

Novel insights on H₂S and COS removal for gas purification applications

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H₂S and COS are well-known gaseous pollutants encountered in many applications. Synthetic fuels produced from thermochemical routes (Fisher-Tropsch based XTL processes) or IGCC technologies for cleaner power production are among the applications for which the removal of H₂S and COS from intermediate synthesis gas products is critical [1]. Indeed, H₂S and COS are known severe poisons for FT catalysts, corrosive for industrial equipments, but also acute environmental pollutants and display high toxicity. Natural gas and biogas applications also face similar issues which require H₂S and/or COS removal.

Some fundamental studies conducted at IFPEN have dealt with sulfur removal issues from gases. A review of some recent studies will be presented.

A part of our work focused on kinetics modeling and solid-state transformations involved in metal oxides sulfidation and oxidizing regeneration reactions, in the scope of H₂S removal processes over metal oxides. We questioned some generally admitted assumptions regarding ZnO sulfidation mechanism and kinetics modeling [2]. With the support of materials characterizations such as 3D-TEM, we evidenced that an outward growth process of the ZnS phase upon ZnO crystals was more likely to occur, compared to inward development mechanisms generally assumed. Updated mechanism and kinetics modeling were also proposed. Regeneration properties of various metal oxides were also studied [3], showing that mixed metal oxides are for this purpose more adapted, and that lower regeneration temperature can be reached.

We also interested in the understanding and the modeling of dedicated COS adsorption and catalytic based removal processes. We focused on the study of the interactions taking place in COS adsorption on doped or undoped oxides (Al₂O₃, TiO₂ and ZrO₂) [4]. Sorbents stability over adsorption-regeneration cycles was also investigated. Besides, we explored the modeling of catalytic processes used for COS and HCN elimination in the field of synthesis gas purification applications [5]. A reaction model was developed considering hydrodynamic, mass transfer limitations, and Langmuir-Hinshelwood reaction mechanisms for both COS and HCN hydrolysis reactions. LH rate laws accounted for the detrimental effect of gaseous reactants and products upon COS and HCN conversion kinetic rates.

Our work included in a multi-technique and multi-scale approach brings novel and rational insights on reactions involved at the heart of H₂S and COS sorption and catalytic removal processes. This finally leads to rational pathways to a better design of industrial sorbents, catalysts and processes.

[1] D. Chiche et al, *Oil Gas Sci Technol*, **2013**, 68(4), 707-723

[2] L. Neveux et al, *Chem Eng J*, **2012**, 181-182, 508-515; L. Neveux et al, *Phys Chem Chem Phys*, **2013**, 15(5), 1532-1545

[3] V. Girard et al, *Fuel*, **2014**, 128, 220-230; V. Girard et al, *Fuel*, **2015**, 140, 453-461

[4] To be published

[5] D. Chiche et al, *Appl Catal B*, **2017**, 205, 189-200