DOE/MT/93005--T3

Simultaneous Removal of H_2S and NH_3 in Coal Gasification Processes

CONTRACT INFORMATION

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OBJECTIVES

The objective of this study is to develop advanced high-temperature coal gas desulfurization mixed-metal oxide sorbents with stable ammonia decomposition materials at 550-800° C (1022-1472° F). The specific objectives of the project are to:

(i) Develop a combined sorbent-catalyst materials shall be capable of removing hydrogen sulfide to less than 20 ppmv and ammonia by at least 90 percent.

(ii) Carry out comparative fixed-bed studies of absorption and regeneration with various formulations of sorbent-catalyst systems and select most promising sorbent-catalyst type.

(iii) Conduct long-term (at least 30 cycles) durability and chemical reactivity in the fixed-bed with the superior sorbent-catalyst.

BACKGROUND INFORMATION

Nitrogen (N_2) occurs in coal in the form of tightly bound organic ring compounds, typically at levels of 1 to 2 wt% on a dry-ash-free basis. During, coal gasification, this fuel-

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bound nitrogen is released principally as ammonia and nitrogen, with smaller levels of HCN. The formation of NH_3 in a coal gasification processes is a function of the fuel gas composition and the gasifier operating conditions. During the use of coal gas to generate electricity in gas-fired turbines, fuel bound(N_2) is converted to nitrogen oxides(NO_x), which are difficult to remove and are highly undesirable as atmospheric pollutants. Recent results indicate that while the efficiency of molten carbonate fuel cell (MCFC) anodes is not effected by exposure to NH_3, NO_X is generated during combustion of the anode exhaust gas. Thus, NH_3 must be removed from the coal gas before it is used in IGCC or MCFC applications.

The product stream from a high temperature, oxygen-blown gasifier, such as Texaco, contains about 2000 ppmv of NH_3, where higher concentrations (about 5000 ppmv) occur when the gasification is conducted at lower temperatures, such as in the Lurgi or GE air-blown gasifier. A range of 1500 to 3000 ppmv is considered for this study.

Removal of H_2S using zinc-based sorbents, particularly zinc titanate, to < 20 ppmv levels has been well established (Lew et al., 1989; Jothimurugesan and Harrison, 1990; Woods et al., 1990; Gupta and Gangwal, 1993,). Previous literature study indicated that catalyst have high activities for NH_3 decomposition (Krishnan et al., 1988). If desulfurization sorbents such as zinc titanate could be used along with the NH_3 decomposition catalysts to decompose ammonia present in hot coal gas, then the number of unit processes necessary to clean hot coal gas could be reduced by one.

The objective of this project is to develop successful combination of an NH_3 decomposition catalyst with the mixed-metal oxide sorbent so that the sorbent-catalyst activity remains stable for NH_3 decomposition in addition to H_2S removal under cyclic sulfidation-regeneration conditions in the temperature range of 550-800° C (1022-1472° F) and pressures up to 20 atm.

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 - Catalyst Preparation and Characterization

Task 2: Experimental Testing

Task 3: Cyclic Testing

Task 2: Experimental Testing

The activities of the HART 42 and HART41 sorbent-catalysts were tested using the simulated coal gas. The H_2S removal ability is shown in Figures 2&4 and its ammonia decomposition activity is shown in Figures 1&3. Figures 2& 4 shows the H_2S breakthrough profiles as a function of time. The pre-breakthrough H_2S level was below 100 ppm. Nearly complete sorbent conversion (100%) was observed at breakthrough.

The HART 42 sorbent-catalysts showed moderate catalytic activity (50% average conversion) for ammonia decomposition as shown in Figure 1. The average conversion decreased from 66 to 47 % as the temperature was increased from 600 to 750° C for HART41 sorbent-catalysts as shown in Figure 3.

FUTURE WORK

Additional sorbent-catalysts containing Mo and W will be tested in the fixed bed reactor.

REFERENCES

1. Gupta, R and Gangwal, S.K., "Fludizable Zinc Titanate Materials with High Chemical Reactivity and Attrition Resistance", U.S. Patent 5,254,516, 1993.

2. Jothimurugesan,K and Harrison, D.P., "The Reaction Between H_2S and Zinc Oxide-Titanium Oxide Sorbents: II. Single Pellet Sulfidation Modelling", Ind.Eng.Chem.Res. 29,1167(1990).

3. Krishnan, G.N., Wood, B.J., Tong, G.T., and McCarty, J.G., "Study of Ammonia

Removal in Coal Gasification Processes", Final Report to U.S. DOE/METC. Contract Number DE-AC21-86MC23087, 1988.

4. Lew,S., Jothimurugesan, K and Flytzani-Stephanopoulos,M., "High-Temperature H_2S Removal from Fuel Gases by Regenerable Zinc Oxide -Titanium Dioxide Sorbents", Ind.Eng.Chem.Res., 28,535(1989).

5. Woods,M.C., Gangwal., S.K., Jothimurugesan,K and Harrison, D.P., "The Reaction Between H_2S and Zinc Oxide-Titanium Oxide Sorbents: I. Single Pellet Kinetic Studies", Ind.Eng.Chem.Res, 29,1160(1990).

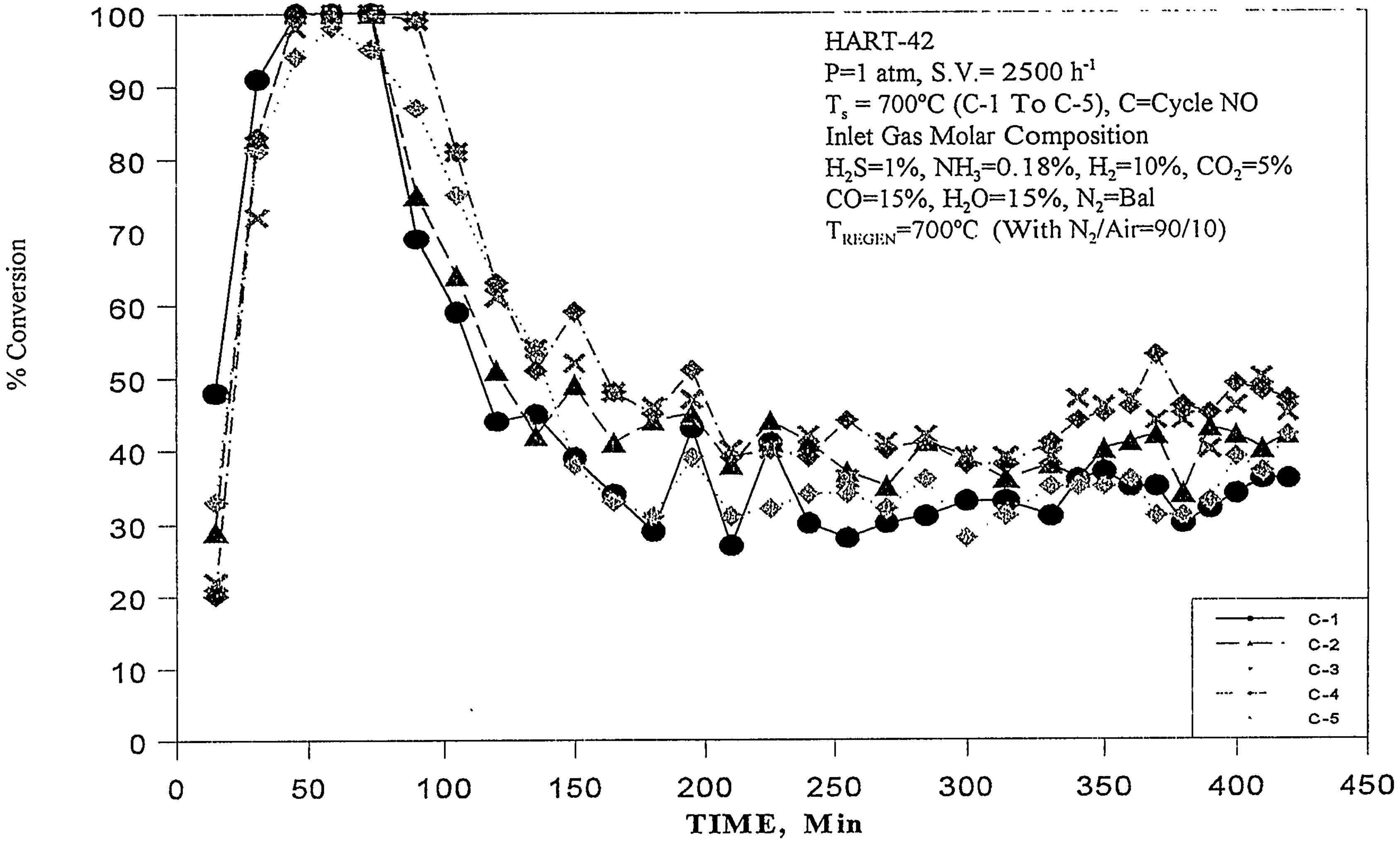

Figure 1.Conversion of ammonia on HART-42 catalyst-sorbents at 700°C

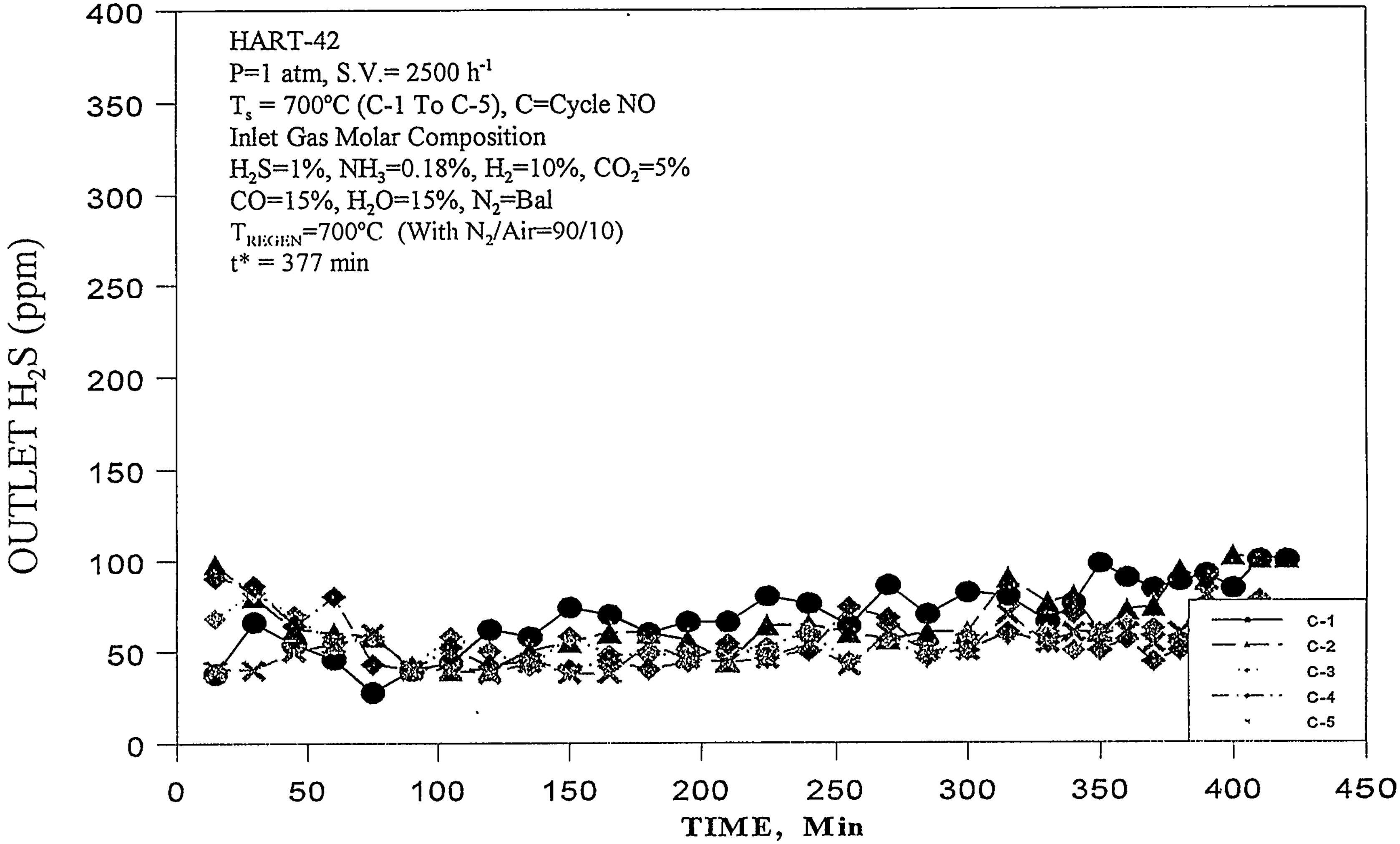

Figure 2. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent-Catalyst HART-42

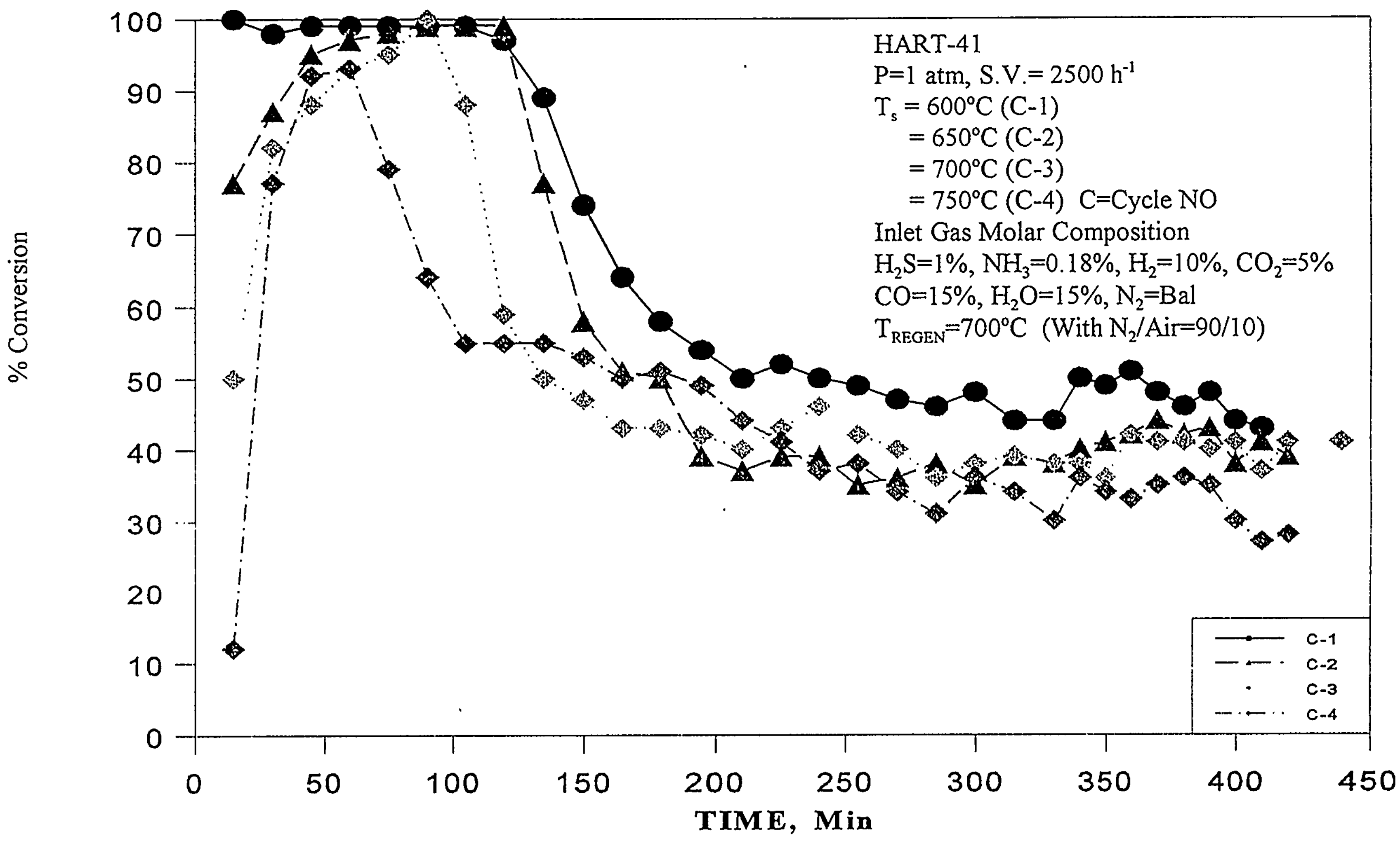

Figure 3.Conversion of ammonia on HART-41 catalyst-sorbents.

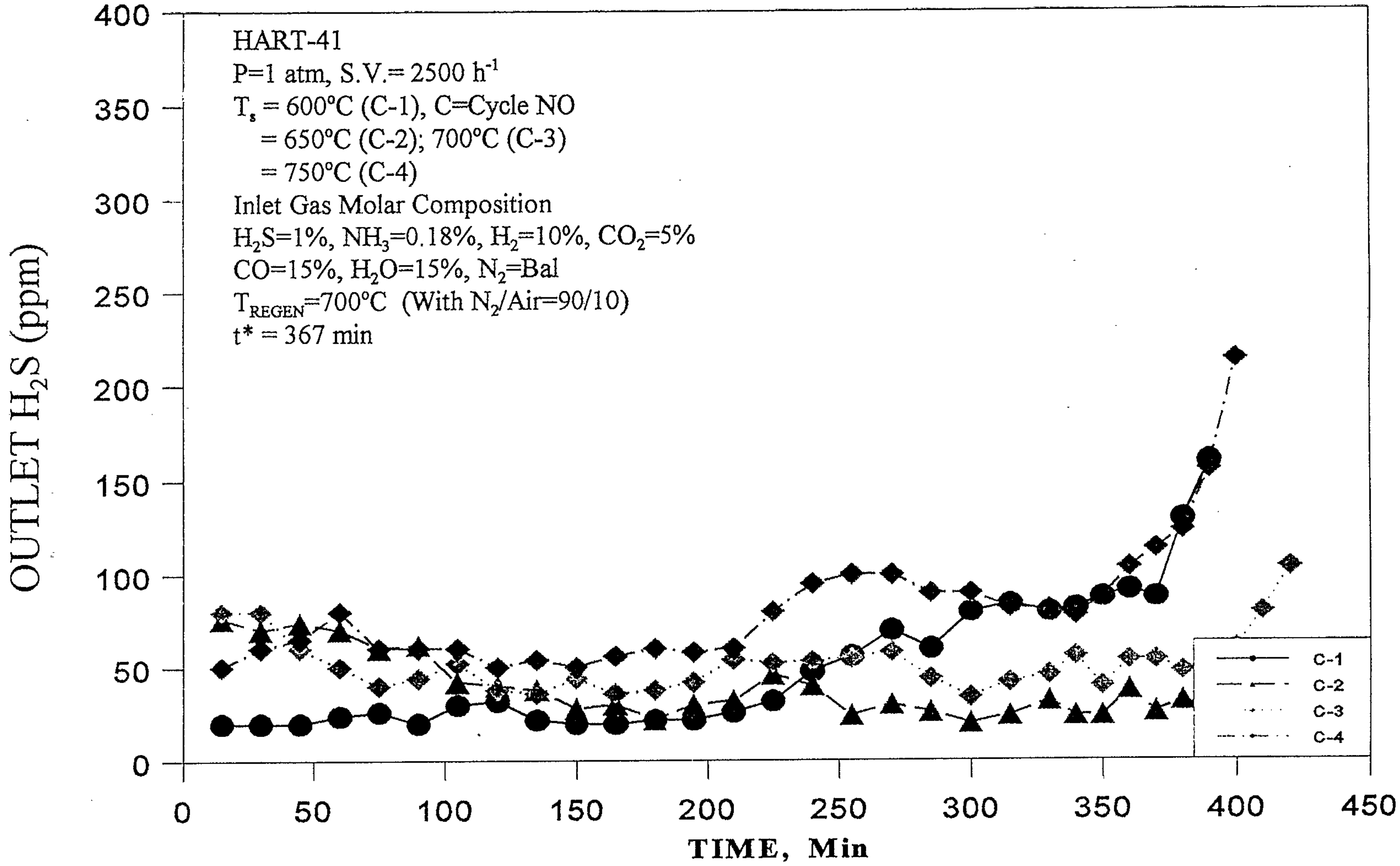

Figure 4. H_2S Breakthrough Curves in Successive Sulfidation Cycles of Sorbent-Catalyst HART-41