

## H<sub>2</sub>S Catalytic Oxidative Decomposition at high temperature

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### Highlights

- H<sub>2</sub>S oxidative decomposition in presence of catalyst was studied at high temperature.
- Alumina-based catalysts were prepared and characterized.
- The influence of the temperature and the contact time were investigated.
- An approach to the equilibrium was obtained in the overall range of temperature.

### 1. Introduction

Hydrogen sulfide is an acid compound, flammable, corrosive and extremely toxic. It's by-product of the refinery processes (e.g. hydrodesulphurization) and due to the nature it is considered a waste stream and therefore the industrial application are very limited [1]. Today the hydrogen sulfide is converted to Sulphur and water by the Claus process ( $\text{H}_2\text{S} + \frac{1}{2} \text{O}_2 \rightarrow \frac{1}{x} \text{S}_x + \text{H}_2\text{O}$ ) for the final production of the sulfuric acid [2]; it isn't a profitable process because the price of the Sulphur is depressed and the H<sub>2</sub> is lost as low grade steam. In order to obtain, not only Sulphur, but also hydrogen, an innovative process could be the catalytic decomposition of H<sub>2</sub>S in presence of a sub-stoichiometric concentration of oxygen, in order to obtain Sulphur, hydrogen and water. The catalyst plays a key role to improve the selectivity of the process towards Sulphur and hydrogen, depressing the Sulphur dioxide (SO<sub>2</sub>) formation, the which presence, related to the oxygen addition, is much favored from a kinetic point of view.

In our previous works, we have studied the reaction of H<sub>2</sub>S oxidative decomposition in homogeneous phase [3-4] and in presence of alumina-based catalyst by developing in both the cases a macroscopic kinetic model able to describe, also if in simplified manner, the complex reaction system [5].

### 2. Methods

The Al<sub>2</sub>O<sub>3</sub> catalyst was synthesized by thermal treatment of pseudoboehmite at 900 °C for 12 h in air in order to obtain the stabilization of the alumina phase. The fresh catalyst was characterized by X-Ray diffraction and adsorption of nitrogen at -196 °C.

The catalytic activity tests were performed in the laboratory apparatus described in detail in our previous work [3-4]. Experiments were carried out in a fixed bed quartz tubular reactor with 300 mm length and internal diameter of 12 mm.

Sulphur and other solid species produced by the reaction were trapped by using a quartz-wool filter placed at the end of the reactor in the quenching zone. In order to avoid SO<sub>2</sub> absorption in the water produced from the reaction, a cold trap was placed after the quenching zone to remove selectively sulphur and water [5].

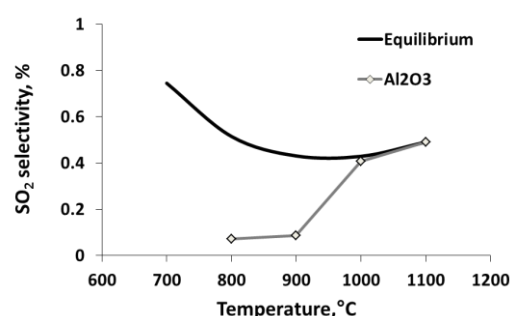
The exhaust stream was analysed by a quadrupole filter mass spectrometer .

The catalytic activity tests were performed by varying the temperature (700-1100°C), contact time (20-40 ms) at fixed H<sub>2</sub>S concentration (10 vol%) and molar feed ratio(O<sub>2</sub>/H<sub>2</sub>S=0.2).

### 3. Results and discussion

The XRD spectra have shown the presence of mixed phases of  $\text{Al}_2\text{O}_3$ ; the structural transformation of a fraction of the  $\gamma$  phase to the  $\theta$  phase structure is also confirmed by the value of SSA that was lower than the one observed for  $\gamma$ -alumina.

The influence of the temperature was studied with and without the catalyst at fixed contact time of 33 ms. The experimental  $\text{SO}_2$  selectivity is even lower than the equilibrium data up to 900 °C, while for higher temperatures it is possible to observe an approach to the equilibrium, maybe because it begins to be significant the contribution of the homogeneous reactions (Figure 1).



**Figure 1.** Comparison of  $\text{SO}_2$  selectivity between catalyst and equilibrium value as a function of temperature ( $\text{H}_2\text{S}_{\text{IN}} = 10$  vol %,  $\text{O}_2/\text{H}_2\text{S} = 0.2$ ).

In homogeneous phase, the  $\text{SO}_2$  selectivity was equal to 4% and higher than that expected from the thermodynamic equilibrium at 1000 °C (~0.5%).

### 4. Conclusions

The oxidative decomposition of hydrogen sulfide was investigated by employing an alumina based-catalyst. Characterization data of the obtained catalyst evidenced the presence of mixed phases of  $\text{Al}_2\text{O}_3$  ( $\gamma$  and  $\theta$ -phase). The catalytic performance has been investigated at different operating conditions (reaction temperature and contact time). In the overall range of the investigated temperature (700–1100 °C), it was observed that  $\text{H}_2\text{S}$  conversion,  $\text{H}_2$  yield, and  $\text{SO}_2$  selectivity were very close to those ones expected by the thermodynamic equilibrium. No significant changes in  $\text{H}_2\text{S}$  conversion,  $\text{H}_2$  yield, and  $\text{SO}_2$  selectivity were obtained by increasing the contact time.

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### Keywords

“ $\text{H}_2$  Production”, “ $\text{H}_2\text{S}$  Oxidative Decomposition”, “Hydrogen Sulphide”, “Catalyst”.

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