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Development of Advanced Hot-Gas Desulfurization Sorbents

Quarterly Report
April - June 1995

K. Jothimurugesan
Adeyinka A. Adeyiga
Santosh K. Gangwal

July 1995

Work Performed Under Contract No.: DE-FG21-94MC31393

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Hampton University
Hampton, Virginia

DISCLAIMER

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P.O. Box 880
Morgantown, West Virginia 26507-0880

By
Hampton University
Department of Engineering
Hampton, Virginia 23668

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CONTRACT INFORMATION

Contract Number:	DE-FG21-94MC31393
Contractor	Hampton University Department of Engineering Hampton, Virginia 23668
Principal Investigators	K. Jothimurugesan Adeyinka A. Adeyiga Santosh K. Gangwal (RTI)
DOE Project Officer	Kamalendu Das
Reporting Period	April 1, 1995-June 30, 1995

OBJECTIVES

The objectives of this project are to develop hot-gas cleanup sorbents for relatively lower temperature application, with emphasis on the temperature range from 343-538°C. The candidate sorbents include highly dispersed mixed metal oxides of zinc, iron, copper, cobalt and molybdenum. The specific objective in the successful preparation of H_2S absorbents will be to generate as high and as stable a surface area as possible, in order to develop suitable sorbent, that are sufficiently reactive and regenerable at the relatively lower temperatures of interest in this work. A number of formulations will be prepared and screened for testing in a 1/2-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 (650°F) to 538°C (1000°F). Each formulation will be tested for up to 5 cycles of absorption and regeneration. To prevent sulfation, catalyst additives will be investigated, which would promote a lower ignition of the regeneration. Selected superior formulation will be tested for long term (up to least 30 cycles) durability and chemical reactivity in the reactor.

BACKGROUND INFORMATION

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. Most of the hot-gas desulfurization research has focused on the development of mixed-metal oxide sorbents for application at 538°C (1000°F) to 760 °C (1400°F).

This project aims to develop hot-gas cleanup sorbents for relatively lower temperature applications, 343°C (650°F) to 538°C (1000°F) with emphasis on the temperature range from 400 to 500°C. There are a number of reasons for development of sorbents suitable for this temperature range. Recent economic evaluations have indicated that the thermal efficiency of IGCC systems increases rapidly with the temperature of hot-gas cleanup up to 350°C and then very slowly as the temperature is increased further. This suggests that the temperature severity of the hot-gas cleanup devices can be reduced without significant loss of thermal efficiency. Another important reason to reduce hot-gas cleanup severity is to reduce the operating temperatures and temperature swings of on-off valves and vessels for cyclic fixed-bed desulfurization/regeneration systems, lock-hopper valves for moving-bed applications, and the turbine load control valves. Finally, the current plans in Europe call for hot-gas desulfurization system operation at 350-600 °C. Also iron-oxide process development studies in Japan have concentrated on a temperature range of 400-600 °C. Thus development of suitable advanced sorbents in the U.S. for this temperature range is necessary for U.S. for maintain their leadership position in hot-gas cleanup.

PROJECT DESCRIPTION/RESULTS AND ACCOMPLISHMENTS

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

Task 1: Sorbent Preparation and Characterization

Task 2: Experimental Testing and Sorbent Evaluation

Task 3: Cyclic Testing

Task 1: Sorbent Preparation and Characterization

A highly promising method was recently developed under this contract to prepare suitable sorbents. Zinc oxide based sorbents was prepared using this proprietary technique. 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: Experimental Testing and Sorbent Evaluation

Various formulation of sorbents with catalyst additives was prepared and then it was investigated for its ability to promote a lower ignition of the regeneration. Figures 1-5 show the H_2S breakthrough profiles as a function of normalized time for various ZnO based sorbents (MCRH-14, 18, 23, 26 and 27). Of the sorbents tested MCRH-27 showed a superior performance. Complete (100%) sorbent conversion was observed at breakthrough at 800°F and the pre-breakthrough H_2S level was below 90 ppm. The regeneration light-off temperature for various sorbents tested is shown in Table 1

Table 1. Regeneration Light-off Temperature

SORBENT	LIGHT-OFF T°©
MCRH-10	650 °C
MCRH-14	600 °C
MCRH-18	600 °C
MCRH-23	600 °C
MCRH-26	600 °C
MCRH-27	550 °C

FUTURE WORK

Various formulations of sorbents with catalyst additives will be prepared and then it will be investigated for its ability to promote a lower ignition of the regeneration.

PUBLICATION/PRESENTATION

1. K. Jothimurugesan, A.A. Adeyiga and S.K. Gangwal "Development of Advanced Hot-Gas Desulfurization Sorbents" Third Annual Historically Black Colleges and Universities/Private Sector/Energy Research and Development Technology Transfer Symposium, Atlanta, Georgia, April 1995.

2. K. Jothimurugesan, A.A. Adeyiga and S.K. Gangwal "Development of Advanced Hot-Gas Desulfurization Sorbents" Advanced Coal-Fired Power Systems'95 Review Meeting, Morgantown, West Virginia, June 1995.

3. K. Jothimurugesan, A.A. Adeyiga and S.K. Gangwal "Development of Advanced Hot-Gas Desulfurization Sorbents" 1995 Annual AIChE Meeting, November-17, Miami Beach, FL. Abstract submitted

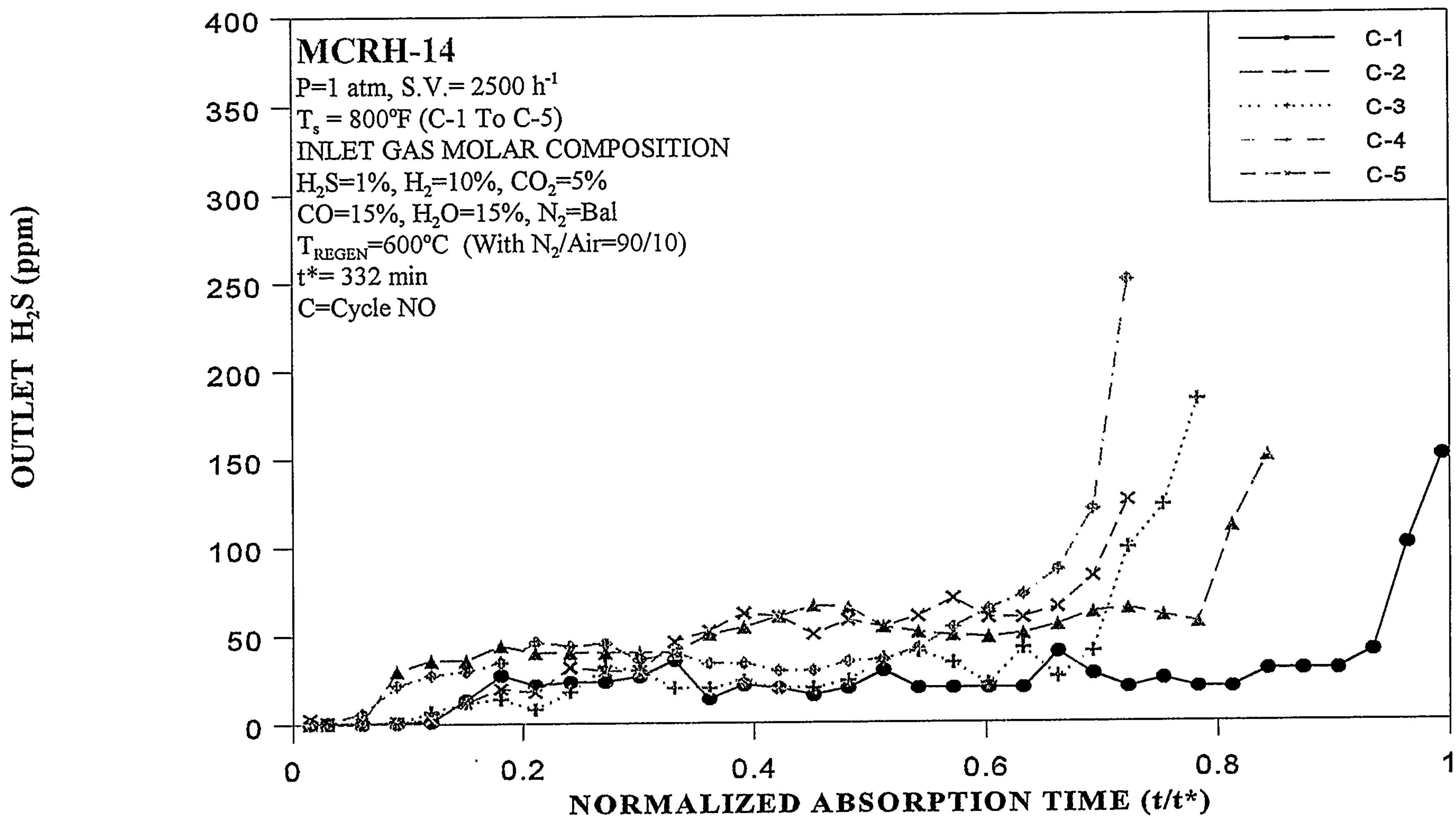

Figure 1. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-14

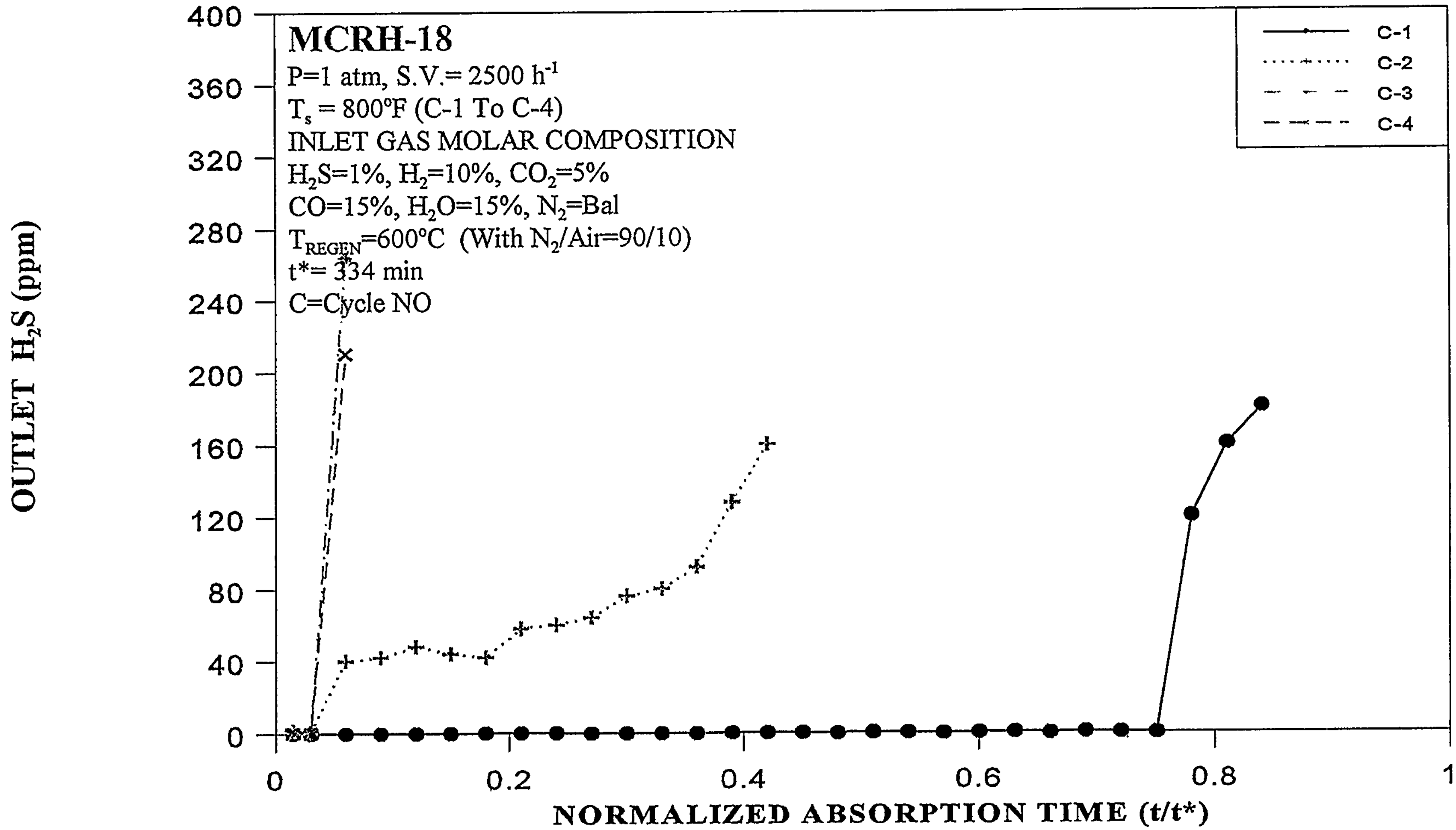

Figure 2. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-18

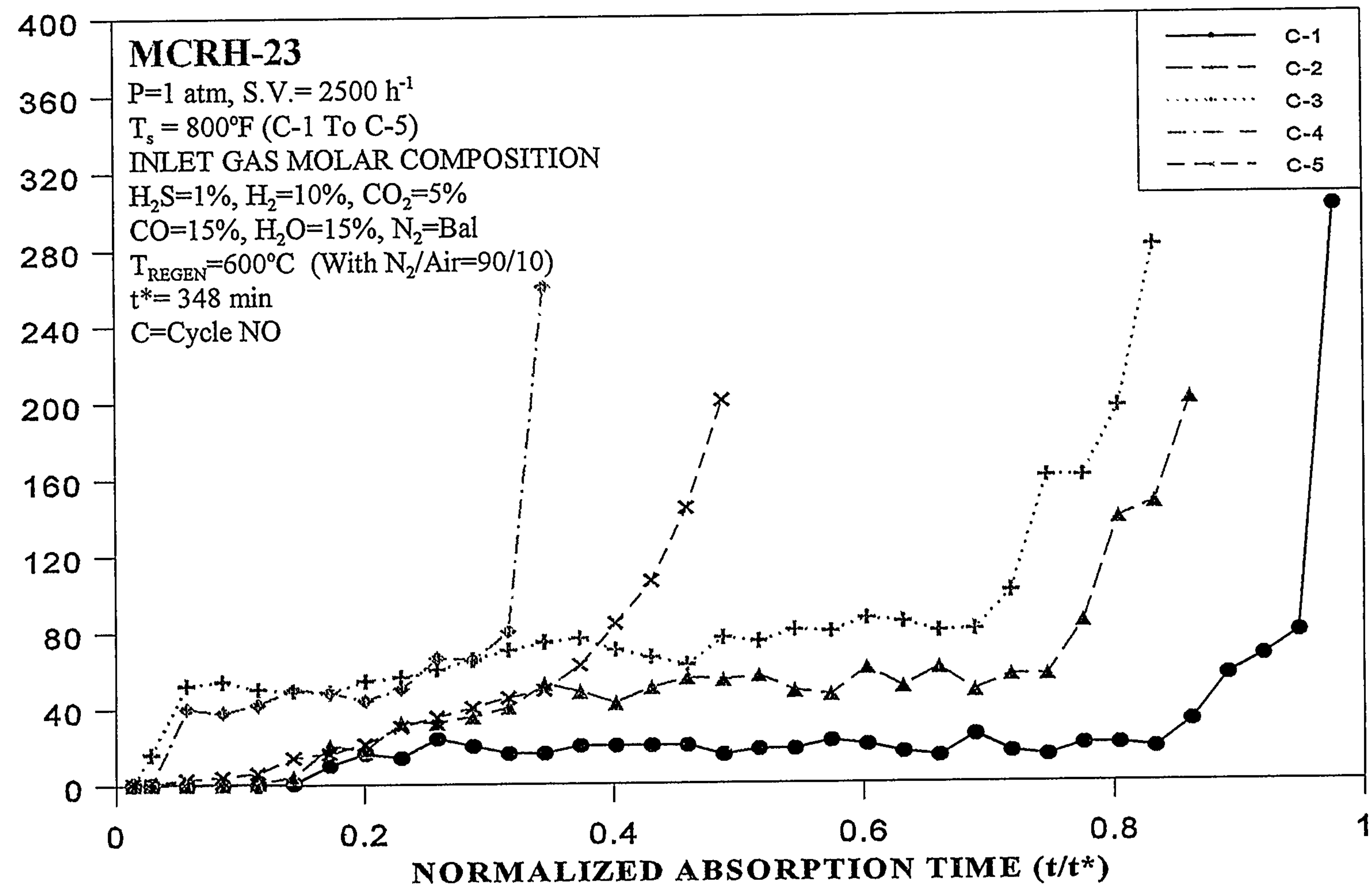

Figure 3.. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-23

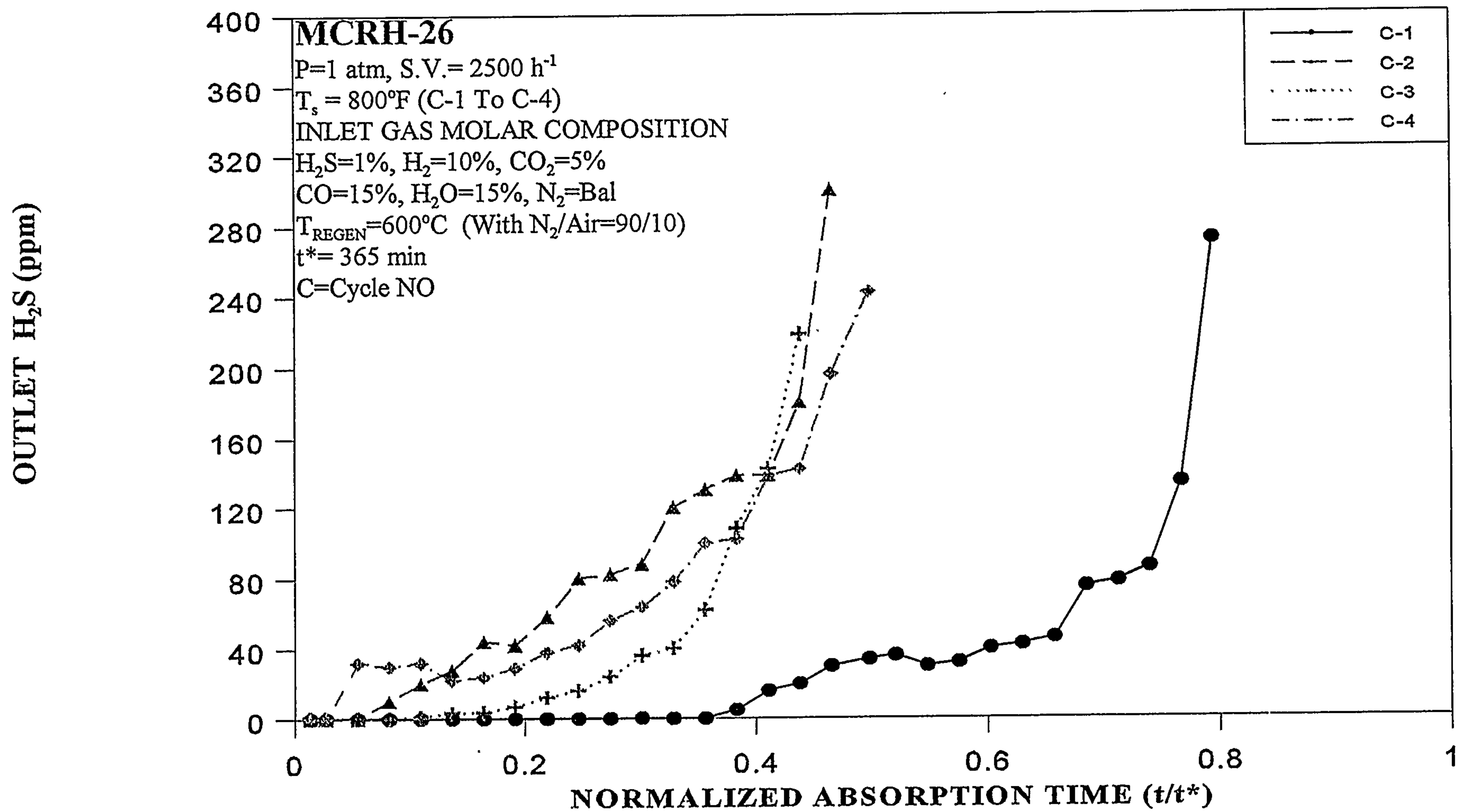

Figure 4. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-26

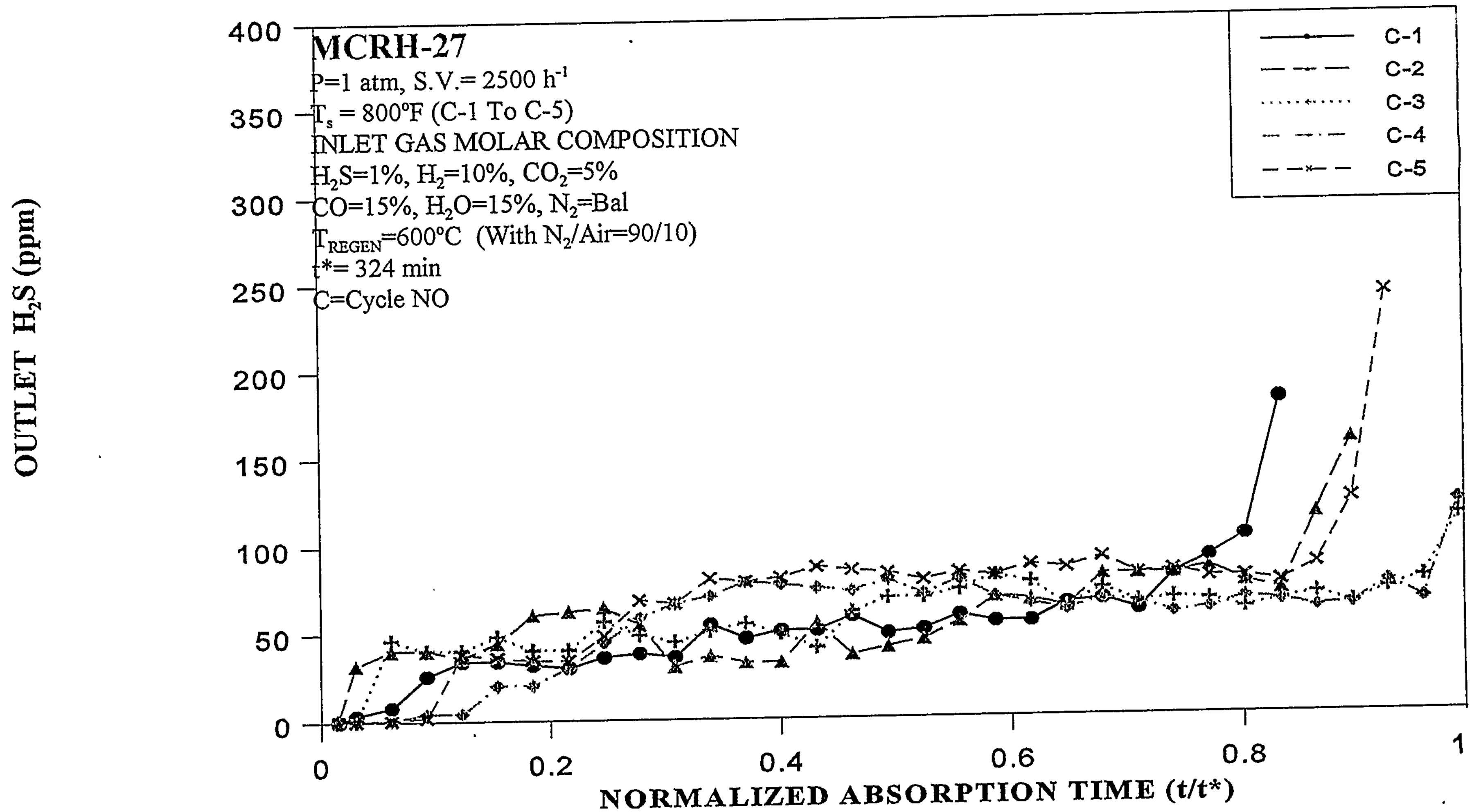

Figure 5. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent MCRH-27