

# ENERGY

## Kinetics of Hot-Gas Desulfurization Sorbents for Transport Reactors

Annual Technical Progress Report for the Period  
October 1, 1999 to September 30, 2000

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January 2001

Work Performed Under Contract No  
DE-FG26-98FT40145

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

By  
Tuskegee University  
Tuskegee, Alabama 36088

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## CONTENTS

	Page
DISCLAIMER	iii
LIST OF TABLES	v
LIST OF FIGURES	vi
SUMMARY	1
INTRODUCTION	1
EXPERIMENTAL SETUPS AND PROCEDURES	2
THEORY	9
RESULTS AND DISCUSSION	10
Effects of Temperature on Initial Dynamic Absorption of H <sub>2</sub> S	12
Effects of Sorbent amounts on Initial Dynamic Absorption of H <sub>2</sub> S	14
Effects of Concentration of H <sub>2</sub> S on Initial Dynamic Absorption of H <sub>2</sub> S	17
Effects of Moisture Concentration on Initial Dynamic Absorption of H <sub>2</sub> S	19
Effects of Hydrogen Concentration on Initial Dynamic Absorption of H <sub>2</sub> S	19
CONCLUSIONS	21
REFERENCES	22
PUBLICATIONS	23

## LIST OF TABLES

Table		Page
1	Experimental conditions for the reaction of hydrogen sulfide with the sorbents.	3
2	Properties of the AHI-5 sorbent from the Research Triangle Institute (RTI).	3
4	Absorption of hydrogen sulfide into AHI-5 sorbent in the presence of hydrogen and moisture at various absorption conditions.	4
5	Surface reaction rate constants and intraparticle diffusivity values calculated with initial absorption data and the reaction model.	8

## LIST OF FIGURES

Figure		Page
1	A differential reactor assembly	2
2	Effects of reaction duration on absorption of 12000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.	11
3	Effects of temperature on absorption of 12000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture.	12
4	Effects of temperature on absorption of 12000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture, using the reaction model.	13
5	Effects of temperature on surface reaction rate constant and intraparticle diffusivity in the absorption of 12000-ppm H <sub>2</sub> S into AHI-5 sorbent with 10-v% H <sub>2</sub> and 10-v% moisture.	13
6	Effects of temperature on surface reaction rate constant in the absorption of 12000-ppm H <sub>2</sub> S into AHI-5 sorbent with 10-v% H <sub>2</sub> and 10-v% moisture, using the Arrhenius equation.	14
7	Effects of sorbent amount on absorption of 18000-ppm H <sub>2</sub> S on AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.	15
8	Effects of sorbent amount on absorption of 18000-ppm H <sub>2</sub> S on AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.	15
9	Effects of sorbent amount on surface reaction rate constant and intraparticle diffusivity in the absorption of 18000-ppm H <sub>2</sub> S into AHI-5 sorbent with 10-v% H <sub>2</sub> and 10-v% moisture at 450°C.	16
10	Effects of H <sub>2</sub> S concentration on absorption of H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.	16
11	Effects of H <sub>2</sub> S concentration on absorption of H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.	17
12	Effects of H <sub>2</sub> S concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H <sub>2</sub> S into 0.01-g AHI-5 sorbent with 10-v% H <sub>2</sub> and 10-v% moisture at 450°C.	17

## LIST OF FIGURES Continued-1

13	Effects of moisture concentration on absorption of 18000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen at 450°C and the space time 0.076 s.	18
14	Effects of moisture concentration on absorption of 18000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% hydrogen at 450°C and the space time 0.076 s, using the reaction model.	18
15	Effects of moisture concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H <sub>2</sub> S into 0.01-g AHI-5 sorbent with 10-v% H <sub>2</sub> at 450°C.	19
16	Effects of hydrogen concentration on absorption of 18000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C and the space time 0.076 s.	20
17	Effects of hydrogen concentration on absorption of 18000-ppm H <sub>2</sub> S on 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.	20
18	Effects of hydrogen concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H <sub>2</sub> S into 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C.	21

## SUMMARY

Hot-gas desulfurization for the integrated gasification combined cycle (IGCC) process has been investigated by many researchers to remove effectively hydrogen sulfide with various metal oxide sorbents at elevated temperatures. Various metal oxide sorbents are formulated with metal oxides such as Fe, Co, Zn, and Ti. Initial reaction kinetics of formulated sorbents with hydrogen sulfide is studied in the presence of various amounts of moisture and hydrogen at various reaction temperatures. The objectives of this research are to study initial reaction kinetics for a sorbent-hydrogen sulfide heterogeneous reaction system, to investigate effects of concentrations of hydrogen sulfide, hydrogen, and moisture on dynamic absorption of H<sub>2</sub>S into sorbents, and to evaluate effects of temperature and sorbent amounts on dynamic absorption of H<sub>2</sub>S into sorbents. Experimental data on initial reaction kinetics of hydrogen sulfide with metal oxide sorbents were obtained with a 0.83-cm<sup>3</sup> differential reactor. In this report, the reactivity of AHI-5 was examined. This sorbent was obtained from the Research Triangle Institute (RTI). The sorbent in the form of 70 μm particles are reacted with 9000 – 18000 ppm hydrogen sulfide at 350 – 500°C. The range of space time of reaction gas mixtures is 0.071 – 0.088 s. The range of reaction duration is 4 – 10800 s.

## INTRODUCTION

Coal is the most plentiful energy resource in the United States and continues to be the major fuel utilized by electrical power plants. The coal resources will become attractive for chemical feedstocks as well as for a source of liquid and gaseous fuels as petroleum resources become continually depleted.

Coal is a very complex and heterogeneous material. Effective utilization of coal requires an understanding of the formation, structure, and purification of coal. Raw coal consists of minerals and organic matter. The minerals include primarily pyrites, silicate (quartz, clays) and various minor minerals, whereas organic matter is composed of carbon, oxygen, hydrogen, nitrogen and sulfur. Sulfur in coal is present in organic, pyritic and sulfate forms. Although the sulfur occurring in the form of relatively coarse pyrite particles is removed largely by density separation, very finely disseminated pyrite and organic sulfur cannot be removed in this way. Sulfur products from combustion not only pollute the environment, but are also detrimental to combustion systems because of the corrosive action of their combustion products. In the gasification process, coal is reacted with steam and oxygen to produce a medium to high calorific value fuel gas. The fuel gas can be subsequently combusted in a gas/steam turbine combined cycle (IGCC). These IGCC-based power generation options offer much higher thermodynamic cycle efficiencies that promise to remove all forms of pollutants released from a coal conversion process through lower fuel consumption.

The removal of hydrogen sulfide from hot coal gas produced in the IGCC- based system by metal oxide sorbents is required to protect downstream combustion turbines from being corroded with sulfur compounds. The removal of sulfur compounds from coal gas products is investigated by using various metal oxide sorbents and membrane separation methods. Current

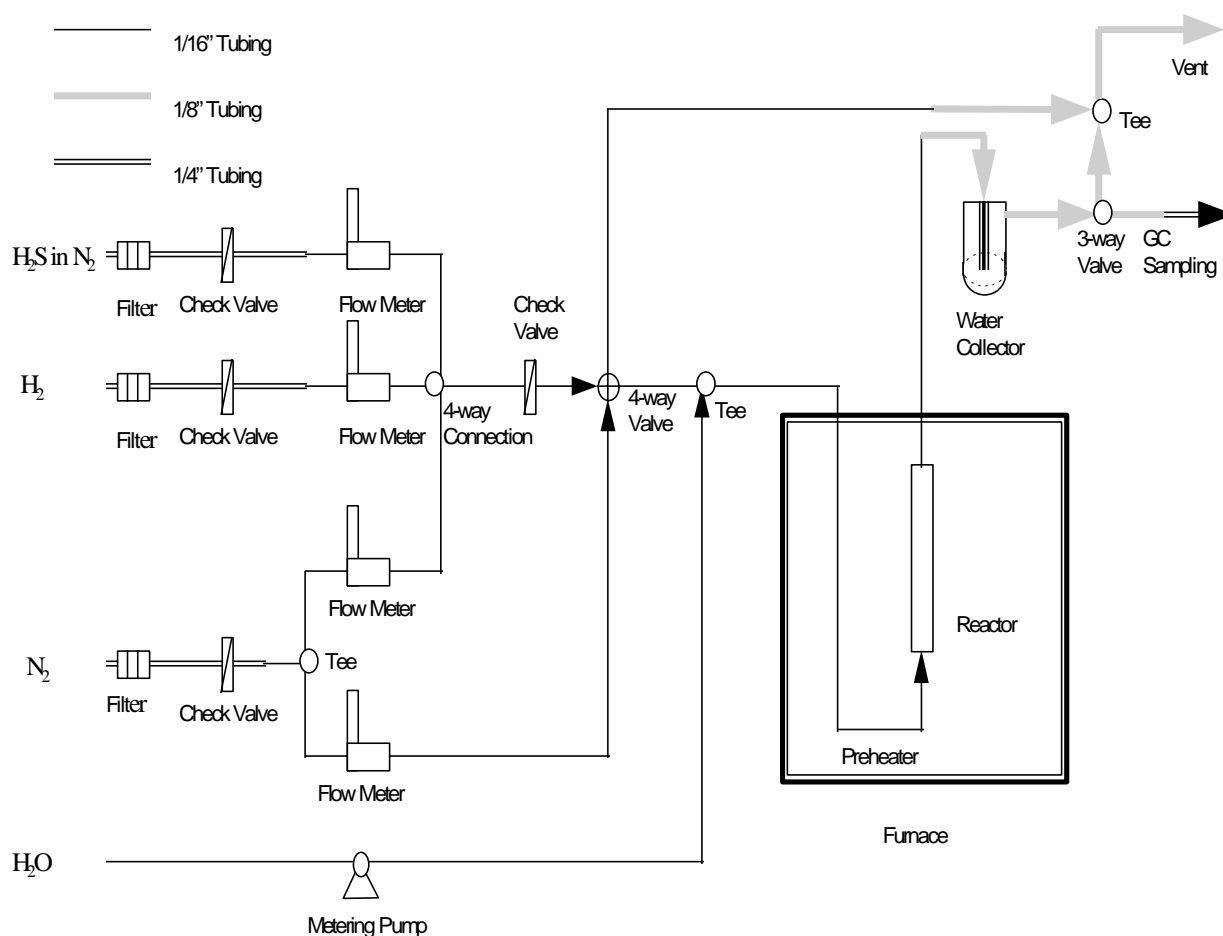


investigations show that the removal of sulfur with metal oxide sorbents appears to deliver the most promising results. The main requirements of these metal oxide sorbents are durability and a high sulfur-loading capacity during absorption-regeneration cycles.

## EXPERIMENTAL SETUPS AND PROCEDURES

A differential micro reactor was fabricated with a titanium grade-2 tube. The volume of reactor is  $0.83 \text{ cm}^3$ . The RTI AHI-5 sorbent in the form of  $70\text{-}\mu\text{m}$  spherical particles was examined. These sorbent particles were reacted with simulated coal gases containing hydrogen sulfide in the  $0.83 \text{ cm}^3$  titanium Grade-2 differential micro reactor for 4 – 10800 seconds at  $350\text{--}500^\circ\text{C}$ . Absorption amounts of hydrogen sulfide into the sorbent were analyzed with the LECO Sulfur Analyzer. The range of space (residence) time of the reaction gas mixture in the reactor was  $0.071\text{--}0.088 \text{ s}$  under the reaction conditions.

Figure 1. A Differential Reactor Assembly



A differential reactor assembly mainly consists of four mass flow meters for gases, one differential reactor, one preheater, one high pressure liquid pump for water, one four-way switch valve, one muffle furnace, three filters for gases, four check valves, and one water collection bottle (see Figure 1). The differential reactor is fabricated with one 5-cm-long ¼-inch titanium grade-2 tubing and two ¼” external column end fittings. The preheater is made of 1/16” Hastelloy C tubing. The reactor was loaded with 70 µm sorbent particles. The reactor, loaded with the sorbent particles, was placed inside the muffle furnace to be heated at a desired temperature. Nitrogen was introduced into the sorbent-loaded reactor during preheating the reactor. When the temperature of the reactor was raised at the desired temperature, a simulated coal gas mixture, containing H<sub>2</sub>S, was introduced into the reactor for desired reaction duration, by switching nitrogen with the simulated coal gas mixture. A reaction was terminated at desired reaction duration, and then absorption of H<sub>2</sub>S into the sorbent was analyzed with the LECO sulfur analyzer. These raw experimental data were shown in Table 3. The typical reaction conditions are shown in Table 1. The properties of the sorbents are shown in Table 2.

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

Reactor Volume, cm <sup>3</sup> :	0.83
Temperature, °C:	350 – 500
Reaction Pressure, psia	40.7
Reaction Time, s:	4 – 10800
Space Time under the absorption conditions, s:	0.071 – 0.088
Mean Particle Size, µm	70
Amount of Sorbent, g	0.005 – 0.02
Gas Flow Rate, cc/min at room temperature and 1 atm	750
Hydrogen, vol %	0 – 27
Moisture, vol %:	5 – 20
Concentration of H <sub>2</sub> S, ppm	9000 – 18000
Nitrogen, vol %	Remainder

Table 2. Properties of the sorbents from the Research Triangle Institute (RTI).

Sorbent	AHI-5
BET Area, m <sup>2</sup> /g	86
Avg. Pore Diameter, Å, based on volume	500
Density, g/cm <sup>3</sup>	2.077
Pore Volume, cm <sup>3</sup> /g	0.224
Mean Particle Size, µm	70
Composition	75 w% Fe <sub>2</sub> O <sub>3</sub> 15 w% ZnO 10 w% Inerts

Table 3. Absorption of hydrogen sulfide into AHI-5 sorbent in the presence of hydrogen and moisture at various absorption conditions.

Run #	Sorbent Amount, g	Reaction Duration, s	Sulfur Absorption, w%	Reaction Temperature, °C	Hydrogen Sulfide, ppm	Hydrogen v%	Moisture v%	Space Time, s	Reaction Rate Constant, cm/s	Intraparticle Diffusivity, cm <sup>2</sup> /s
1075	0.0108	4.5	0.53	450	12000	10	10	0.076	1.2034	0.002868
1076	0.0101	8.3	4.002							
1077	0.0108	12.27	7.703							
1078	0.0103	16.11	9.123							
1079	0.0105	20.29	11.89							
1080	0.0105	25.28	14.02							
1081	0.0102	30.41	16.88							
1082	0.0108	60.16	24.64							
1083	0.0105	180.16	29.02							
1093	0.0104	4.31	0.6983	400	12000	10	10	0.081	0.8832	0.001731
1084	0.0107	3.94	1.443							
1085	0.0104	8.24	2.211							
1086	0.0109	12.23	4.436							
1087	0.0104	16.13	7.772							
1088	0.0105	20.3	7.492							
1089	0.0105	25.1	12.57							
1090	0.0108	30.06	7.671							
1091	0.0107	60.07	22.3							
1092	0.0106	180.41	26.8							
1094	0.0109	4.37	1.075	350	12000	10	10	0.088	0.1591	0.000222
1095	0.0108	8.41	1.218							
1096	0.0107	12.1	1.348							
1097	0.0106	16.21	1.687							
1098	0.0107	20.37	1.579							
1099	0.0106	25.42	2.577							
1100	0.0104	30.27	2.508							
1101	0.0105	60.27	5.764							
1102	0.01	180.17	16.41							
1122	0.0102	360.2	22.73							
1104	0.0106	4.28	1.688	500	12000	10	10	0.071	1.7153	0.001953
1105	0.0102	8.34	4.623							
1106	0.0105	12.46	9.144							
1107	0.0108	16.26	12.2							
1108	0.0103	20.28	15.19							
1109	0.0106	25.34	16.04							
1110	0.0104	30.38	17.09							
1111	0.0108	60.29	22.28							
1112	0.0108	180.17	27.5							
1113	0.0104	4.44	3.168	450	18000	10	10	0.076	0.9374	0.002317
1114	0.0102	8.15	8.023							

Table 3. continued-1

Run #	Sorbent Amount, g	Reaction Duration, s	Sulfur Absorption, w%	Reaction Temperature, °C	Hydrogen Sulfide, ppm	Hydrogen v%	Moisture v%	Space Time, s	Reaction Rate Constant, cm/s	Intraparticle Diffusivity, cm <sup>2</sup> /s
1115	0.0102	12.51	14.44							
1116	0.0103	16.2	15.57							
1117	0.0104	20.33	17.8							
1118	0.0108	25.42	20.46							
1119	0.0107	30.23	20.92							
1120	0.0104	60.34	26.83							
1121	0.0105	180.34	29.34							
1123	0.0056	4.36	3.886	450	18000	10	10	0.076	1.1134	0.001787
1124	0.0054	8.4	10.07							
1125	0.0053	12.46	14.34							
1126	0.0052	16.17	14.52							
1127	0.0057	20.2	22.63							
1128	0.0053	25.33	20.61							
1129	0.0054	30.18	21.36							
1130	0.0056	60.2	27.44							
1131	0.0056	180.47	29.56							
1132	0.0154	4.34	2.239	450	18000	10	10	0.076	0.8855	0.002505
1133	0.0153	8.6	7.733							
1134	0.0155	12.2	10.47							
1135	0.0152	16.28	13.71							
1136	0.0151	20.14	15.79							
1137	0.0152	25.24	19.41							
1138	0.0154	30.48	20.41							
1139	0.0156	60.2	25.84							
1140	0.015	180.27	28.74							
1141	0.0101	4.17	2.242	450	15000	10	10	0.076	0.9436	0.004041
1142	0.0108	8.27	5.645							
1143	0.0107	12.19	10.9							
1144	0.0103	16.15	13.18							
1145	0.0102	20.34	16							
1146	0.0106	25.12	16.9							
1147	0.0104	30.22	20.19							
1148	0.0104	60.14	25.79							
1149	0.0108	180.26	31.68							
1150	0.0103	4.39	2.017	450	9000	10	10	0.076	1.3843	0.003022
1151	0.0103	8.31	3.372							
1152	0.0101	12.41	5.578							
1153	0.0105	16.38	7.639							
1154	0.0107	20.14	10.84							
1155	0.0108	25.37	12.19							
1156	0.0102	30.14	13.78							
1157	0.0109	60.34	23.42							
1158	0.0104	180.24	29.17							
1159	0.02	4.51	2.29	450	18000	10	10	0.076	0.8583	0.001924
1160	0.0208	8.2	5.153							

Table 3. continued-2

Run #	Sorbent Amount, g	Reaction Duration, s	Sulfur Absorption, w%	Reaction Temperature, °C	Hydrogen Sulfide, ppm	Hydrogen v%	Moisture v%	Space Time, s	Reaction Rate Constant, cm/s	Intraparticle Diffusivity, cm <sup>2</sup> /s
1161	0.0207	12.24	9.176							
1162	0.0204	16.35	13.57							
1163	0.0206	20.29	14.37							
1164	0.0202	25.33	17.69							
1165	0.0205	29.87	19.61							
1166	0.0204	60.18	24.85							
1167	0.0201	180.47	26.74							
1168	0.0106	4.24	1.861	450	18000	10	20	0.076	0.9575	0.002519
1169	0.0105	8.16	5.797							
1170	0.0108	12.34	7.959							
1171	0.0107	16.4	10.62							
1172	0.0106	20.1	14.69							
1173	0.0107	31.46	20.87							
1174	0.0102	25.21	17.31							
1175	0.0102	60.14	26.69							
1176	0.0105	180.31	30.53							
1177	0.0103	4.3	4.379	450	18000	10	5	0.076	0.8174	0.001944
1178	0.0102	8.19	8.8							
1179	0.01	12.25	13.07							
1180	0.0104	16.16	15.95							
1181	0.0105	20.35	17.13							
1182	0.0107	25.06	20.71							
1183	0.0105	29.99	19.8							
1184	0.0105	60.14	25.41							
1185	0.0105	180.07	29.07							
1186	0.0103	4.21	1.907	450	18000	10	15	0.076	0.9071	0.002137
1187	0.0102	8.38	7.569							
1188	0.01	12.07	10.17							
1189	0.0104	16.08	12.13							
1190	0.0105	20.14	16.73							
1191	0.0107	25.16	17.07							
1192	0.0105	30.24	20.22							
1193	0.0105	60.14	26.07							
1194	0.0105	180.3	28.86							
1195	0.0107	4.24	2.421	450	15000	10	10	0.076	0.9933	0.002510
1196	0.0103	8.2	4.817							
1197	0.0106	12.17	10.86							
1198	0.0105	16.21	13.7							
1199	0.0102	20.35	13.82							
1200	0.0107	25.24	17.65							
1201	0.0103	30.17	19.24							
1202	0.0104	60.27	25.16							
1203	0.0101	180.02	29.13							
1204	0.0103	4.09	1.228	375	12000	10	10	0.085	0.2453	0.000622
1205	0.0104	8.12	1.869							

Table 3. continued-3

Run #	Sorbent Amount, g	Reaction Duration, s	Sulfur Absorption, w%	Reaction Temperature, °C	Hydrogen Sulfide, ppm	Hydrogen v%	Moisture v%	Space Time, s	Reaction Rate Constant, cm/s	Intraparticle Diffusivity, cm <sup>2</sup> /s
1206	0.0102	12.13	3.083							
1207	0.0102	16.42	3.451							
1208	0.0101	20.1	4.17							
1209	0.0106	25.02	6.56							
1210	0.0102	30.07	5.491							
1211	0.0104	60.31	10.11							
1212	0.0105	180.39	21.1							
1213	0.0103	4.17	2.196	475	12000	10	10	0.073	1.1354	0.002899
1214	0.0106	8.14	4.557							
1215	0.01	12.05	8.316							
1216	0.0101	16.11	11.53							
1217	0.0102	20.15	13.09							
1218	0.0101	25.17	16.56							
1219	0.0101	30	18.19							
1220	0.0103	59.93	23.42							
1221	0.0107	180.18	28.95							
1222	0.0103	4.23	1.245	425	12000	10	10	0.079	1.0484	0.002614
1223	0.01	8.27	3.604							
1224	0.0102	12.1	6.156							
1225	0.0106	16.06	9.076							
1226	0.0102	20.21	11.51							
1227	0.0104	25.21	14.85							
1228	0.0107	30.2	16.78							
1229	0.0109	60.28	23.18							
1230	0.01	180.33	28.69							
1231	0.0102	4.2	1.581	400	12000	10	10	0.081	0.7405	0.001863
1232	0.0104	8.12	2.556							
1233	0.0102	12.38	5.003							
1234	0.0104	16.17	6.465							
1235	0.0108	20.23	5.529							
1236	0.0108	25.32	10.77							
1237	0.0102	25.34	8.219							
1238	0.0102	30.14	13.04							
1239	0.0103	60.17	20.44							
1240	0.0102	180.21	28.66							
1241	0.0104	4.08	1.69	450	18000	20	10	0.076	0.8832	0.002365
1242	0.0101	8.3	5.236							
1243	0.0102	12.25	12.38							
1244	0.0101	16.9	15.75							
1245	0.0101	20.2	17.77							
1246	0.0103	25.15	15.46							
1247	0.0103	30.05	21.86							
1248	0.0104	60.29	26.01							
1249	0.0104	180.3	29.08							

Table 3. continued-4

Run #	Sorbent Amount, g	Reaction Duration, s	Sulfur Absorption, w%	Reaction Temperature, °C	Hydrogen Sulfide, ppm	Hydrogen v%	Moisture v%	Space Time, s	Reaction Rate Constant, cm/s	Intraparticle Diffusivity, cm <sup>2</sup> /s
1250	0.0103	4.03	3.092							
1251	0.0103	3.57	2.392	450	18000	27	10	0.076	0.8384	0.002021
1252	0.0104	8.23	6.747							
1253	0.0105	12.32	11.83							
1254	0.0105	16.24	12.72							
1255	0.0102	20.1	16.56							
1256	0.01	25.33	18.98							
1257	0.0106	30.12	19.93							
1258	0.0102	60.16	25.16							
1259	0.0106	180.17	28.68							
1260	0.0103	4.31	4.729	450	18000	15	75	0.076	0.8802	0.001971
1261	0.0103	8.14	6.653							
1262	0.0101	12.25	12.4							
1263	0.0106	16.13	15.39							
1264	0.0106	20.15	16.66							
1265	0.0103	25.3	18.87							
1266	0.0106	30.3	20.88							
1267	0.0105	60.3	25.98							
1268	0.0106	180.37	29.04							
1269	0.0106	4.19	0.842	450	18000	5	10	0.076	1.0054	0.002317
1270	0.0108	8.18	7.318							
1271	0.0104	11.92	11.69							
1272	0.0101	16.16	14.7							
1273	0.0106	20.3	17.86							
1274	0.0104	25.17	18.46							
1275	0.0108	30.34	20.5							
1276	0.0106	60.35	27.47							
1277	0.0106	180.14	29.31							
1278	0.0101	4.28	3.028							

Table 4. Surface reaction rate constants and intraparticle diffusivity values calculated with initial absorption data and the reaction model.

Temperature, °C	Sorbent Amount, g	Pressure, psia	Concentration, v%			Reaction Rate Constant cm/s	Intraparticle Diffusivity, cm/s <sup>2</sup>
			H <sub>2</sub> S	H <sub>2</sub>	H <sub>2</sub> O		
450	0.01	40.6	1.2	10	10	1.2034	0.0029
400	0.01	40.6	1.2	10	10	0.8832	0.0017
350	0.01	40.6	1.2	10	10	0.1591	0.0002
500	0.01	40.6	1.2	10	10	1.7153	0.0020
450	0.01	40.6	1.8	10	10	0.9374	0.0023

Table 4. Continued-1

Temperature, °C	Sorbent Amount, g	Pressure, psia	Concentration, v%			Reaction Rate Constant cm/s	Intraparticle Diffusivity, cm/s <sup>2</sup>
			H <sub>2</sub> S	H <sub>2</sub>	H <sub>2</sub> O		
450	0.005	40.6	1.8	10	10	1.1134	0.0018
450	0.015	40.6	1.8	10	10	0.8855	0.0025
450	0.01	40.6	1.5	10	10	0.9436	0.0040
450	0.01	40.6	0.9	10	10	1.3843	0.0030
450	0.02	40.6	1.8	10	10	0.8583	0.0019
450	0.01	40.6	1.8	10	20	0.9575	0.0025
450	0.01	40.6	1.8	10	5	0.8174	0.0019
450	0.01	40.6	1.8	10	15	0.9071	0.0021
450	0.01	40.6	1.5	10	10	0.9933	0.0025
375	0.01	40.6	1.2	10	10	0.2453	0.0006
475	0.01	40.6	1.2	10	10	1.1354	0.0029
425	0.01	40.6	1.2	10	10	1.0484	0.0026
400	0.01	40.6	1.2	10	10	0.7405	0.0019
450	0.01	40.6	1.8	20	10	0.8832	0.0024
450	0.01	40.6	1.8	27	10	0.8384	0.0020
450	0.01	40.6	1.8	15	10	0.8802	0.0020
450	0.01	40.6	1.8	5	10	1.0054	0.0023

## THEORY

Levenspiel[1] described a reaction equation for a gas-solid reaction of spherical particles, using three reaction rate-controlling steps such as mass transfer of gaseous reactants through gaseous reaction mixtures, diffusion of reactants through porous solid reactants, and surface reaction of gaseous reactants with solid reactants (see Equation 1). Equation 1 was modified to obtain mass transfer rate of H<sub>2</sub>S, intraparticle diffusivity of H<sub>2</sub>S, and reaction rate of H<sub>2</sub>S with the sorbent using the physical properties (see Table 2), and the experimental data (see Table 3). This equation is developed under the assumptions that the shrinking core model and a quasi-steady state are valid.

$$\left(-\frac{d\left(\frac{r_c}{R}\right)}{dt}\right)^{-1} = \left(\frac{C_s R^2}{C_G}\right) \left(\frac{1}{Rk_g} - \frac{1}{d}\right) \left(\frac{r_c}{R}\right)^2 + \left(\frac{C_s R^2}{C_G d}\right) \left(\frac{r_c}{R}\right) + \left(\frac{C_s R}{C_G k_b}\right) \quad (1)$$

Equation 2 is obtained from Equation 1, when mass transfer rates are much higher than both diffusion rates of gaseous reactants and reaction rates of solid reactants. Equation 2 was applied to the experimental data shown in Table 3 to obtain both intraparticle diffusivity of H<sub>2</sub>S and reaction rate of H<sub>2</sub>S with the AHI-5 sorbent.

$$\left(-\frac{d\left(\frac{r_c}{R}\right)}{dt}\right)^{-1} = \left(\frac{C_s R^2}{C_G d}\right) \left(\frac{r_c}{R} - \left(\frac{r_c}{R}\right)^2\right) + \left(\frac{C_s R}{C_G k_b}\right) \quad (2)$$



Equations 3 and 6 are obtained from Equation 1, when both intraparticle diffusion controlling step and mass transfer controlling step of gaseous reactants are negligible.

Equation 7 is obtained from Equation 1, when both surface reaction step and mass transfer controlling step of gaseous reactants are negligible.

$$\left( -\frac{d\left(\frac{r_c}{R}\right)}{dt} \right)^{-1} = \left( \frac{C_s R}{C_G k_b'} \right) \quad (3)$$

$$t = \left( \frac{C_s R}{C_G k_b'} \right) \left( 1 - \frac{r_c}{R} \right) \quad (4)$$

$$\left( \frac{r_c}{R} \right) = (1 - x_B)^{1/3} \quad (5)$$

$$t = \left( \frac{C_s R}{C_G k_b'} \right) \left( 1 - (1 - x_B)^{1/3} \right) \quad (6)$$

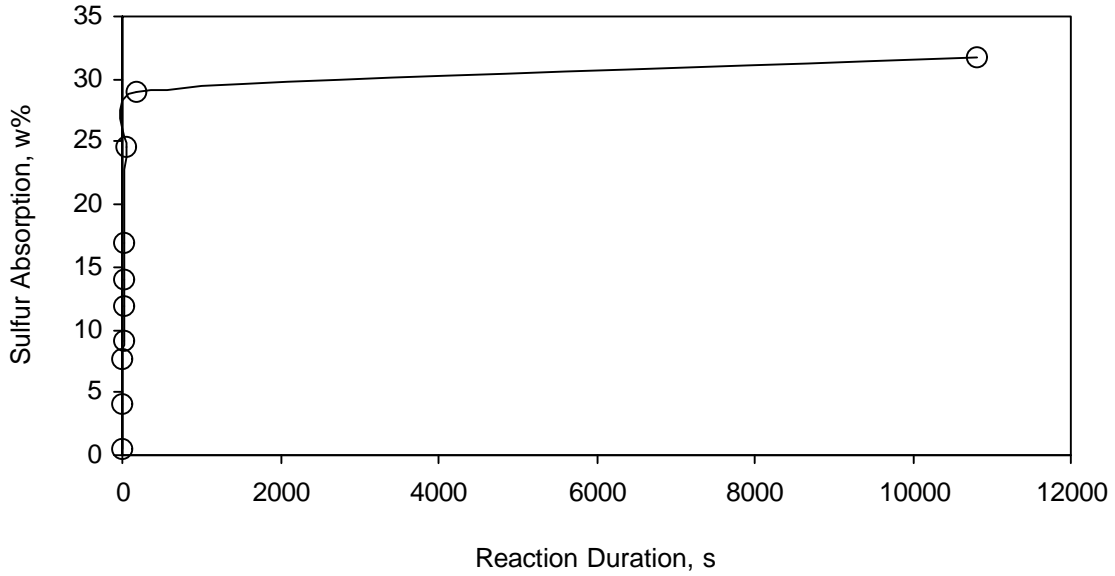
$$t = \left( \frac{C_s R^2}{6 C_G d} \right) \left( 1 - 3(1 - x_B)^{2/3} + 2(1 - x_B) \right) \quad (7)$$

where	$r_c$ : radius of an unreacted core $R$ : radius of spherical sorbent $t$ : reaction time $C_G$ : concentration of gaseous reactant $x_B$ : conversion of solid reactant	$k_b'$ : mass transfer rate of gaseous reactant $C_s$ : molar concentration of solid reactant $d$ : intraparticle diffusivity of gaseous reactant $k_b'$ : reaction rate constant
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## RESULTS AND DISCUSSION

Experiments on absorption of hydrogen sulfide into the AHI-5 sorbent was carried out for 4 – 10800 seconds at 350 - 500°C (see Table 1) to evaluate effects of concentration of H<sub>2</sub>S, H<sub>2</sub> and moisture, temperature, and sorbent amount on initial dynamic absorption of hydrogen sulfide into the sorbent. A typical simulated coal gas mixture consists of 12000-ppm hydrogen sulfide, 10-vol % moisture, 10 vol % hydrogen, and nitrogen as remainder. A volumetric feed rate of a simulated coal gas mixture to the reactor is 750 cm<sup>3</sup>/min at room temperature and atmospheric pressure. The temperature of the reactor is controlled in the furnace oven at 350 - 500°C. The pressure of the reactor is maintained at 40.7 psia. The total capacity of the sorbent for absorption of H<sub>2</sub>S is determined with the experimental data shown in Figure 2. Most reaction of H<sub>2</sub>S with the sorbent appears to occur for the first 3-min reaction duration. The entire reaction experimental data for this report were obtained for the 3-min reaction duration.

Figure 2. Effects of reaction duration on absorption of 12000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.



Raw experimental data on absorption of H<sub>2</sub>S into the sorbent, obtained at various reaction durations and experimental conditions, were shown in Figures 3, 7, 10, 13 and 16.

Conversion of the sorbent,  $x_B$ , is calculated by dividing absorption of H<sub>2</sub>S into the sorbent with the total absorption capacity of the sorbent. The ratio of the radius of an unreacted core  $r_c$  to the radius of the sorbent  $R$ ,  $r_c/R$ , is obtained with the conversion value of the sorbent (see Equation 5)

The values of  $r_c/R$  are plotted against absorption durations, and then the values of  $(-d(r_c/R)/dt)^{-1}$  in the left-hand side of Equation 2 are obtained from the values of the slopes of this plotted curve at various reaction durations.

The values of  $(-d(r_c/R)/dt)^{-1}$  as dependent variable and the values of  $(r_c/R - (r_c/R)^2)$  as independent variable, as shown in Equation 2, are plotted, as shown in Figures 4, 8, 11, 14 and 17, and then are applied to the linear least squares method to find both the value of the slope and the value of the intercept of this linear curve.

The value of the slope, obtained from the above-mentioned linear least squares method, is equal to the value of  $(C_S R^2 / (C_G \delta))$  in Equation 2. The value of intraparticle diffusivity  $\delta$  can be computed with the value of the sorbent radius ( $R$ ), the value of the H<sub>2</sub>S concentration ( $C_G$ ), and the total absorption capacity of the sorbent ( $C_S$ ). The values of intraparticle diffusivity  $\delta$  are shown in Table 4.

The value of the intercept, obtained from the above-mentioned linear least squares method, is equal to the value of  $(C_S R / (C_G k_b'))$  in Equation 2. The value of surface reaction rate constant  $k_b'$  can be computed with the value of the sorbent radius ( $R$ ), the value of the  $H_2S$  concentration ( $C_G$ ), and the total absorption capacity of the sorbent ( $C_S$ ). The values of surface reaction rate constant  $k_b'$  are shown in Table 4.

### Effects of Temperature on Initial Dynamic Absorption of $H_2S$

Experiments on initial absorption of  $H_2S$  into the sorbent were carried out for 4 – 180 seconds at 40.7 psia and 350 - 500°C to find effects of temperature on initial dynamic absorption of  $H_2S$  into the sorbent. The concentrations of both moisture and hydrogen are maintained at 10-volume percent. The concentration of hydrogen sulfide in the simulated coal gas mixture is maintained at 12000 ppm.

Initial absorption of  $H_2S$  into the AHI-5 sorbent is affected with absorption temperature for short absorption duration of 4 - 180 seconds (see Figure 3). Equation 2 was applied to the experimental data to obtain surface reaction rate constants of  $H_2S$  with the sorbent and intraparticle diffusivity values through pores of the sorbent (see Figure 4). The range of surface reaction rate constant is 0.16 – 1.2 cm/s (see Table 4). The range of intraparticle diffusivity values of the sorbent is 0.0002 – 0.0029  $cm^2/s$ . These observations may indicate that reactivity of the AHI-5 sorbent be dependent on reaction temperature in the temperature range of 350 – 500°C. However, reactivity of the sorbent is much lower at 350°C than that at 500°C. These facts also suggest that the controlling steps for the initial absorption of  $H_2S$  into sorbent particles are surface reaction of  $H_2S$  with reactive sites of sorbent particles and intraparticle diffusivity through pores of the sorbent.

Figure 3. Effects of temperature on absorption of 12000-ppm  $H_2S$  on 0.01-g AHI-5 sorbent with 10-v-% hydrogen and 10-v-% moisture.

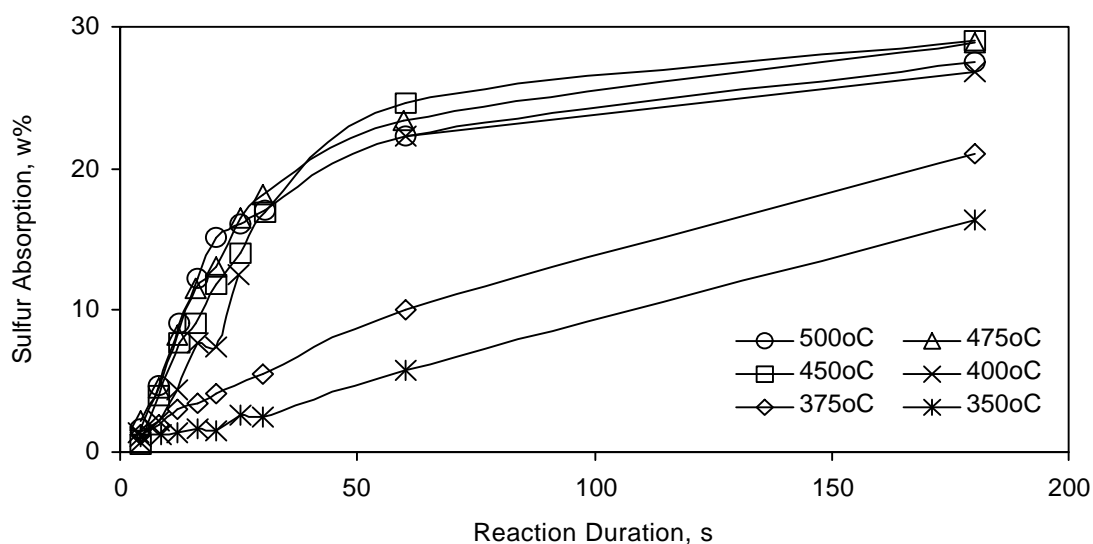
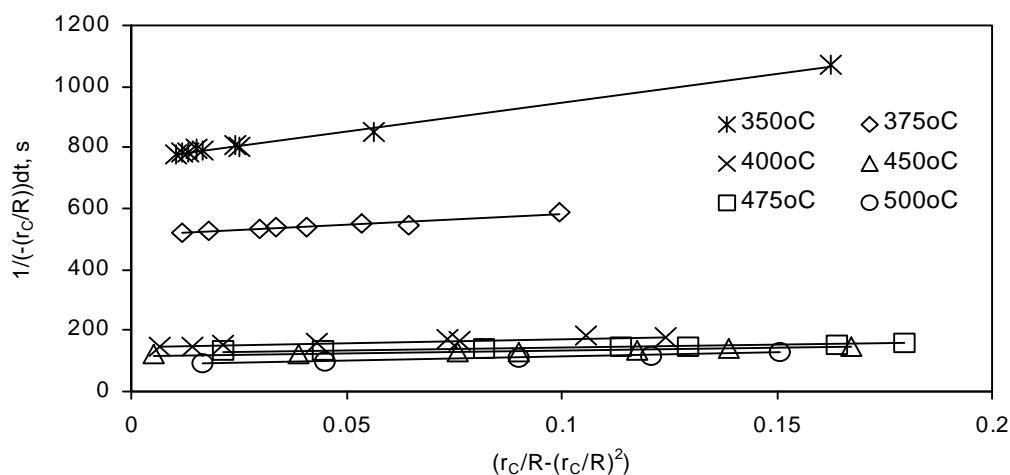
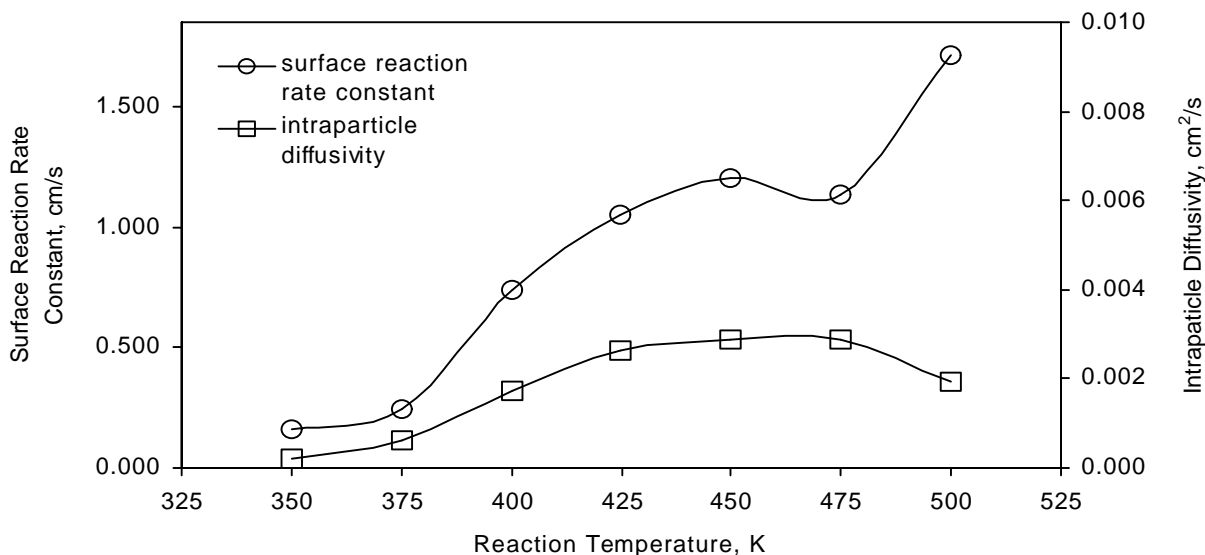


Figure 4. Effects of temperature on absorption of 12000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture, using the reaction model.



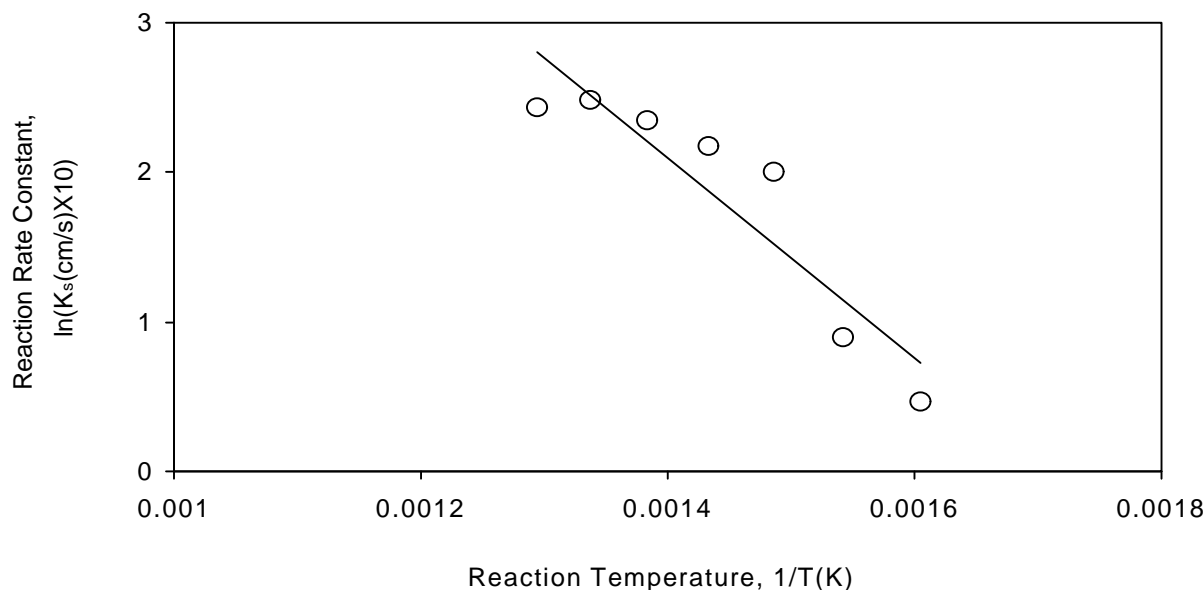
The surface reaction rate constant and the intraparticle diffusivity value increases with the reaction temperatures up to 450°C. However, the surface reaction rate constant increases steeply, and the intraparticle diffusivity value decreases in the temperature range of 475 – 500°C. These facts may indicate that collapsing the pore structure of the sorbent results in the decreased intraparticle diffusivity value in the temperature range of 475 – 500°C. The surface reaction rate constant and the intraparticle diffusivity value levels off in the temperature range of 425 – 475°C. This observation may indicate that both surface reaction rate constant and intraparticle diffusivity value appear to independent of absorption temperature.

Figure 5. Effects of temperature on surface reaction rate constant and intraparticle diffusivity in the absorption of 12000-ppm H<sub>2</sub>S into AHI-5 sorbent with 10-v% H<sub>2</sub> and 10-v% moisture.



The surface reaction rate constants, obtained from the reaction model, are applied to the Arrhenius equation (see Figure 6). The surface reaction rate constants appear to agree fairly with the equation in the temperature range of 350 – 475°C. However, the surface reaction rate constant at 500°C is not agreeable with the equation, and is not included in Figure 6.

Figure 6. Effects of temperature on surface reaction rate constant in the absorption of 12000-ppm H<sub>2</sub>S into AHI-5 sorbent with 10-v% H<sub>2</sub> and 10-v% moisture, using the Arrhenius equation.



### Effects of Sorbent amounts on Initial Dynamic Absorption of H<sub>2</sub>S

Experiments on effects of sorbent amounts on initial dynamic absorption of H<sub>2</sub>S into the sorbent were conducted for the reaction duration of 4 – 180 seconds at 450°C and the space-time of 0.076 s. Amounts of the AHI-5 sorbent affect slightly initial dynamic absorption of H<sub>2</sub>S into the sorbent in the sorbent amount range of 0.005– 0.02 g (see Figure 7). Equation 2 was applied to the experimental data to obtain surface reaction rate constants and intraparticle diffusivity values of H<sub>2</sub>S (see Figure 8). The range of the surface reaction rate constant is 0.86 – 1.11 cm/s, and the range of the intraparticle diffusivity values is 0.0018 – 0.0025 cm<sup>2</sup>/s. The concentrations of both hydrogen and moisture were maintained at 10-v percent. The concentration of H<sub>2</sub>S was maintained at 18000 ppm. Absorption of H<sub>2</sub>S into the sorbent increases with decreased amount of the sorbent in the reactor (see Figure 7). The surface reaction rate constant decrease with increased amount of the sorbent. The intraparticle diffusivity values increase generally with increased amount of the sorbent (see Figure 9).

These observations may suggest that mass transfer of H<sub>2</sub>S from the bulk gaseous phase to the surface of the solid sorbent affects somewhat the overall reaction of H<sub>2</sub>S with the sorbent

Figure 7. Effects of sorbent amount on absorption of 18000-ppm H<sub>2</sub>S on AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.

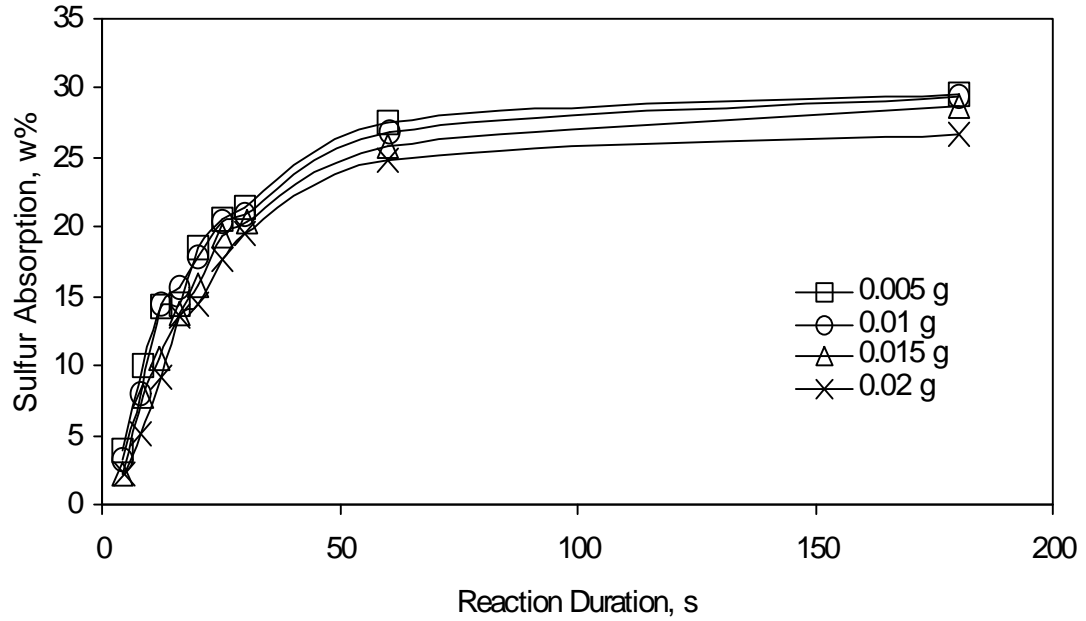


Figure 8. Effects of sorbent amount on absorption of 18000-ppm H<sub>2</sub>S on AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.

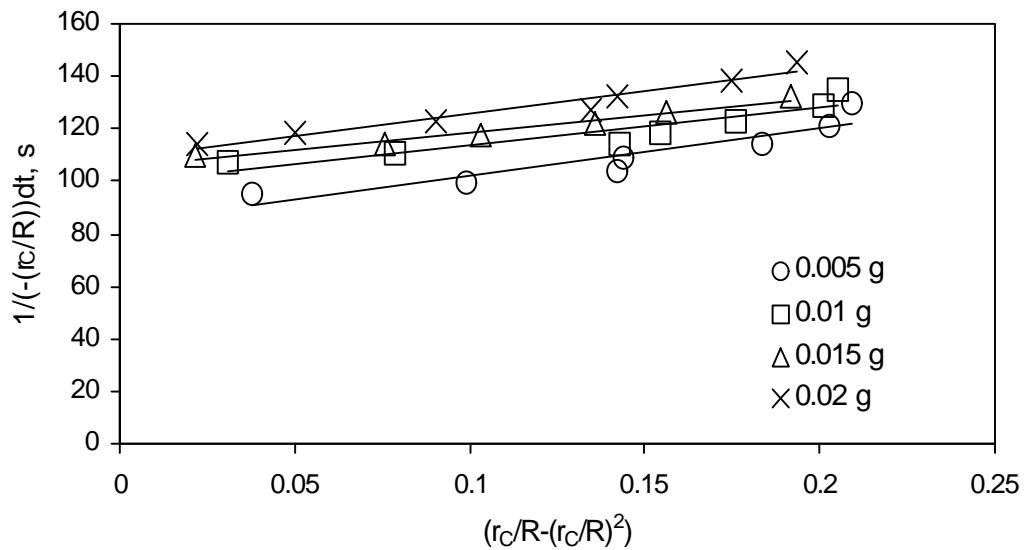


Figure 9. Effects of sorbent amount on surface reaction rate constant and intraparticle diffusivity in the absorption of 18000-ppm H<sub>2</sub>S into AHI-5 sorbent with 10-v% H<sub>2</sub> and 10-v% moisture at 450°C.

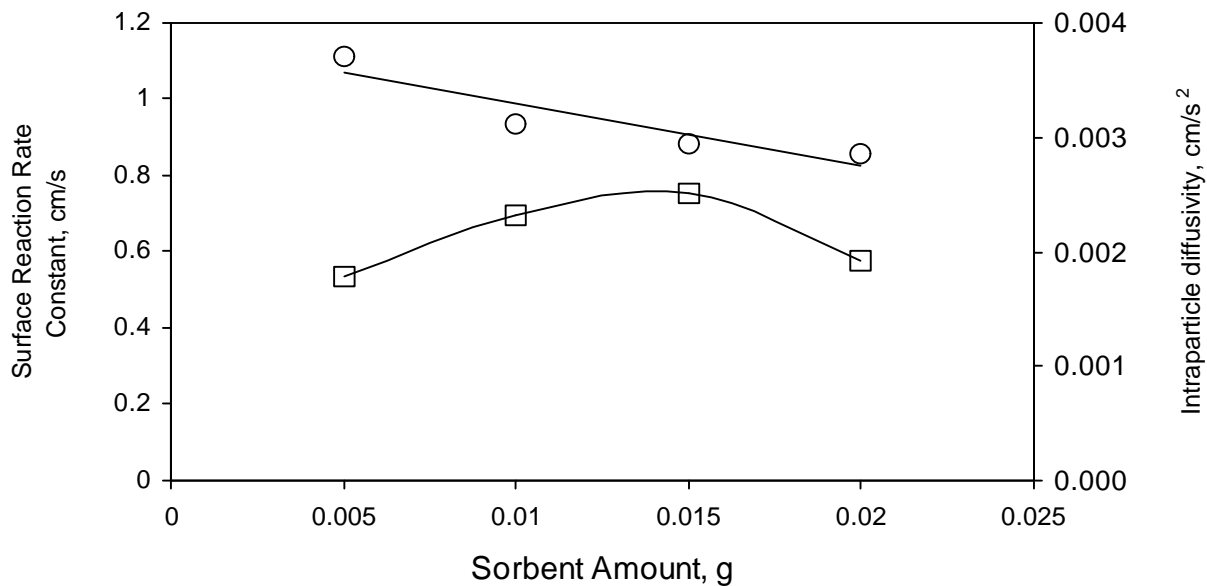
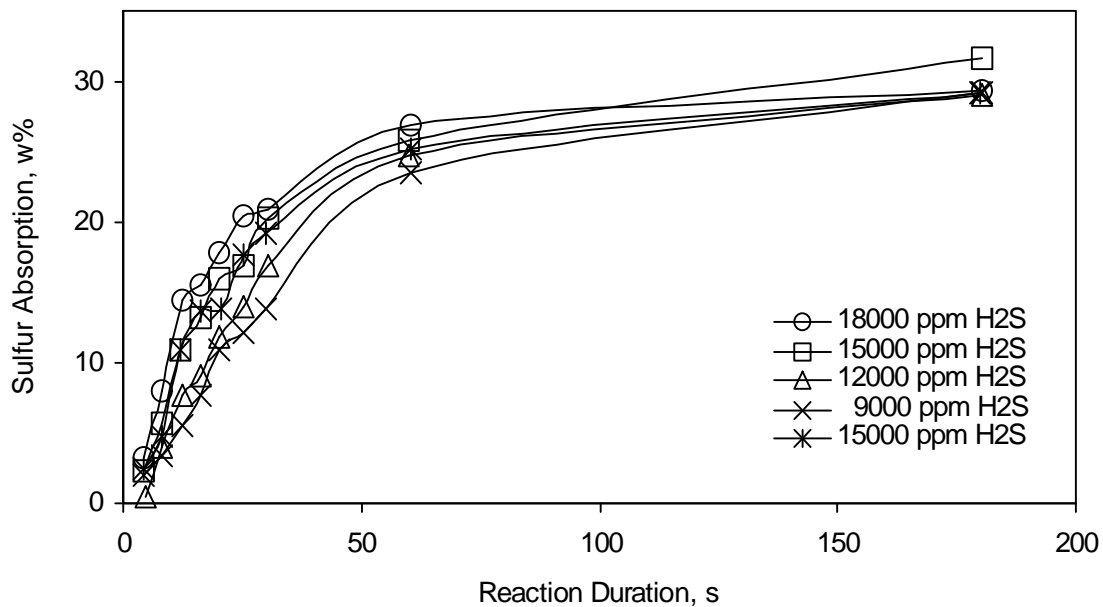


Figure 10. Effects of H<sub>2</sub>S concentration on absorption of H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s.



## Effects of Concentration of H<sub>2</sub>S on Initial Dynamic Absorption of H<sub>2</sub>S

Experiments on effects of concentration of H<sub>2</sub>S on initial dynamic absorption of H<sub>2</sub>S into the sorbents were conducted for the reaction duration of 4 – 180 seconds at 450°C and the space-time of 0.076 s. The concentrations of hydrogen and moisture were maintained at 10-volume percent, respectively

Figure 11. Effects of H<sub>2</sub>S concentration on absorption of H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen and 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.

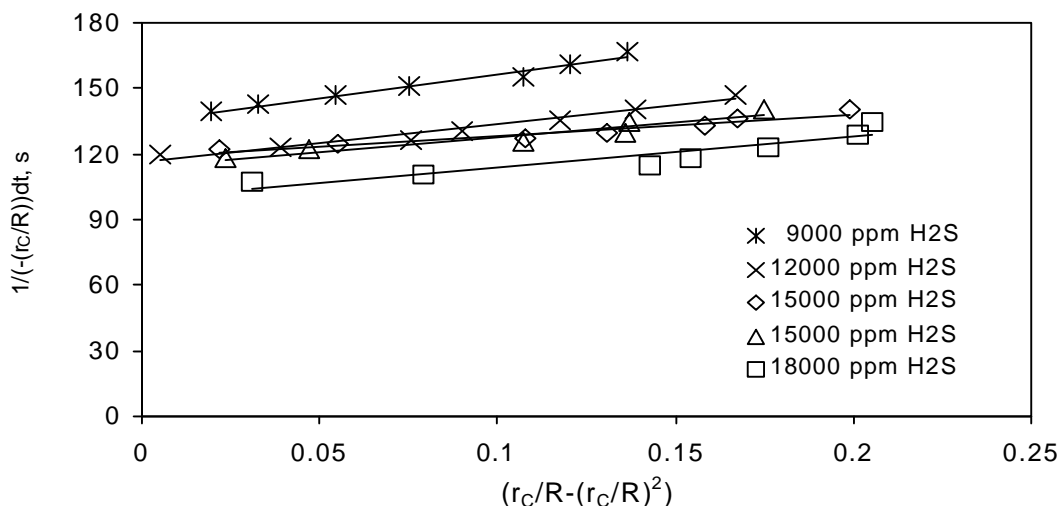
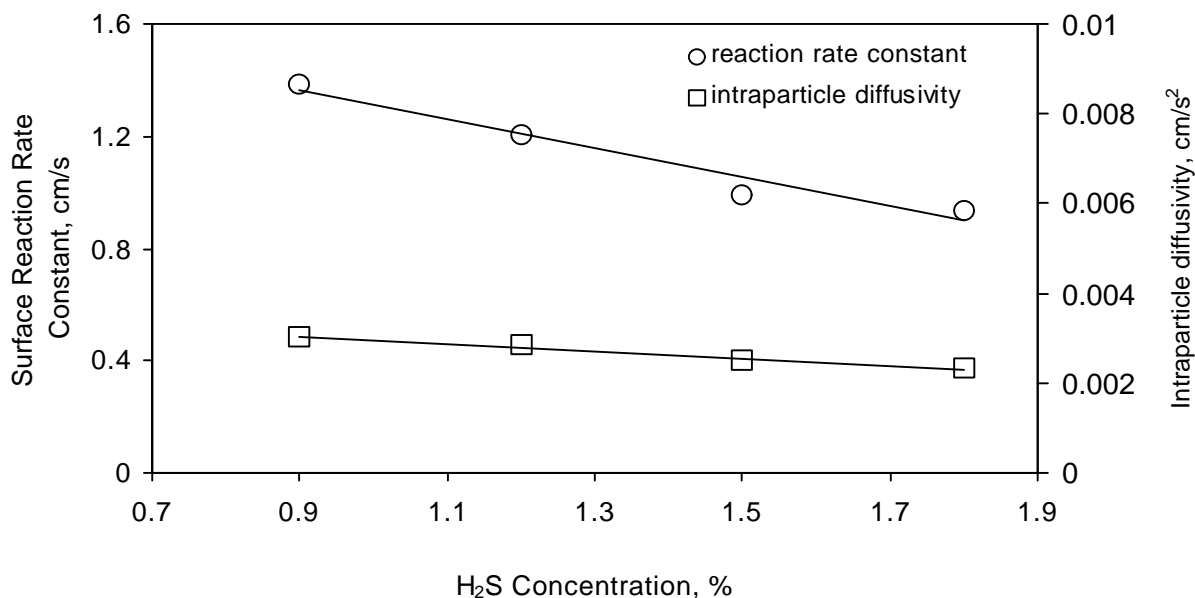


Figure 12. Effects of H<sub>2</sub>S concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H<sub>2</sub>S into 0.01-g AHI-5 sorbent with 10-v% H<sub>2</sub> and 10-v% moisture at 450°C.





Concentrations of H<sub>2</sub>S affect significantly initial dynamic absorption of H<sub>2</sub>S into the AHI-5 sorbent in the H<sub>2</sub>S concentration range of 9000 – 18000 ppm (see Figure 10). Equation 2 is applied to the experimental data. The range of surface reaction rate constant is 0.94 – 1.38 cm/s, and the range of intraparticle diffusivity values is 0.0023 – 0.0031 cm<sup>2</sup>/s. The reaction rate constants of the sorbent, obtained from Equations 2, decrease with increased H<sub>2</sub>S concentration (see Figure 12). The intraparticle diffusivity values of H<sub>2</sub>S through porous sorbents also decrease with increased H<sub>2</sub>S concentrations in the H<sub>2</sub>S concentration range of 9000 – 18000 ppm.

Figure 13. Effects of moisture concentration on absorption of 18000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen at 450°C and the space time 0.076 s.

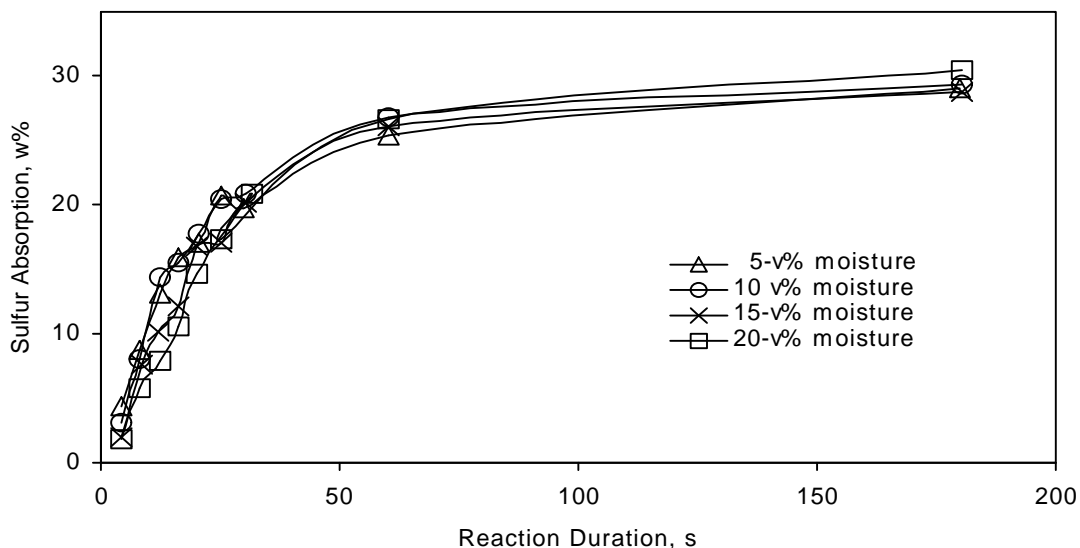
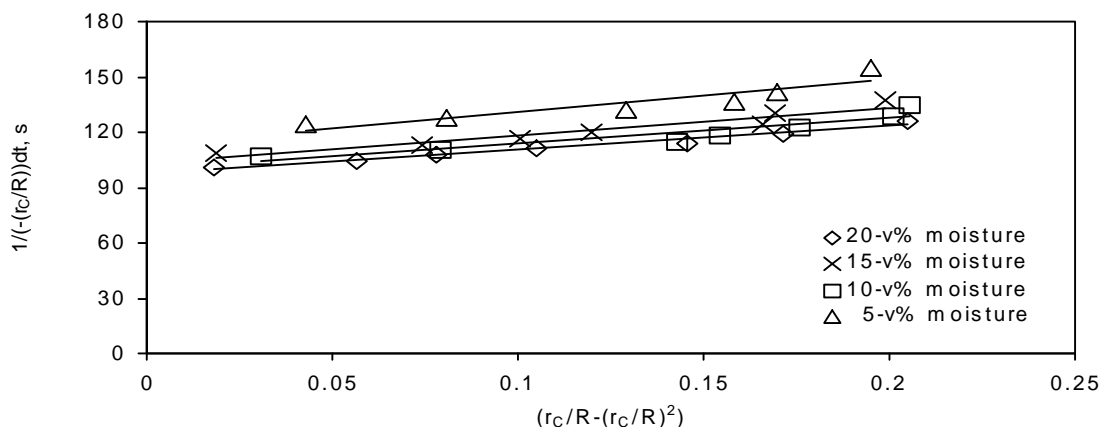


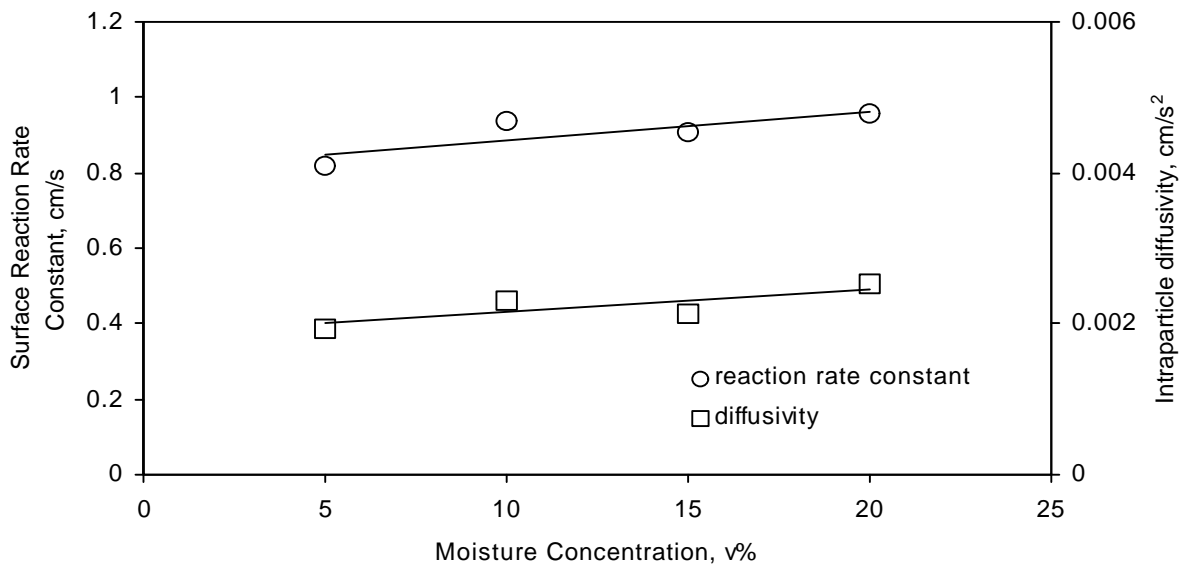
Figure 14. Effects of moisture concentration on absorption of 18000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% hydrogen at 450°C and the space time 0.076 s, using the reaction model.



## Effects of Moisture on Initial Dynamic Absorption of H<sub>2</sub>S

Experiments on initial absorption of H<sub>2</sub>S into the sorbents were carried out for 4 – 180 seconds at 40.7 psia and 450°C to find effects of moisture concentrations on initial dynamic absorption of H<sub>2</sub>S with the sorbent. The concentrations of H<sub>2</sub>S and hydrogen are maintained at 18000 ppm and 10-vol %, respectively. The concentration range of moisture in the simulated coal gas mixture is 5 – 20 volume percent.

Figure 15. Effects of moisture concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H<sub>2</sub>S into 0.01-g AHI-5 sorbent with 10-vol% H<sub>2</sub> at 450°C.



Initial absorption of H<sub>2</sub>S into the AHI-5 sorbent is affected with concentrations of moisture for short absorption duration of 4 – 180 seconds (see Figure 13). These facts may indicate that moisture is not inert for the reaction of H<sub>2</sub>S with the AHI-5 sorbent. Equation 2 was applied to the experimental data to obtain surface reaction rate constants and intraparticle diffusivity values of H<sub>2</sub>S with the sorbent (see Figure 14). The range of surface reaction rate constants is 0.82 – 0.96 cm/s, and the range of intraparticle diffusivity values is 0.0019 – 0.0025 cm<sup>2</sup>/s (see Table 4). These observations may indicate that reactivity of the AHI-5 sorbent and intraparticle diffusivity increase slightly with increased moisture concentrations. These facts also suggest that the controlling steps for the absorption of H<sub>2</sub>S into sorbent particles are surface reaction of H<sub>2</sub>S with reactive sites of sorbent particles and diffusion of H<sub>2</sub>S through porous sorbent particles in the presence of moisture.

## Effects of Hydrogen Concentration on Initial Dynamic Absorption of H<sub>2</sub>S

Experiments on effects of hydrogen concentrations on initial dynamic absorption of H<sub>2</sub>S into the sorbents were conducted for the reaction duration of 4 – 180 s at 450°C and the space-

time of 0.076 s. The concentration of moisture was maintained at 10-volume percent. The concentration of H<sub>2</sub>S was maintained at 18000 ppm.

Figure 16. Effects of hydrogen concentration on absorption of 18000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C and the space time 0.076 s.

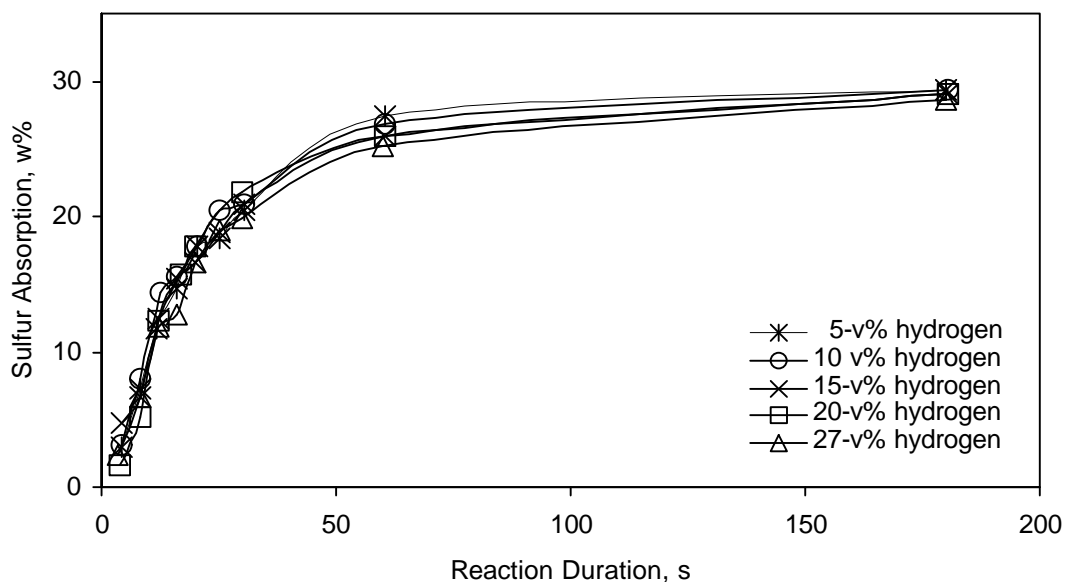


Figure 17. Effects of hydrogen concentration on absorption of 18000-ppm H<sub>2</sub>S on 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C and the space time 0.076 s, using the reaction model.

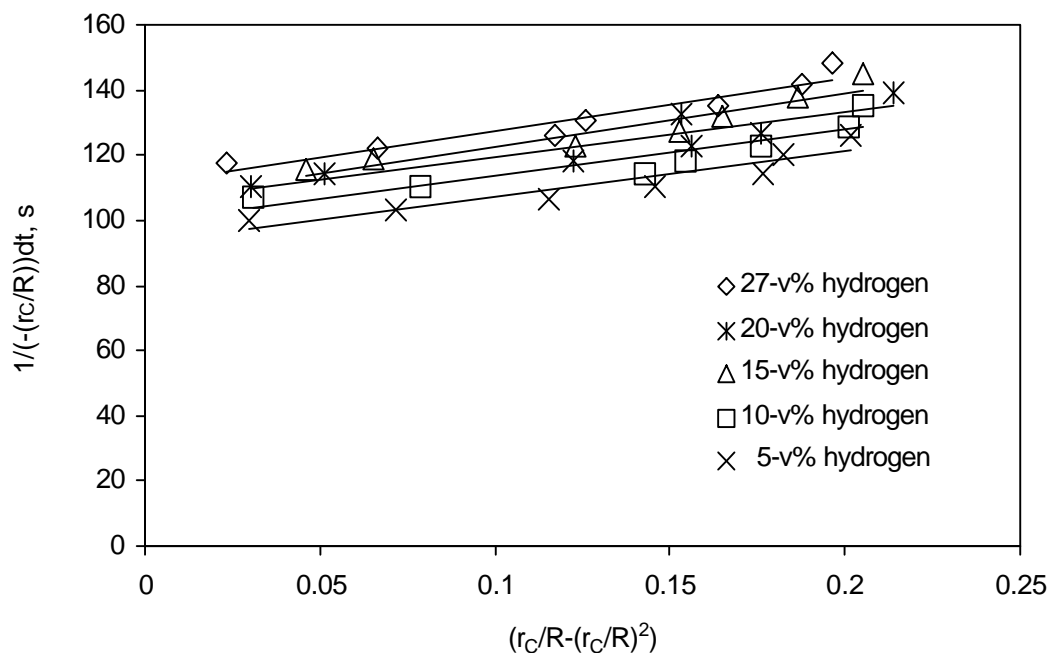
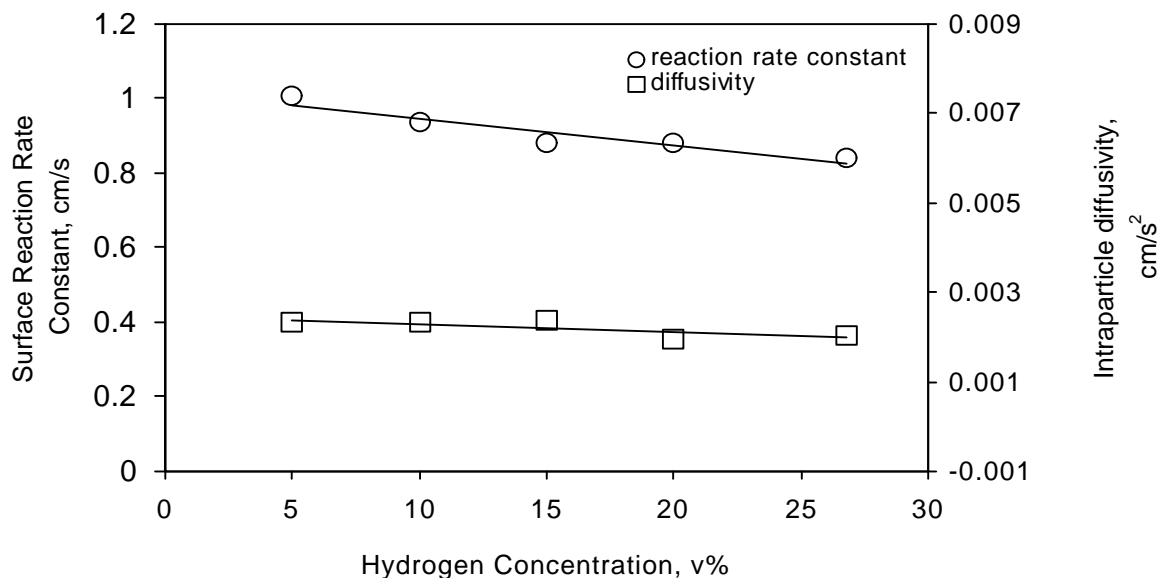


Figure 18. Effects of hydrogen concentration on surface reaction rate constant and intraparticle diffusivity in the absorption of H<sub>2</sub>S into 0.01-g AHI-5 sorbent with 10-v% moisture at 450°C.



Concentrations of hydrogen affect slightly initial dynamic absorption of H<sub>2</sub>S into the AHI-5 sorbent in the hydrogen concentration range of 5 – 27 v% (see Figure 16). Equation 2 was applied to the experimental data to obtain initial surface reaction rate constants and intraparticle diffusivity values (see Figure 17). The range of the initial surface reaction rate constant is 0.84 – 1.01 cm/s, and the range of the intraparticle diffusivity values is 0.0044 – 0.0052 cm<sup>2</sup>/s (see Table 4). Surface reaction rate constant and intraparticle diffusivity slightly decrease with hydrogen concentration. These facts may indicate that hydrogen affects both surface reaction of H<sub>2</sub>S with the sorbent and intraparticle diffusivity of H<sub>2</sub>S through the porous sorbent (see Figure 18).

## CONCLUSIONS

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

- Initial absorption of H<sub>2</sub>S into the AHI-5 sorbent is affected with absorption temperature for short absorption duration of 4 - 180 seconds. The range of surface reaction rate constant is 0.16 – 1.2 cm/s. The range of intraparticle diffusivity values of the sorbent is 0.0002 – 0.0029 cm<sup>2</sup>/s. These observations may indicate that reactivity of the AHI-5 sorbent be dependent on reaction temperature in the temperature range of 350 – 500°C. However, reactivity of the sorbent is much lower at 350°C than that at 500°C. These facts also suggest that the controlling steps for the initial absorption of H<sub>2</sub>S into sorbent particles are surface reaction of H<sub>2</sub>S with reactive sites of sorbent particles and intraparticle diffusivity through pores of the sorbent.

- The surface reaction rate constant and the intraparticle diffusivity value increases with the reaction temperatures up to 450°C. However, the surface reaction rate constant increases steeply, and the intraparticle diffusivity value decreases in the temperature range of 475 – 500°C. These facts may indicate that collapsing the pore structure of the sorbent results in the decreased intraparticle diffusivity value in the temperature range of 475 – 500°C.
- Amounts of the AHI-5 sorbent affect slightly initial dynamic absorption of H<sub>2</sub>S into the sorbent in the sorbent amount range of 0.005– 0.02 g. The range of the surface reaction rate constant is 0.86 – 1.11 cm/s, and the range of the intraparticle diffusivity values is 0.0018 – 0.0025 cm<sup>2</sup>/s. Absorption of H<sub>2</sub>S into the sorbent increases with decreased amount of the sorbent in the reactor. The surface reaction rate constant decrease with increased amount of the sorbent. The intraparticle diffusivity values increase generally with increased amount of the sorbent. These observations may suggest that mass transfer of H<sub>2</sub>S from the bulk gaseous phase to the surface of the solid sorbent affects somewhat the overall reaction of H<sub>2</sub>S with the sorbent
- Concentrations of H<sub>2</sub>S affect significantly initial dynamic absorption of H<sub>2</sub>S into the AHI-5 sorbent in the H<sub>2</sub>S concentration range of 9000 – 18000 ppm. The range of surface reaction rate constant is 0.94 – 1.38 cm/s, and the range of intraparticle diffusivity values is 0.0023 – 0.0031 cm<sup>2</sup>/s. The reaction rate constants of the sorbent decrease with increased H<sub>2</sub>S concentration. The intraparticle diffusivity values of H<sub>2</sub>S through the porous sorbent also decrease with increased H<sub>2</sub>S concentrations in the H<sub>2</sub>S concentration range of 9000 – 18000 ppm.
- Initial absorption of H<sub>2</sub>S into the AHI-5 sorbent is affected with concentrations of moisture for short absorption duration of 4 –180 seconds. These facts may indicate that moisture is not inert for the reaction of H<sub>2</sub>S with the AHI-5 sorbent. The range of surface reaction rate constants is 0.82 – 0.96 cm/s, and the range of intraparticle diffusivity values is 0.0019 – 0.0025 cm<sup>2</sup>/s. These observations may indicate that reactivity of the AHI-5 sorbent and intraparticle diffusivity increase slightly with increased moisture concentrations.
- Concentrations of hydrogen affect slightly initial dynamic absorption of H<sub>2</sub>S into the AHI-5 sorbent in the hydrogen concentration range of 5 – 27 v%. The range of the initial surface reaction rate constant is 0.84 – 1.01 cm/s, and the range of the intraparticle diffusivity values is 0.0044 – 0.0052 cm<sup>2</sup>/s. Surface reaction rate constant and intraparticle diffusivity slightly decrease with hydrogen concentration. These facts may indicate that hydrogen affects both surface reaction of H<sub>2</sub>S with the sorbent and intraparticle diffusivity of H<sub>2</sub>S through porous sorbents.

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#### PUBLICATIONS AND PRESENTATIONS

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