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Advanced Low-Temperature Fluid Bed Sorbents

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Advanced Low-Temperature Fluid Bed Sorbents

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Abstract

The Integrated Gasification Combined Cycle (IGCC) process is emerging as the most promising technology for advanced electric power generation from coal that is likely to replace conventional coal combustion. The commercialization of this technology and realization of its full potential, however, depend to a great extent on the development of regenerable sorbents capable of reducing the H_2S content of gasifier fuel gas from several thousand ppmv levels down to a few ppmv at elevated temperatures (i.e., >3500C), over hundreds or even thousands of sulfidation/regeneration cycles.

Over the last two decades, development of high temperature regenerable desulfurization sorbents has been pursued, primarily using various combinations of transition metal oxides. The primary focus has been directed toward application above 550°C, limiting the choice of metal oxides to those which possess favorable thermodynamic equilibria. These efforts led to the emergence of zinc-based sorbents, such as zinc titanates, as the leading candidates. However, because of problems with sorbent deterioration, further improvement is needed to minimize the overall cost of desulfurization in the IGCC process.

Recent studies on total IGCC system integration have indicated that system components become prohibitively expensive with increasing operating temperature and that the overall process efficiency gains of conducting desulfurization at above 550°C may not be sufficient to justify operation at such high temperatures. The optimum desulfurization temperature appears to be in the range of 350 °-550°C, where technical viability and process efficiency result in lower overall process costs. In addition, because of the more favorable thermodynamic equilibria at the lower temperature range, a large number of metal oxides can be considered for coal gas desulfurization, increasing the likelihood of developing suitable sorbents.

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The three most advanced reactor configurations for hot gas desulfurization include fixed-bed, moving-bed, and fluidized-bed systems. The fluidized-bed approach offers advantages over the moving-bed and fixed-bed reactors because of its ability to control temperature particularly during the highl y exothermic regeneration step; however, a more durable, attrition-resistant sorbent is required.

This paper discusses the results obtained in an ongoing study geared towards developing advanced mixed-metal oxide sorbents for desulfurization of coal-derived fuel gases in the temperature range of 350° to 550°C. The paper focuses on the study related to the development of durable sorbents suitable for fluidized-bed application and addresses thermodynamic considerations, sulfidation kinetics, regenerability, and the physical and chemical characteristics of a number of novel sorbents.

Acknowledgments

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4A.3 Advanced Low-Temperature Fluid Bed Sorbents

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Overall Objective

Develop **regenerable sorbents** for removal of hydrogen sulfide from **coal-**derived fuel gases in **fluidized** bed reactor in the temperature range of **343-538°C (650-1000 “F)**

Why Lower Temperature?

- Alkali condensation and removal
- More favorable H_2S Equilibria
- Greater number of suitable metal oxides
- Lower stress on sorbent
- Higher sorbent stability in long run
- Less expensive system components

Specific Objectives

- Selection of potential candidate sorbents
- Screening of potential candidate sorbents
- Laboratory evaluation of selected sorbents
- Bench scale testing of superior sorbent formulations
- Development of cost assessment/Market Plan
- Long-term testing of most promising sorbent

Thermodynamic Calculations

- H_2S equilibria calculations
  - Pure metal oxides
  - Sorbent/H_2S =2
  - 350-5500C, 20 bar
  - Texaco & U-Gas

- Phase stability diagrams

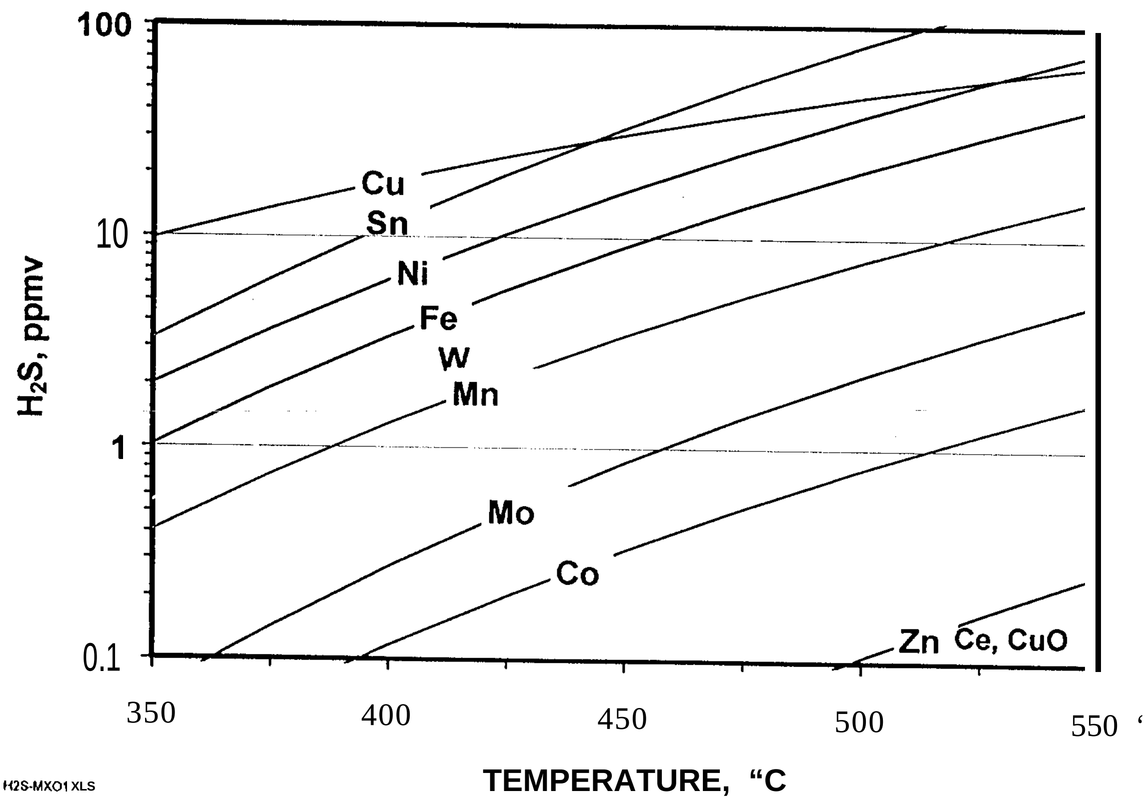
SULFIDATION FEED GAS COMPOSITION

Component	Mel%
H_2S	0.5-2
H_2	10
CO	20
CO_2	10
H_2O	10
N_2	48.49.5

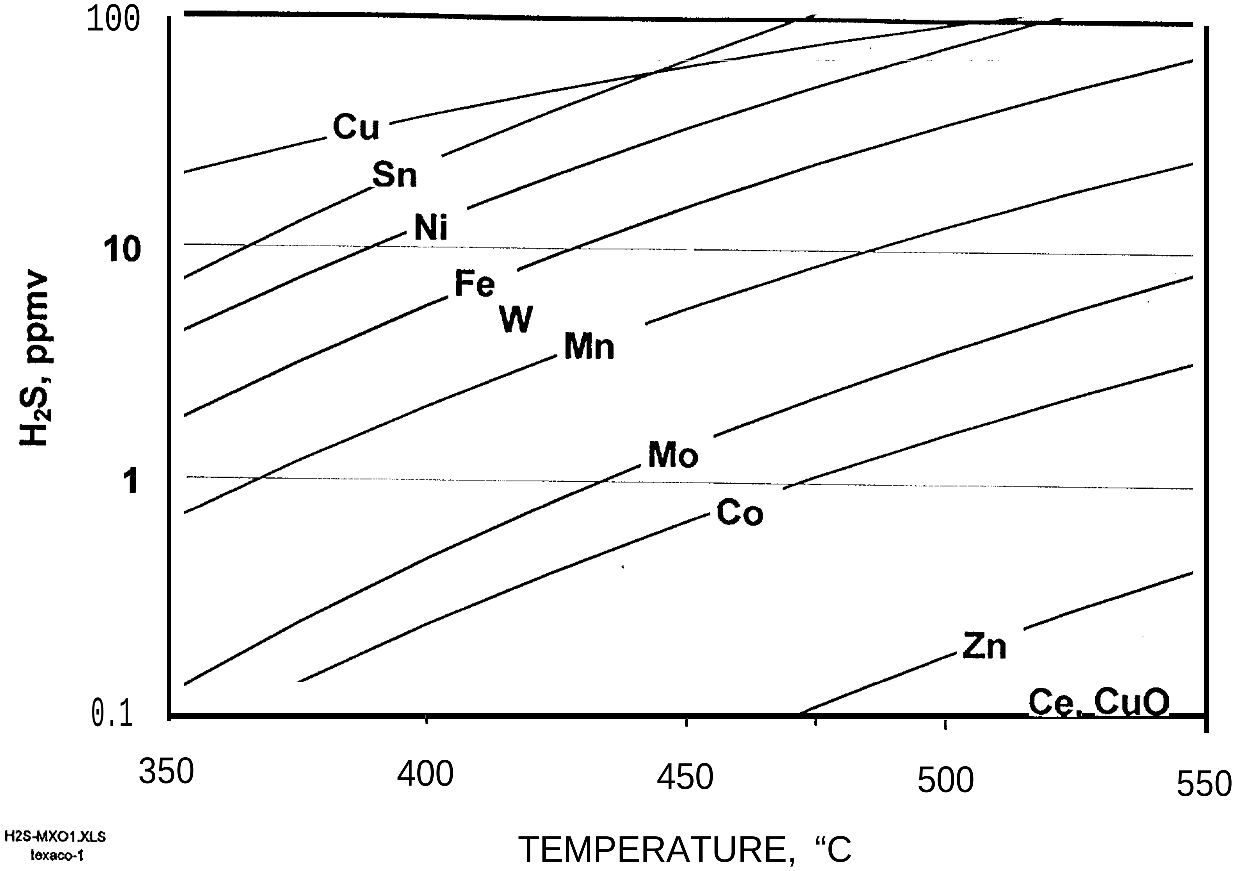
METAL OXIDE SCREENING

Function	Metals Chosen
Primary Desulfurization	Cu, Fe, Zn, Mn, Ce
Secondary Additives	Cu, Fe, Mn, Cr, Ce, Co, Mo
Support/Stabilizers	$Ti0_2$, ZrO_2, Al_2O_3, Binders

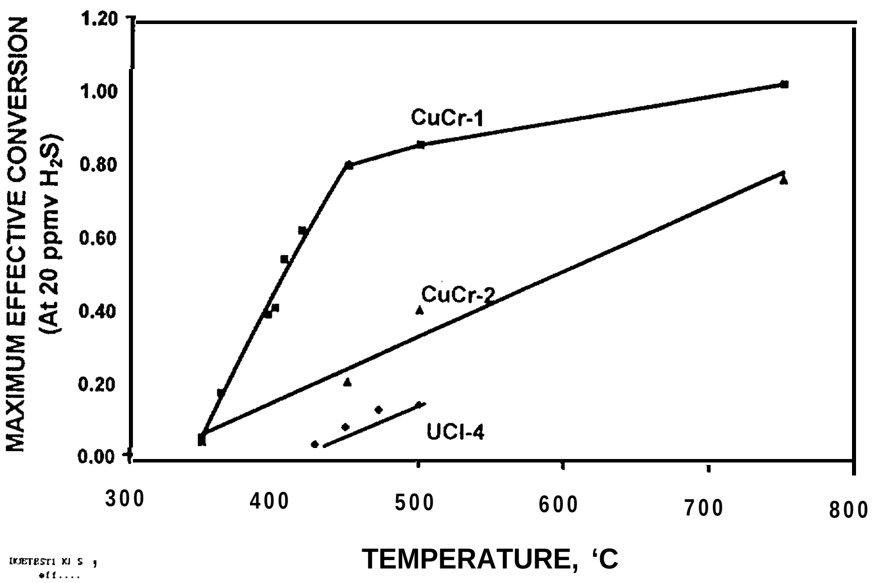
H_2S Equilibrium Concentration: U-GAS



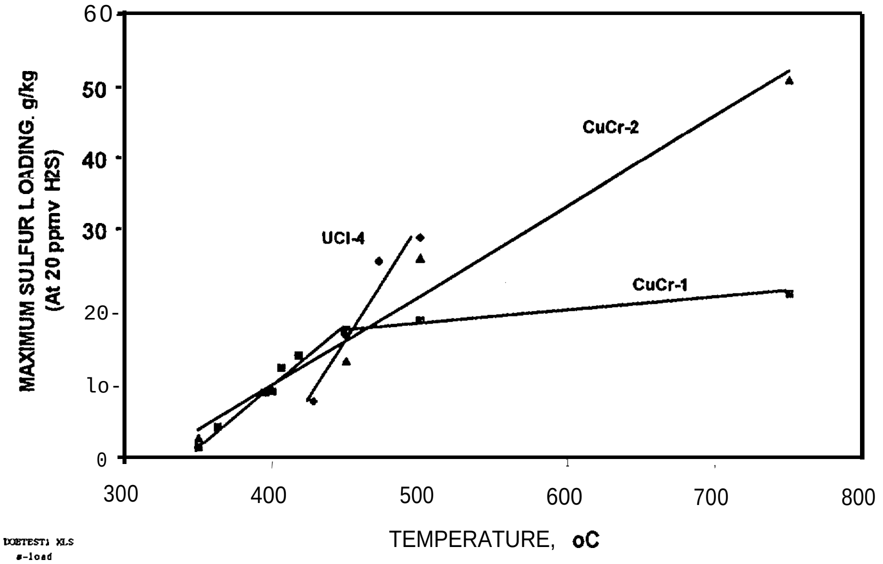
H_2S Equilibrium Concentration: TEXACO



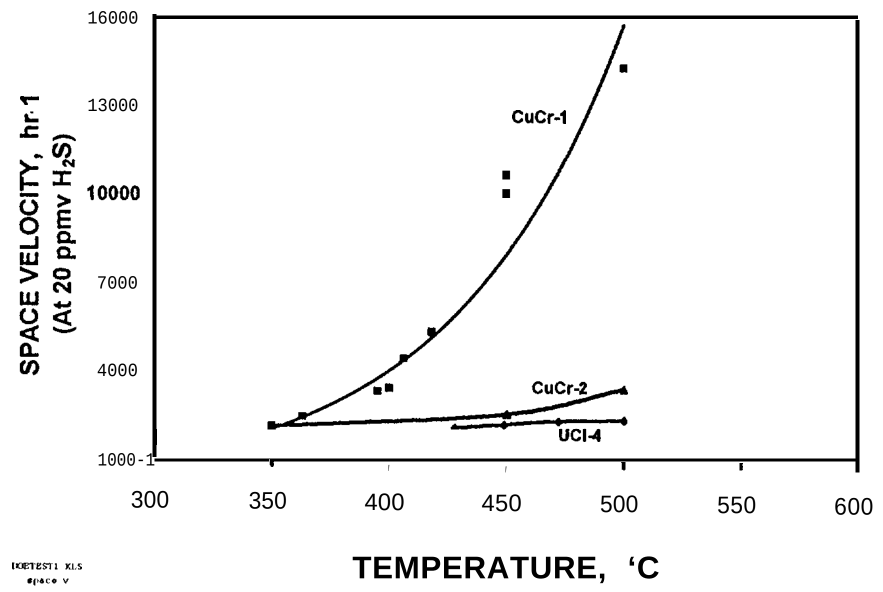
Comparison of effective conversion



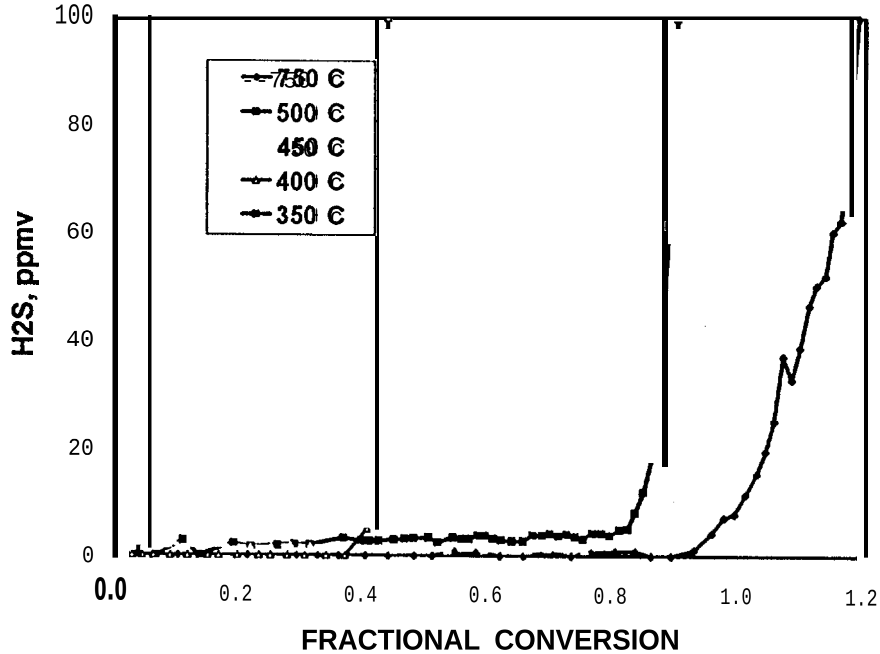
Comparison of Sulfur Loading



Sorbent Performance at different Space Velocity

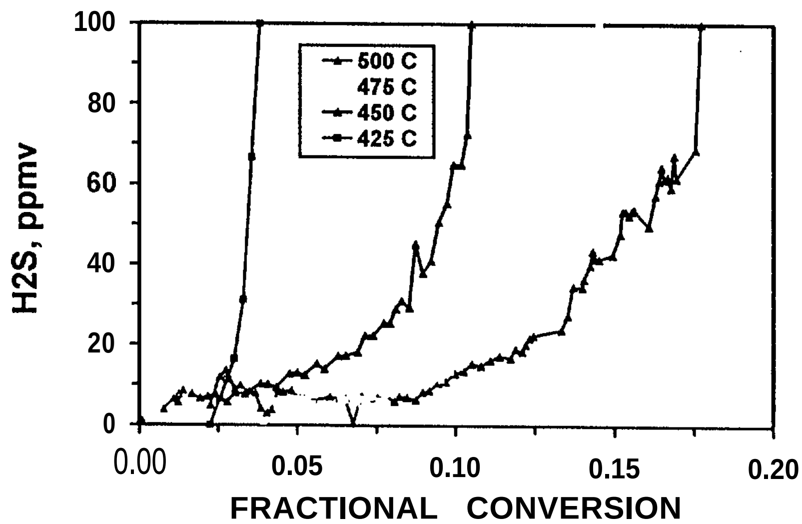


Performance of CuCr-1 at Different

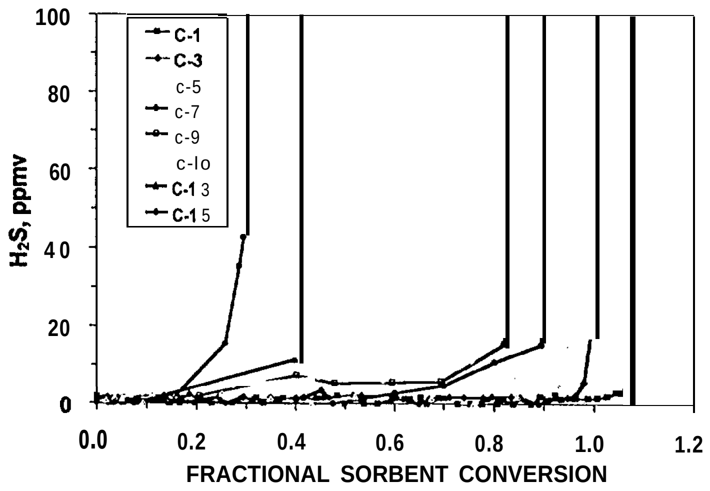


DOETEST1 XLS

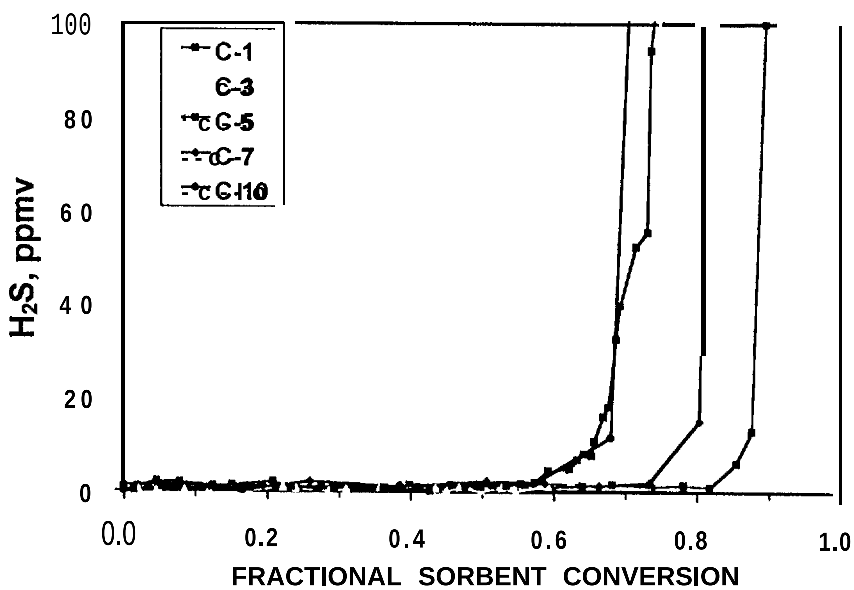
Performance of Zinc Titanate at Different Temperatures



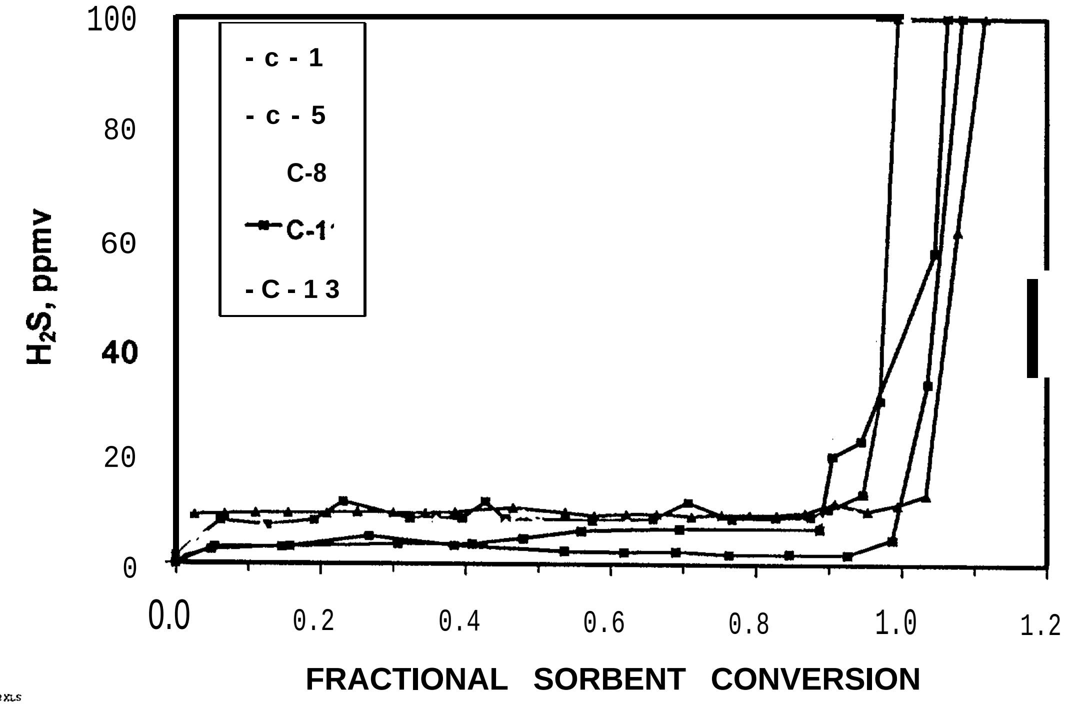
Performance of IGTSS-1 at 350“C



Performance of IGTSS-6 at 350“C

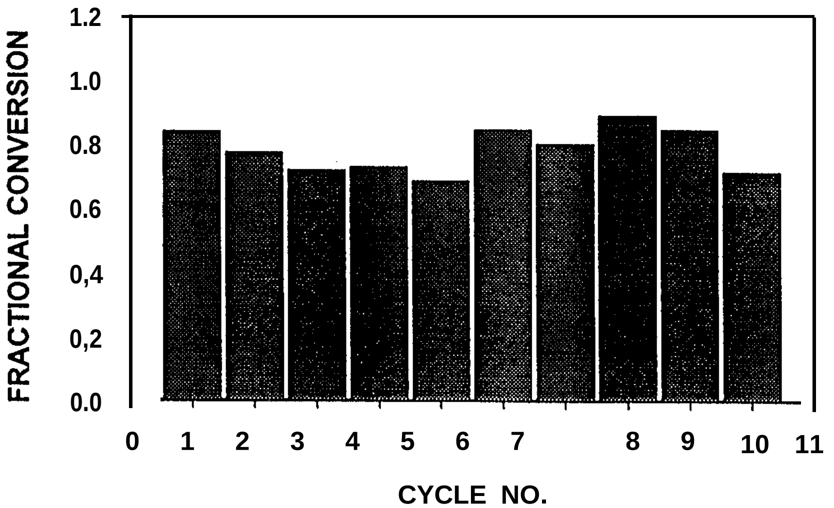


Performance of IGTSS-11 at 450“C



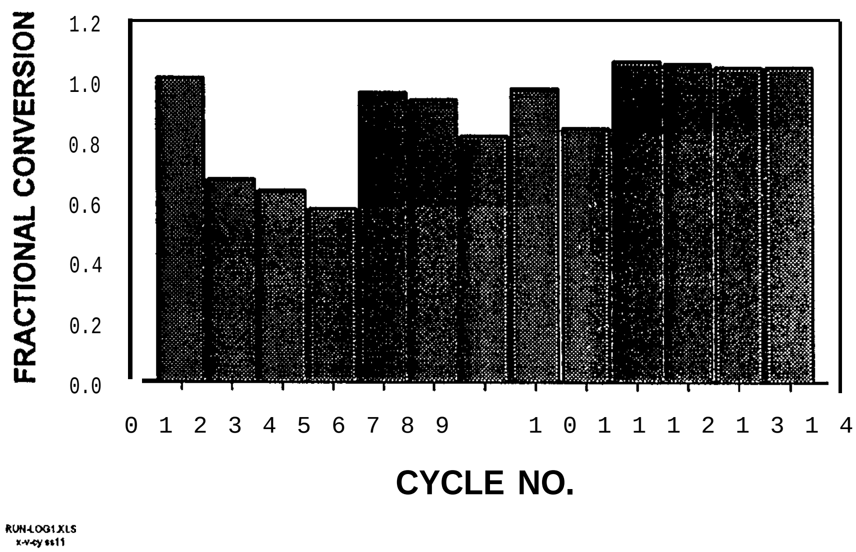
SERIE S2XLS

Breakthrough Conversion of IGTSS-6 at 350 “C

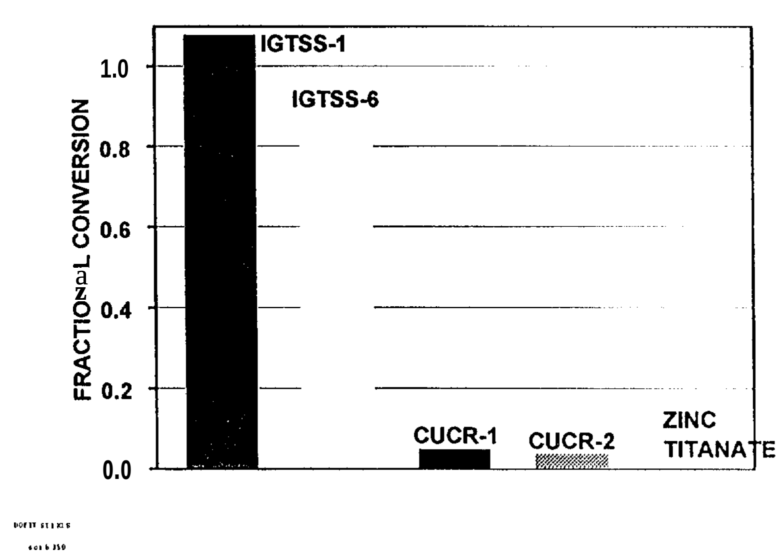


RUN-LOG1.XLS

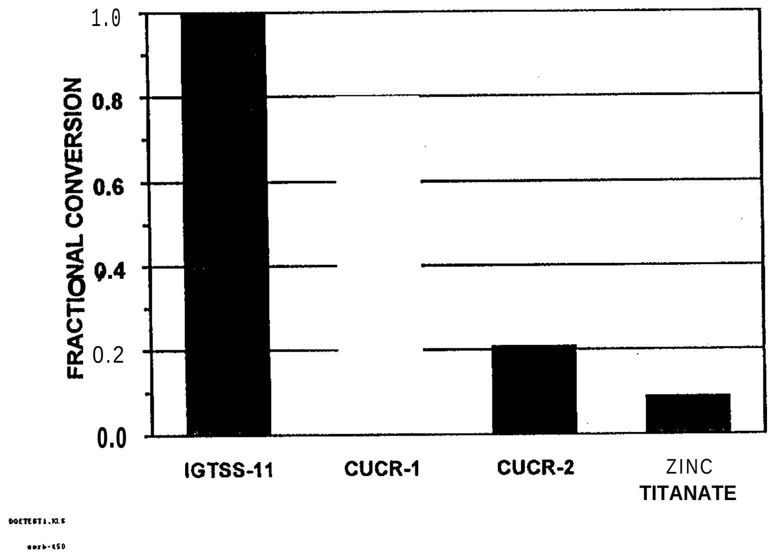
Breakthrough Conversion of IGTSS-11 at 450 *C



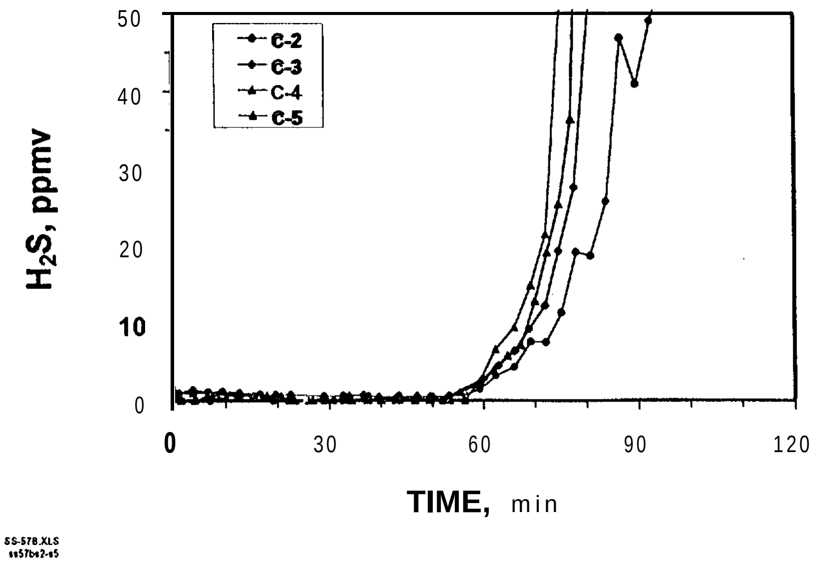
Comparison of Sorbent Conversion at 350 *C



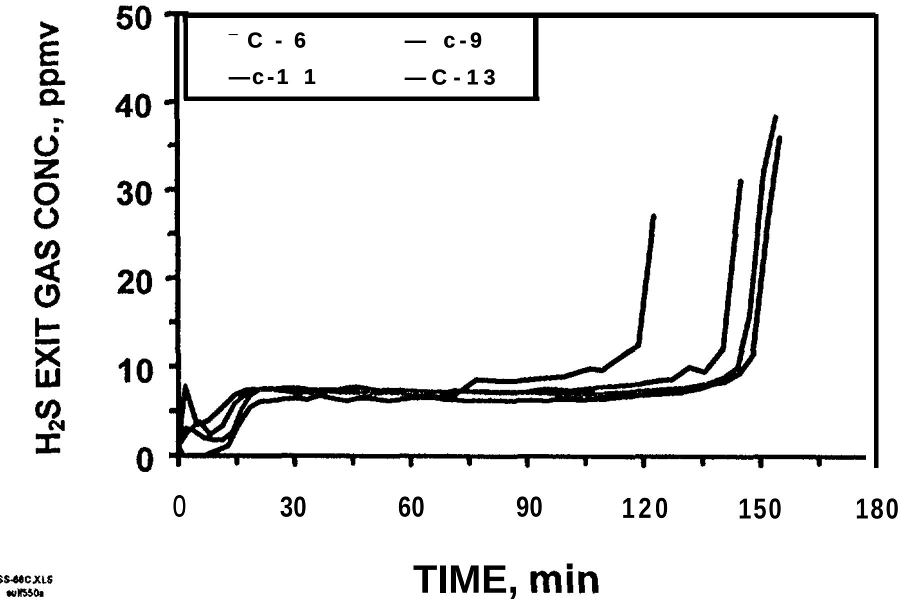
Comparison of Sorbent Conversion at 450“C



Performance ofIGTSS-57at450“C



Performance ofIGTSS-68at550“C



Conclusions

- Three promising sorbents have been developed for temperature range of 350^o-550^oC

- The sorbents reactivities and sulfur capacity appear to be stable or improving during the first 10 to 15 cycles

- The attrition resistance of the sorbent are better than UCI-4169 Zinc Titanate sorbent.