

Scale-Up of Advanced Hot-Gas Desulfurization Sorbents

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ABSTRACT

The overall objective of this project is to develop regenerable sorbents for hot gas desulfurization in IGCC systems. The specific objective of the project is to develop durable advanced sorbents that demonstrate a strong resistance to attrition and chemical deactivation, and high activity at temperatures as low as 343°C(650°F). A number of formulations will be prepared and screened in a ½-inch fixed bed reactor at high pressure (1 to 20 atm) and high temperatures using simulated coal-derived fuel-gases. Screening criteria will include, chemical reactivity, stability, and regenerability over the temperature range of 343°C to 650°C. After initial screening, at least 3 promising formulations will be tested for 25-30 cycles of absorption and regeneration. One of the superior formulations with the best cyclic performance will be selected for investigating scale up parameters. The scaled-up formulation will be tested for long term durability and chemical reactivity.

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EXECUTIVE SUMMARY

Additional sorbents were prepared, characterized and then tested for its hydrogen sulfide removal efficiency. Based on the results of the extensive testing conducted on this project so far, the zinc oxide-based sorbent is the best sorbent for hot gas application in the temperature range of 343°C to 538°C.

INTRODUCTION

Advanced high-efficiency integrated gasification combined cycle (IGCC) power systems are being developed to produce power from coal under the U.S. Department of Energy's (DOE's) multibillion dollar Clean Coal Technology (CCT) Program. In these advanced systems, coal is gasified to produce a gas at high temperature and high pressure (HTHP) conditions. The hot gas is cleaned of contaminants, primarily particulates and sulfur gases such as hydrogen sulfide (H_2S) and burned in a combustion turbine. IGCC systems are capable of higher thermal efficiency and lower gaseous, liquid, and solid discharges than conventional pulverized-coal-fired power plants. Hot gas cleanup offers the potentially key advantages of higher plant thermal efficiencies and lower costs due to the elimination of fuel gas cooling and associated heat exchangers.

Sorbents based on zinc oxide are currently the leading candidates and are being developed for moving-, and fluidized-bed reactor applications. Zinc oxide based sorbents can effectively reduce the H_2S in coal gas to 10 ppm levels and can be regenerated for multicycle operation. However, all of the current first-generation leading sorbents undergo significant loss of reactivity with cycling, as much as 50% or greater loss in only 25-30 cycles. Stability of the hot-gas desulfurization step over 100s of cycles is essential for improved IGCC economics over conventional power plants. Thus a pressing need exists for developing durable second generation sorbents with much improved and stable reactivity during cyclic operation.

The overall objective of this project is to develop regenerable sorbents for hot gas desulfurization in IGCC systems. The specific objective of the project is to develop durable advanced sorbents that demonstrate a strong resistance to attrition and chemical deactivation, and high activity at temperatures as low as 343°C(650°F).

RESULTS AND DISCUSSIONS

The project consists of three major experimental tasks (Tasks 1-3) addressing the contract objectives described above.

Task 1: Optimization of Preparation

Task 2: Investigation of Scale-Up

Task 3: Preparation of 100 lb Batch

Task 1: Optimization of Preparation

A highly promising method was recently developed to prepare suitable sorbents. Various sorbents were prepared by our proprietary method. The main parameters we have varied was various concentrations of starting materials and ageing conditions. The starting compounds was chosen based on water solubility, commercial availability and low costs and that avoid introducing elements that may be deleterious in the final sorbent or that cause difficulties in subsequent processing. These prepared sorbents were tested in the fixed bed reactor.

The following analytical techniques was used to characterize the fresh, sulfided and regenerated sorbents

1. X-ray Diffraction (XRD) for crystalline phase.
2. Surface area measurement will be based on the standard BET method.
3. Hg-porosimetry for pore volume, bulk density, average pore diameter and pore size distribution determination.
4. Atomic Absorption (AA) Spectrometry for elemental composition analysis.

Effect of Concentration

The summary of the BET, pore volume results are presented in Table 1. As shown in Table 1, as the concentration of starting solutions is increased (at fixed ageing time) from 1.0 to 2.5 molar, the surface area of the sorbent decreases from 8.9 to 5.1 m²/g. Also the pore volume decreases from 0.87 to 0.34. The bulk density increases from 0.67 to 1.43 g/mL. The SEM morphology (figure not shown here) of the MCRH 41, 42, 43, 44 and 45 shows that fine crystals are at the surface.

Currently we are analyzing the data as well as started writing the final report. All the properties of the sorbents will be reported in the final report.

Table 1. BET and Pore Volume of MCRH Sorbents

Sorbent	BET Surface Area, m ² /g	Concen tration	Ageing, min	Pore volume, mL/g	Avg Pore Dia (Å)	Bulk Density , g/mL	Porosity, %
MCRH-41	8.2	0.05 M	0	0.75	1904	0.7882	59.2
MCRH-42	8.9	1.0 M	0	0.87	2833	0.6753	58.9
MCRH-43	7.7	1.5 M	0	0.68	1398	0.7290	50.1
MCRH-44	6.0	2.0 M	0	0.52	1602	1.0149	52.9
MCRH-45	5.1	2.5 M	0	0.34	1320	1.4338	49.7
MCRH-46	7.3	1.0 M	30	0.82	2401	0.6967	57.5
MCRH-47	7.3	1.0 M	60	0.83	2368	0.6991	58.2
MCRH-48	6.4	1.0 M	90	0.84	2837	0.7098	59.6
MCRH-49	6.7	1.0 M	120	0.77	2526	0.7919	61.2

CONCLUSION

Based on the results of the extensive testing conducted on this project so far, the zinc oxide-based sorbent is the best sorbent for hot gas application in the temperature range of 343°C to 538°C.

FUTURE WORK

The grant expires on December 31, 1997. A final report is being prepared.

PUBLICATION/PRESENTATION

1. K. Jothimurugesan, A.A. Adeyiga and S.K. Gangwal "Regenerable Sorbents for Desulfurization of Coal Gas", Fourth Annual HBCUs/Private Sector Energy Research and Development Technology Transfer symposium, Greensboro, NC, April 2-4, 1996.
2. K. Jothimurugesan, A.A. Adeyiga and S.K. Gangwal, "Removal of Hydrogen Sulfide from Hot Coal Gas Streams", Thirteenth Annual International Pittsburgh Coal Conference Proceedings, p.596-601, 1996.
3. K. Jothimurugesan "Performance of Zinc Based Sulfur Sorbents on Coal Derived Fuel Gas", Fifth Annual Historically Black Colleges and Universities and Other

Minority Institutions Program-Energy Research and Development Technology Transfer Symposium, March 4-6, 1997.

4. Jothimurugesan, K., Gangwal, S.K., Gupta, R., and Turk, B.S., “Advanced Hot-Gas Desulfurization Sorbents”, Coal-Based Power and Environmental Systems ‘97 Conference, July 22-24, 1997, Pittsburgh, PA.
5. Gangwal, S.K., Gupta, R.P., Portzer, J.W., Turk, B.S and Jothimurugean, K., “Simultaneous Removal of H₂S and NH₃ Using Metal Oxide Sorbents”, Coal-Based Power and Environmental Systems ‘97 Conference, July 22-24, 1997, Pittsburgh, PA.