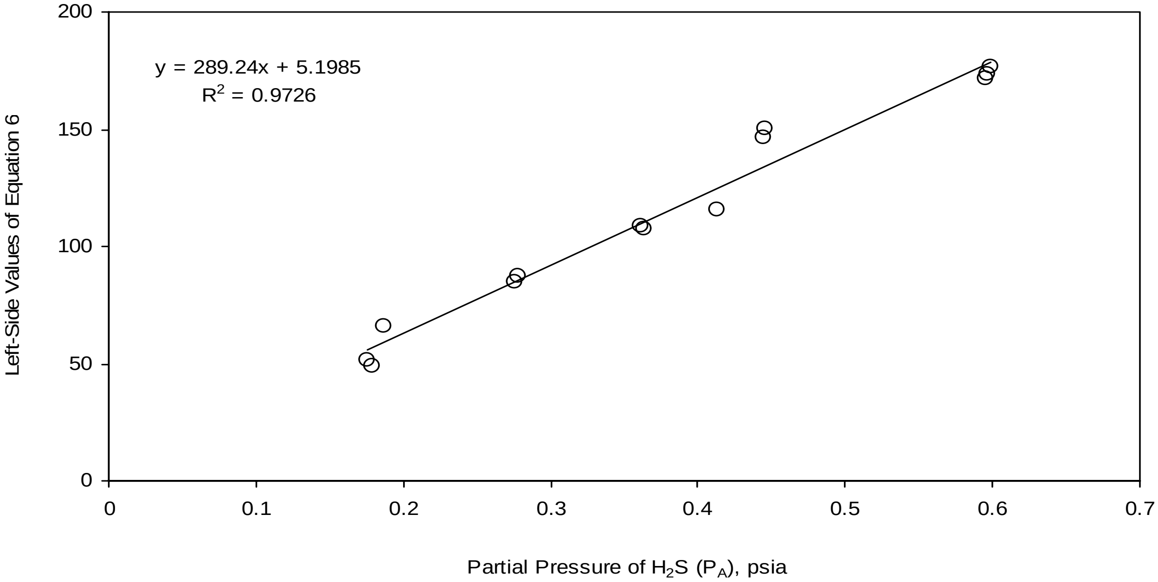
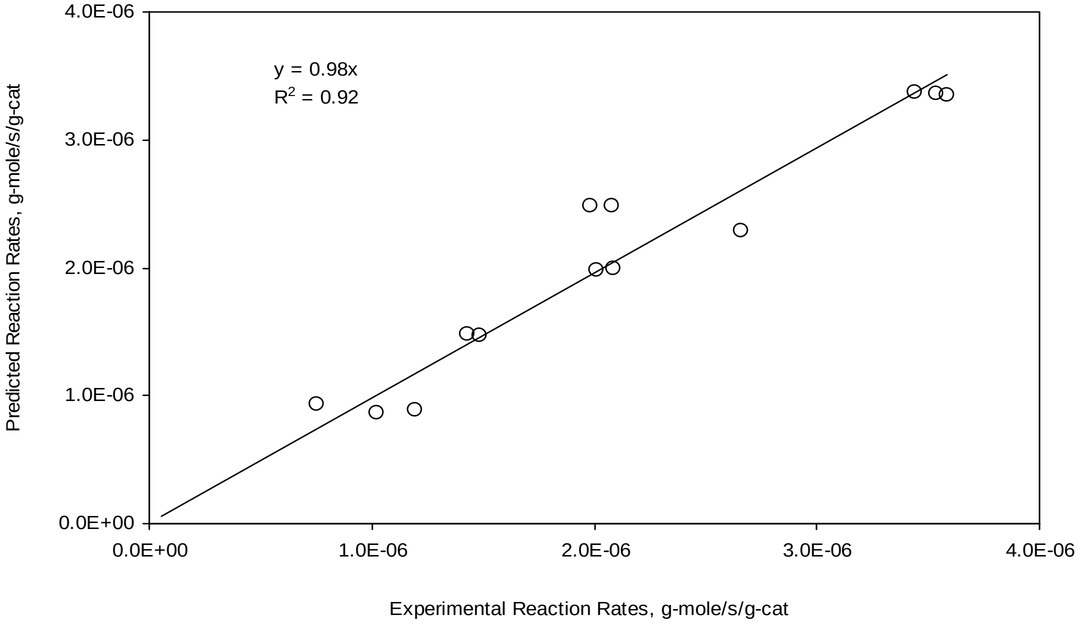


Figure 7. Predicted reaction rates vs. experimental reaction rates.



Predicted surface reaction rates for conversion of H_2S to elemental sulfur in Table 7 were calculated with the surface reaction rate constant k and the equilibrium absorption constant k_A of

H_2S determined by applying experimental data to the reaction model. Predicted surface reaction rates for conversion of H_2S to elemental sulfur were compared with experimental surface reaction rates for conversion of H_2S to elemental sulfur, calculated with experimental data of conversion of H_2S, catalyst amount, and molar feed rate of H_2S, as shown in Figure 7.

Figure 8. Effects of catalyst amount on conversion of 5000-ppm H_2S with 2500-ppm SO_2 wi th a 50-SCCM feed stream containing 10-v% moisture and 70-v% H_2 at 140°C, 115 - 123 psia and 0.169 - 0.849 s space time.

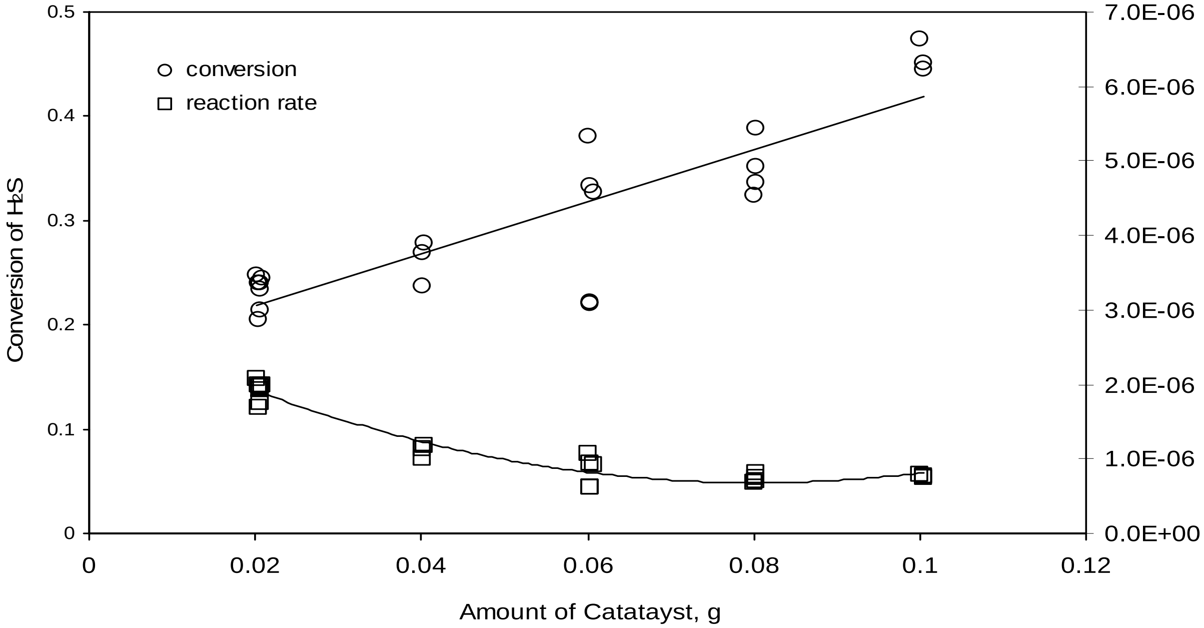
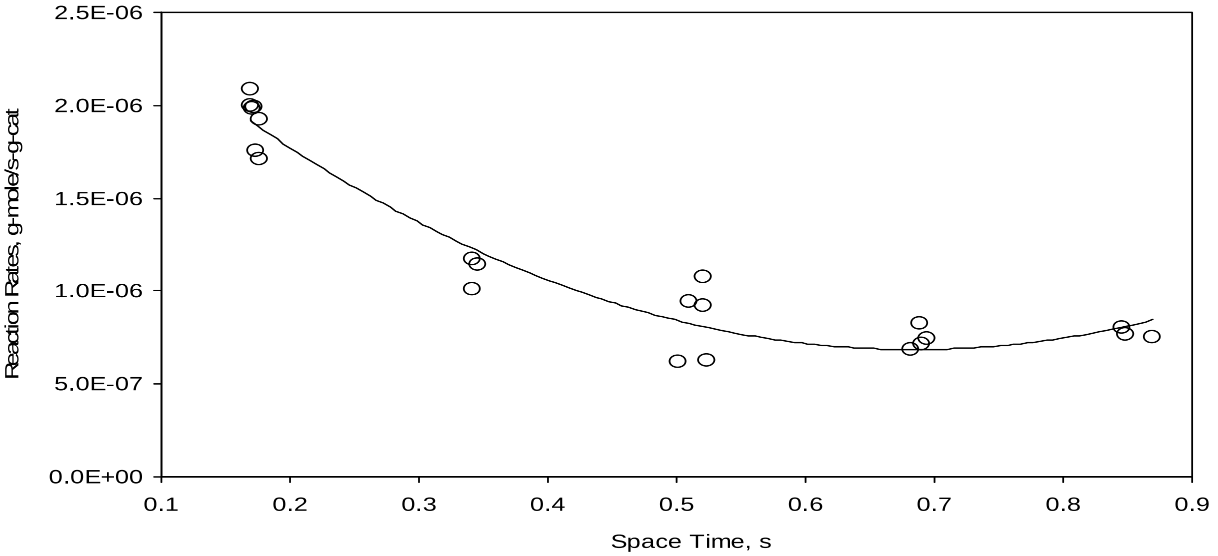


Figure 9. Effects of space time on surface reaction rates of H_2S with 0.02 - 0.1 g catalyst and a 50-SCCM feed stream containing 5,000-ppmv H_2S and 2,500-ppmv SO_2, 10-v% moisture, and 70-v% H_2 at 115 - 123 psia and 140°C.



Effects of catalyst loading and space time on conversion of H_2S

Effects of catalyst loading on conversion of H_2S to elemental sulfur were examined at 140°C and 115 - 123 psia. The feed gas mixture to a micro bubble reactor contains 5,000-ppmv H_2S, 2,500-ppmv SO_2, 10-v% moisture, and 70-v% H_2. The volumetric feed rate of the feed gas mixture to the micro bubble reactor is 50 SCCM. The amount range of fresh catalyst particles loaded in the micro bubble reactor is 0.02 – 0.1 g. Conversion of H_2S increases with catalyst loading. However, experimental reaction rate of H_2S, obtained with Equation 3, decreases with increased catalyst loading in the catalyst loading range of 0.02 – 0.06 g, whereas experimental reaction rate of H_2S appears to be independent of catalyst loading in the catalyst loading range of 0.06 – 0.1 g, as shown in Figure 8. This result may indicate that both surface reaction rate k and equilibrium adsorption constant k_A of H_2S for conversion of H_2S with SO_2 to elemental liquid sulfur in the catalyst loading range of 0.06 – 0.1 g are not significantly affected by amount of catalyst particles loaded in the micro bubble reactor. These data also indicate that experimental reaction rate of H_2S with SO_2 in the space time range of 0.05 –0.85 s be independent of space time of gaseous reaction mixtures in the micro bubble reactor, as shown in Figure 9.

CONCLUSIONS

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

- Reaction temperature affects conversion of H_2S to elemental sulfur, but conversion of H_2S to elemental sulfur does not follow the Arrhenius' equation. Conversion of H_2S to elemental sulfur is highest at 145°C, while conversion of H_2S to elemental sulfur is lowest at 125°C over the reaction temperature range of 125 – 155°C. Conversion of H_2S to elemental sulfur increases with increased reaction temperature over the temperature ranges of 125 – 135°C, 140 – 145°C, and 150 – 155°C, whereas conversion of H_2S to elemental sulfur decreases with increased reaction temperature over the temperature ranges of 135 – 140°C and 145 – 150°C

- Reaction pressure affects significantly conversion of H_2S to elemental sulfur in the pressure range of 40 -170 psia. Conversion of H_2S to elemental sulfur increases moderately with increased total reaction pressure over the pressure range of 40 – 120 psia, whereas conversion of H_2S to elemental sulfur increases sharply with increased total reaction pressure over the pressure range of 120 – 170 psia.

- A reaction rate equation for the conversion of H_2S to element sulfur in the presence of SO_2 over the total reaction pressure range of 40 – 120 psia was developed with the surface reaction mechanisms mentioned below. Gaseous hydrogen sulfide is predominantly attached to active sites on the surface of catalyst particles, and then the H_2S attached to active sites on the surface of catalyst particles is reacted with gaseous SO_2 from a bulk gaseous reaction mixture to produce liquid elemental sulfur and water. Water on active sites, produced from the reaction of H_2S with SO_2, is mostly evaporated into the gaseous bulk reaction mixture.

- The developed reaction model suggests that H_2S is strongly adsorbed onto active sites of catalyst particles in the preference over SO_2, and the reaction for conversion of H_2S to elemental sulfur is second order with respect to partial pressure of H_2S and first order with respect to partial pressure of SO_2 over the total reaction pressure range of 40 – 120 psia . In other words, this reaction may be called element reaction.

- Conversion of H_2S increases with catalyst loading. Experimental reaction rate of H_2S decreases with increased catalyst loading in the catalyst loading range of 0.02 – 0.06 g, whereas experimental reaction rate of H_2S appears to be independent of catalyst loading in the catalyst loading range of 0.06 – 0.1 g..

- Both surface reaction rate k and equilibrium adsorption constant k_A of H_2S for conversion of H_2S with SO_2 to elemental liquid sulfur in the catalyst loading range of 0.06 – 0.1 g are not significantly affected by amount of catalyst particles loaded in the micro bubble reactor. Experimental reaction rates of H_2S with SO_2 in the space time range of 0.05 –0.85 s appear to be independent of space time of gaseous reaction mixtures in the micro bubble reactor.

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PUBLICATIONS AND PRESENTATIONS

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