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 343ºC(650ºF). 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 343ºC to 650ºC. 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 and then tested for its hydrogen sulfide removal efficiency. Based on the results of the extensive testing conducted on this project so for, the zinc oxide-based sorbent is the best sorbent for hot has application in the temperature range of 343ºC to 538ºC.

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 343ºC(650ºF).

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.

Task 2. Investigation of Scale-Up

Forming operations to making particles for both fluidized-bed and moving bed was investigated by a balance among several factors, including rheological properties of the mixture, and the necessity to achieve satisfactory strength, an open-pore structure, and high activity for hydrogen sulfide removal. Suitable rheological properties was obtained by incorporating certain organic (e.g. methocel) and inorganic binders (e.g. kaolin and bentonite) and micropore formers.

The ZnO-based sorbentsMCRH-41, 42, 43, 44, 45, 46, 47, 49, were evaluated in a sulfidation gas mixture containing (in vol%): H_2=10%, CO=15%, CO_2=5%, H_2S=1.0%,

H_2O=15% and bal N_2. Figures 1-9 show the H_2S breakthrough profiles as a function of time. Of the sorbents tested MCRH-42 (Fig2.) showed excellent sulfidation behaviour. The pre-breakthrough H_2S level was less than 100 ppm and the breakthrough conversion was 100%. Regeneration of this sorbent was conducted with 2 volume percent O_2 in N_2 at 550ºC.

CONCLUSION

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

FUTURE WORK

The future activity will include, development and testing of attrition resistance zinc oxide based sorbents for fludized bed applications.

TIME SCHEDULE FOR YEAR 2

TASK	**Semiannual**	
	1	**2**
Sorbent Preparation	━━━━━━━━	━━━━━━━━
Sorbent Characterization	━━━━━━━━	━━━━━━━━
Sorbent Evaluation	━━━━━━━━	━━━━━━━━
Investigation of Scale-Up	━━━━━━━━	━━━━━━━━
Final Report		━━
Semiannual Reports	✍	✍

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.

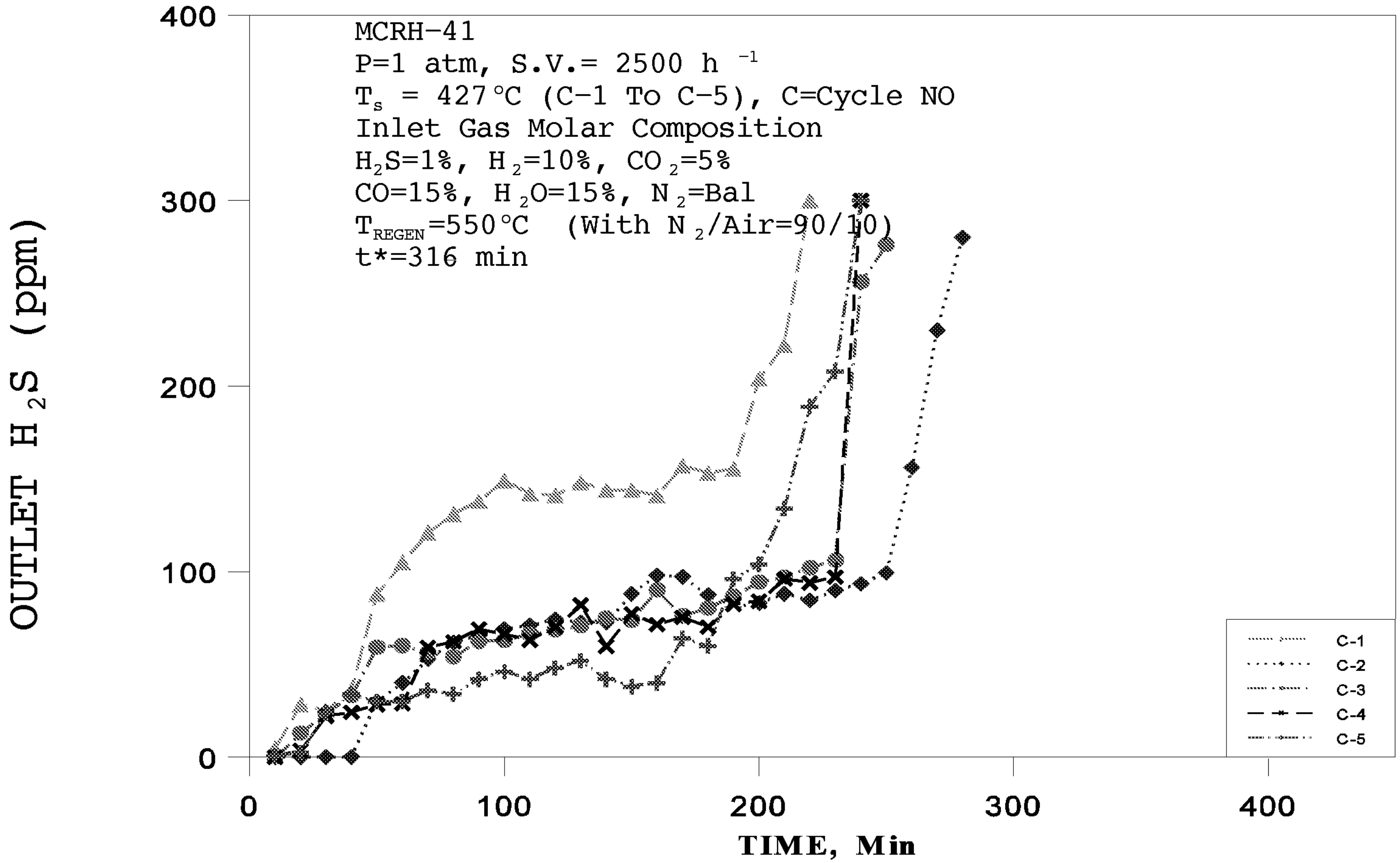

Figure 1. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-41

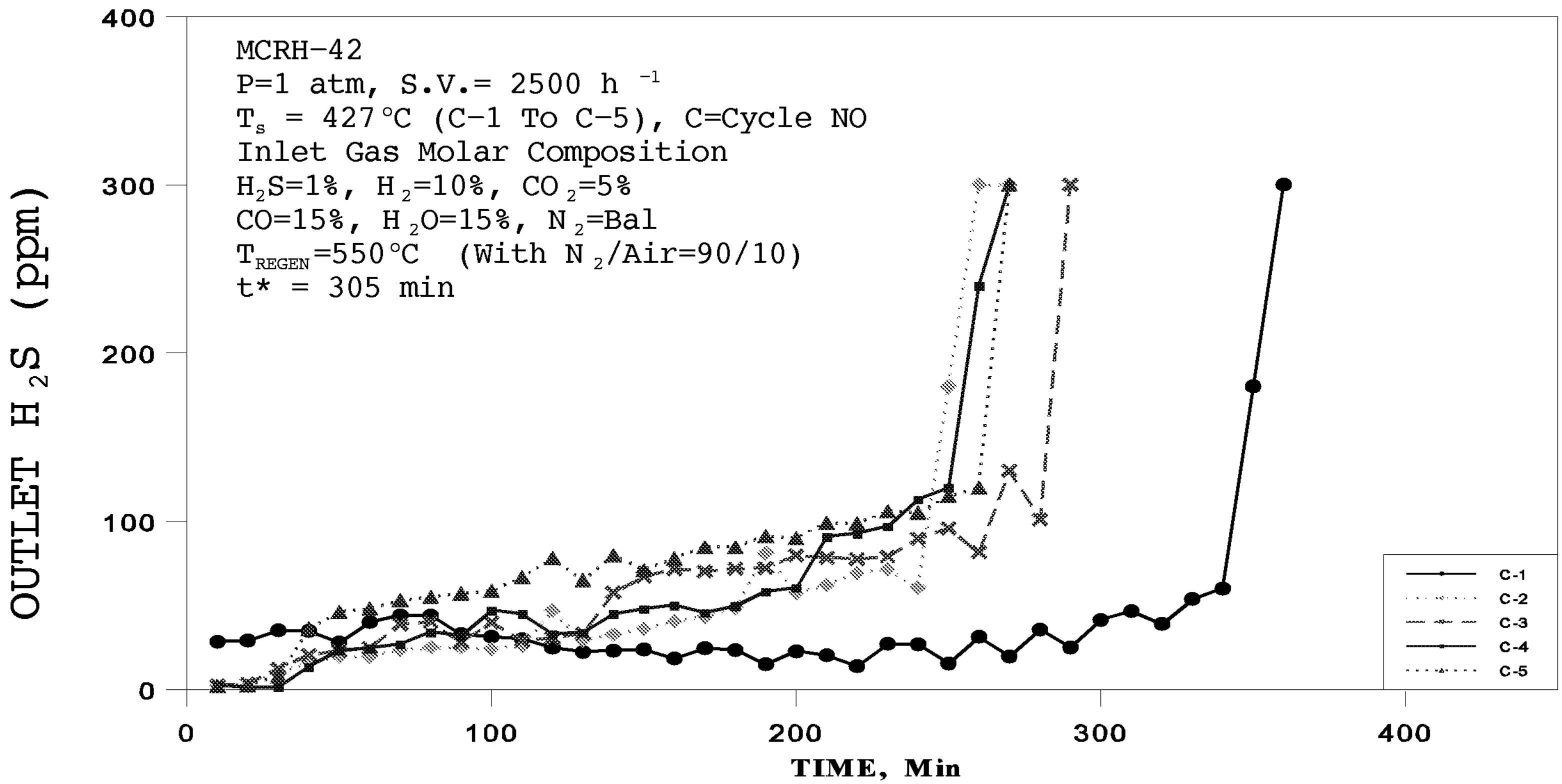

Figure 2. H_2S Breakthrough Curves in Successive Sulfidation Cycles of SorbentMCRH-42

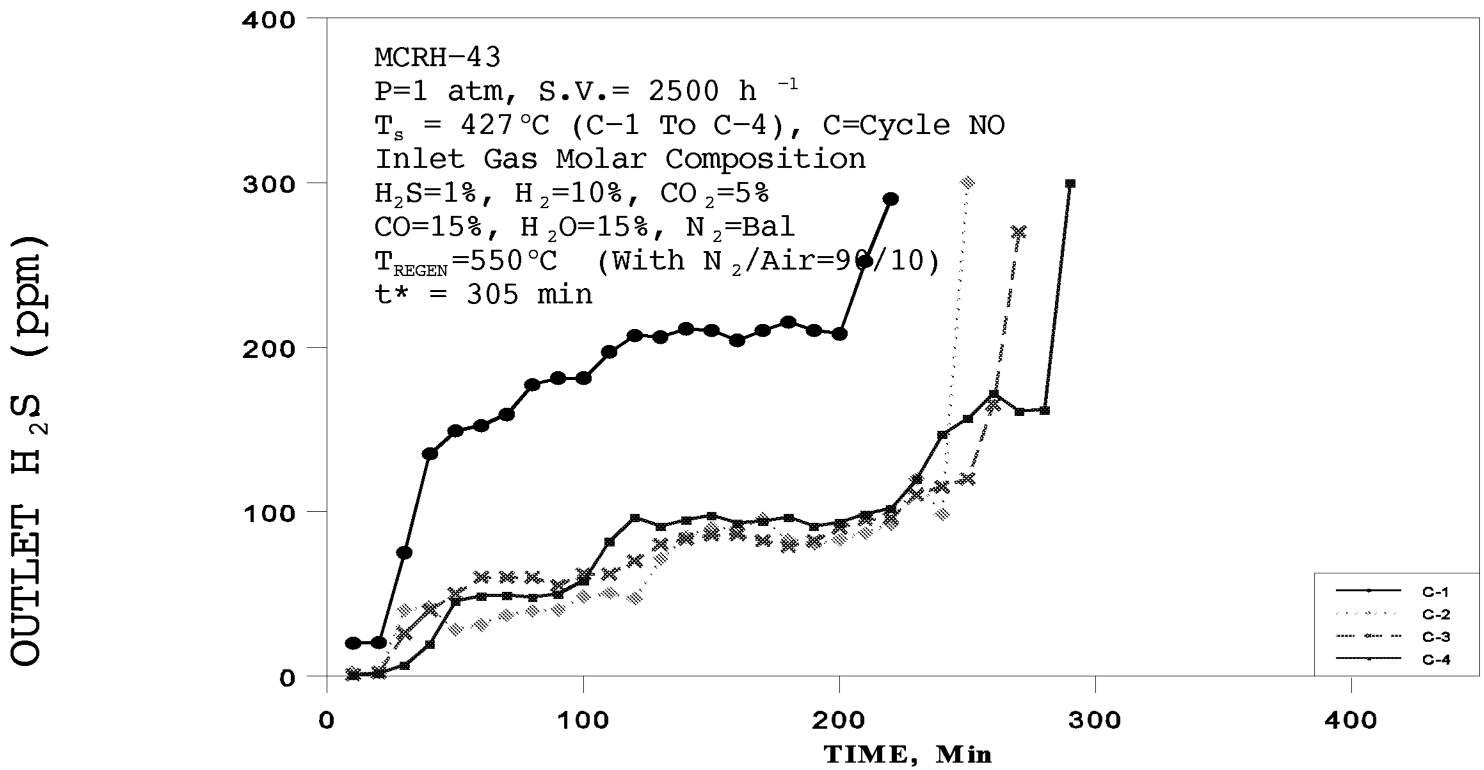

Figure 3. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-43

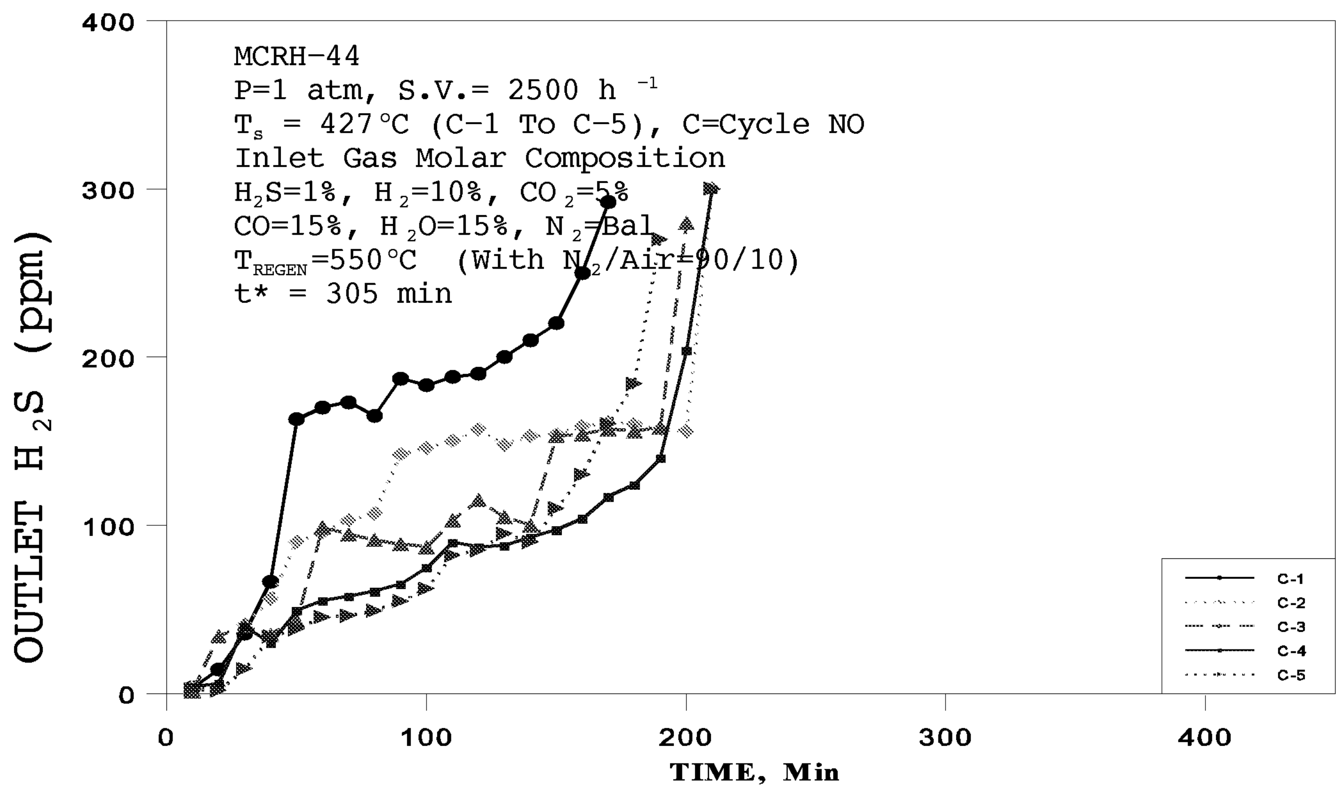

Figure 4. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-44

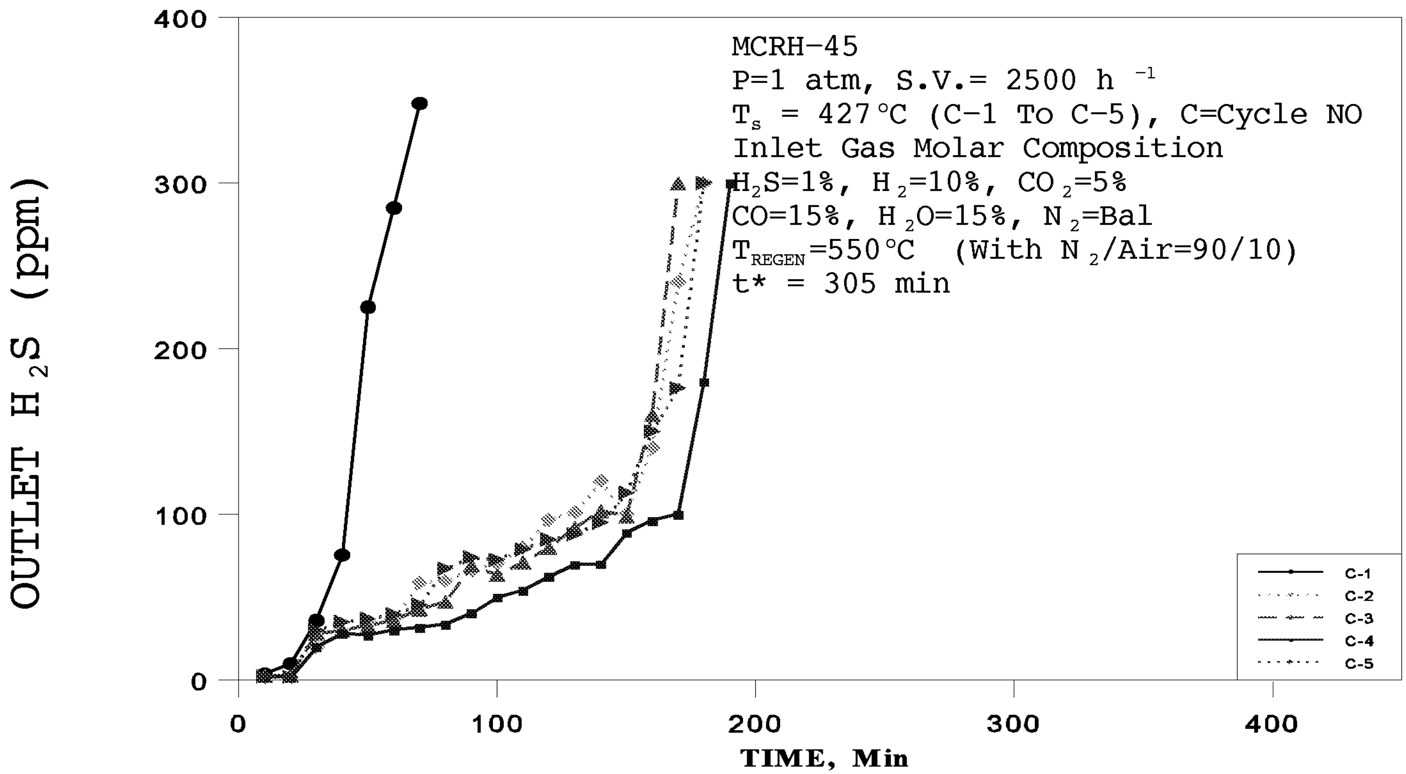

Figure 5. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-45

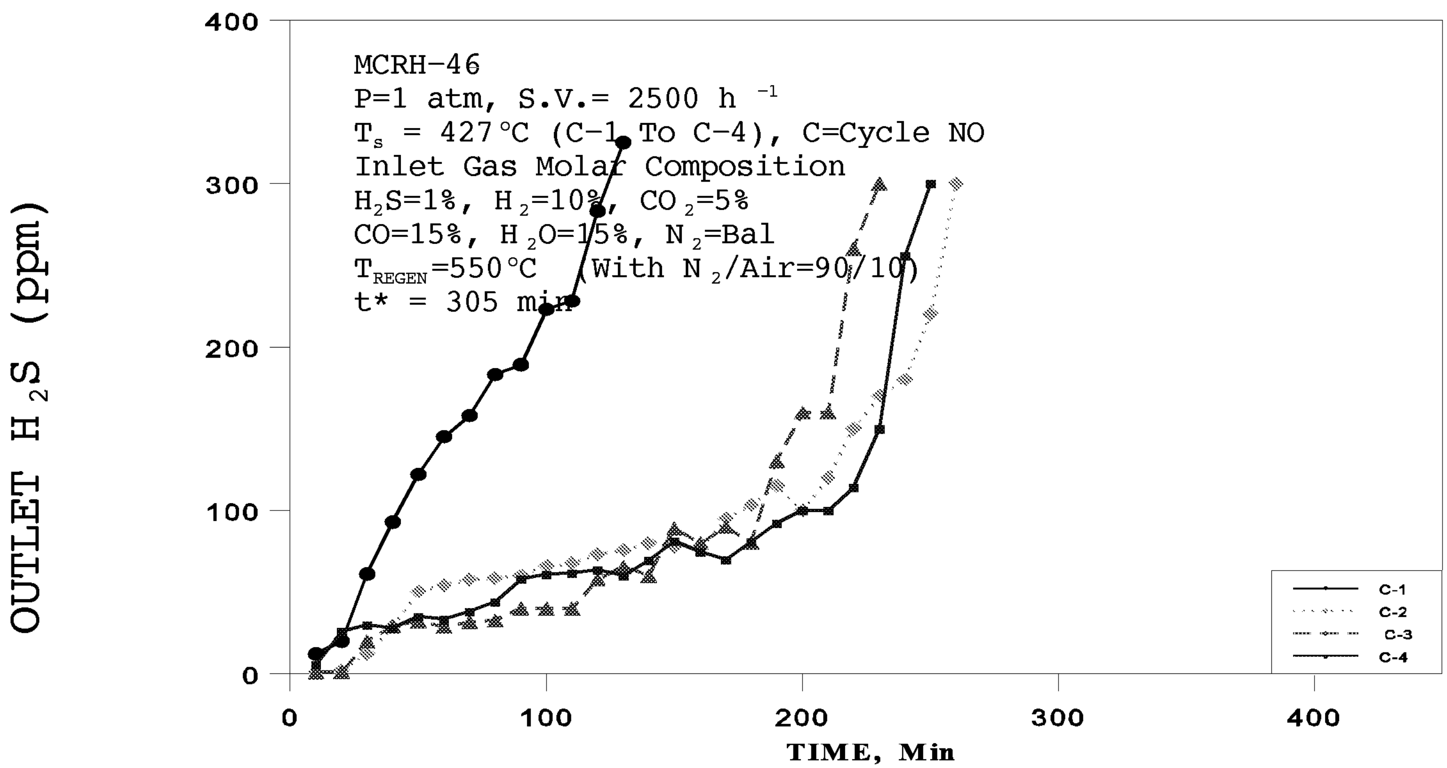

Figure 6. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-46

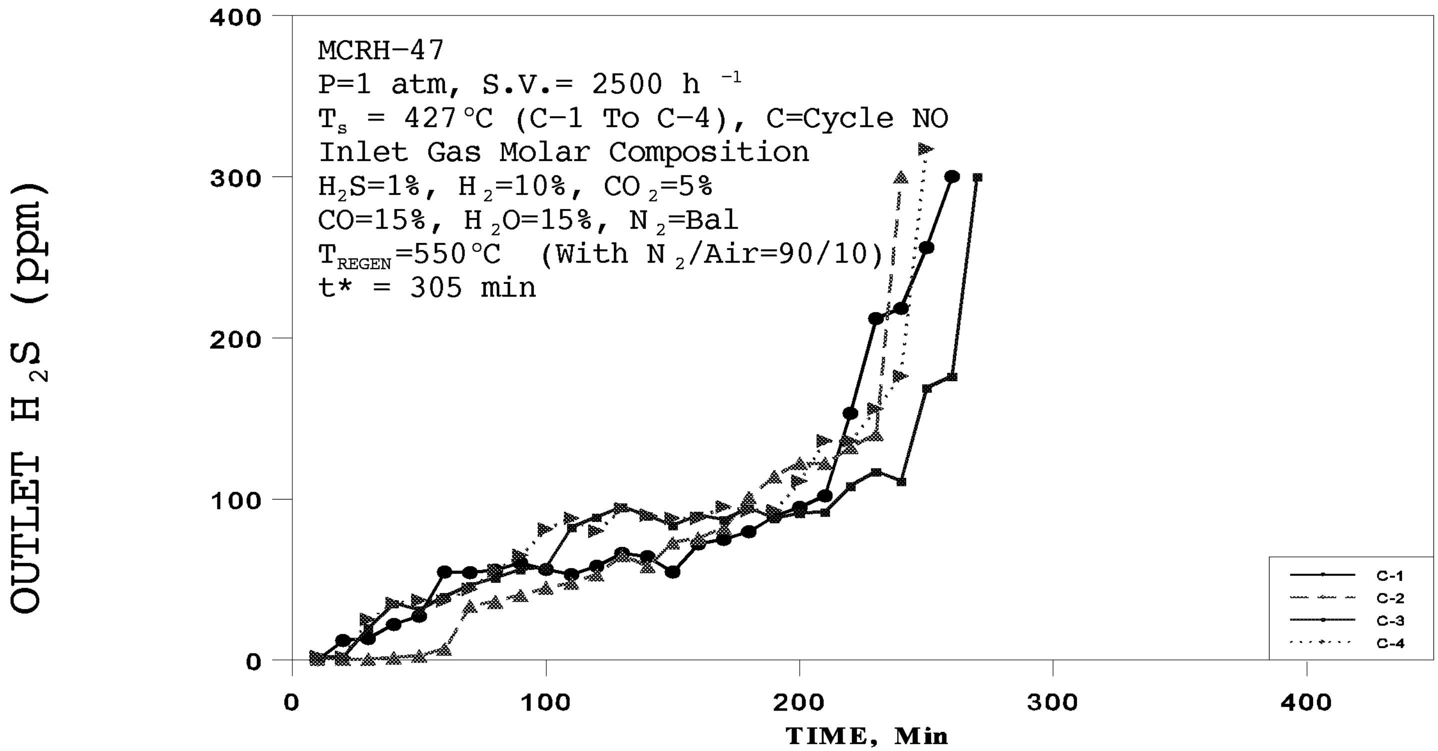

Figure 7. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-47

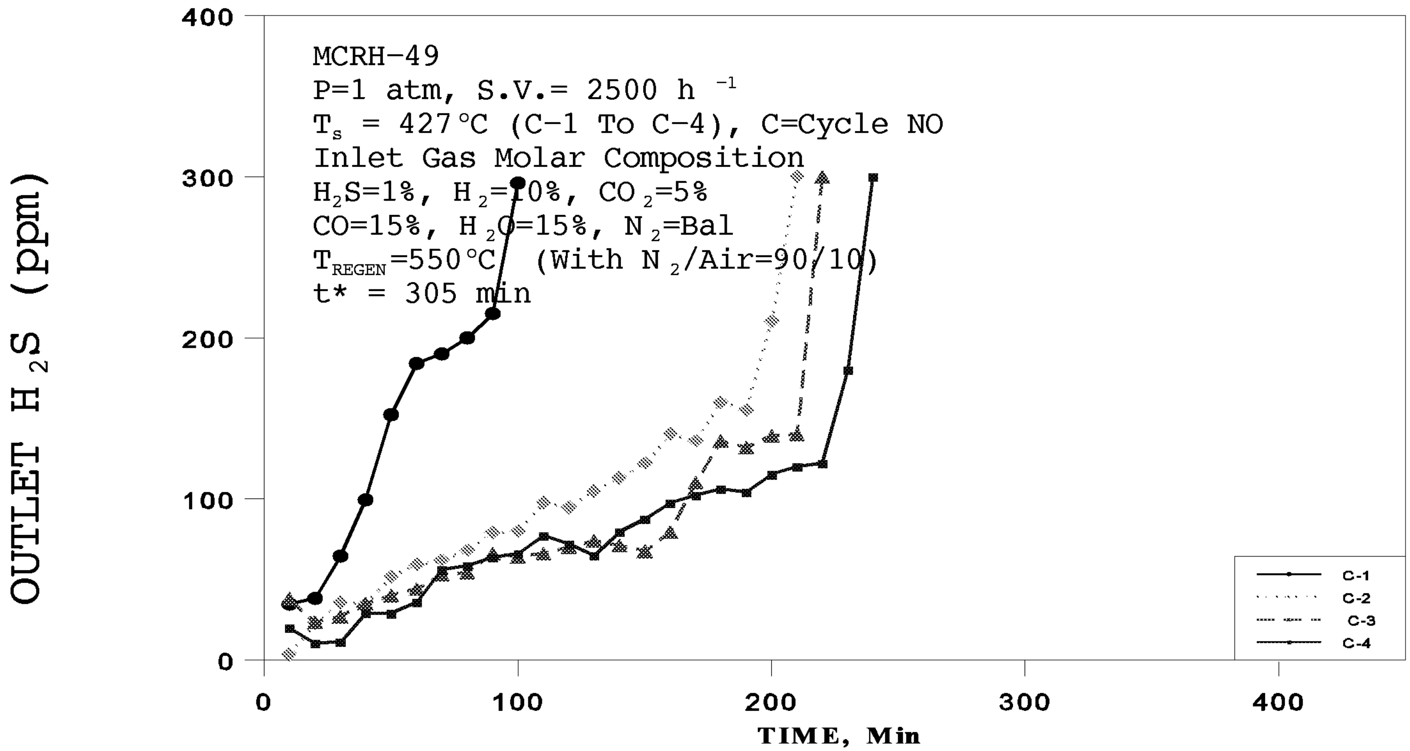

Figure 8. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-49