Analysis of Transport-Kinetics Interactions in Complex Commercial Catalyst Shapes for Improved H_2SO_4 Manufacturing Processes

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Introduction

Development of next-generation chemical processes that have zero emissions is a key environmental objective for sustainable development. The manufacture of H_2SO_4 by the air oxidation of SO_2 to SO_3 is an important technology where an opportunity exists for new catalyst development and process innovation by reducing emissions of unconverted SO_2 in process reactor tail gases owing to the sheer number (> 1500) and scale (ca. 500 to 4500 metric tons/day) of typical plants. The global supply of H_2SO_4 is projected to grow from 200 MM tonnes in 2006 to more than 258 MM tonnes in 2015 with a value of > \$10 MMM [1]. Hence, an opportunity exists to develop new innovations in environmental catalysis and reaction engineering for an important technology that has a rich and long history with positive economic growth.

H_2SO_4 catalyst manufacturers offer a variety of catalyst shapes for the air oxidation of SO_2 to SO_3 using adiabatic multi-stage reactor systems. These shapes include solid and hollow cylinders as well as those involving hollow multi-lobe ribs, such as 5-lobe and 6-lobed shapes. These shapes have evolved over time due to accumulated experience and the use of heuristic knowledge without any notable application of environmental catalytic reaction engineering principles. The availability of advanced modeling tools and experimental techniques provides an opportunity to invent next-generation catalysts and processes.

The primary objective of this presentation is three-fold: (1) to review the current state-of-the art in modeling transport-kinetic interactions for catalyst particle shapes utilized in the oxidation of SO_2 to SO_3; (2) to develop a rigorous modeling framework that accounts for diffusion and non-isothermal reaction in various realistic 3-D commercial catalyst shapes for the SO_2 oxidation, such as solid cylindrical pellets, rings, and multi-lobe particles, using various diffusion flux models; and (3) to employ this framework to compare the performance of these various catalyst shapes under typical multi-pass convertor operation.

Methods

Some catalyst shapes that have been proposed for SO_2 oxidation to SO_3 are illustrated in Figure 1 [2]. The ones shown here are not exhaustive, but provide a perspective of shapes that have been proposed in various patents and the open literature. In the case of catalyst shapes involving multiple lobes, the shape parameters include the number and shape function of the lobes, the characteristic dimension of a given lobe, the distance from the lobe center to true particle center,
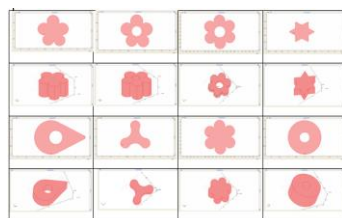

Figure 1-Illustration of catalyst shapes

and the diameter of the hollow center region.

COMSOL Multiphysics is used to model non-isothermal diffusion and reaction with compositional and temperature dependence of all model parameters, which builds upon our previous work [2]. The reaction kinetic model is based upon the work of Collina *et al.* [3] since it accounts for the dependence of the SO_2 oxidation rate on the partial pressures of SO_2, O_2, and SO_3 with inhibition by both SO_2 and SO_3. The reaction rate and adsorption equilibrium parameters in this model are valid from 420 to 550°C, which is within the commercial operating range of most convertors.

Various diffusion flux models, namely, the Wilke model, the Wilke-Bosanquet, and the Dusty Gas model, are not available in COMSOL so these were coded to compare the effect of the flux model on the particle concentration and temperature profiles as well as the particle effectiveness factor over process conditions encountered in multi-pass convertor operation.

Results and Discussion

Typical SO_2 concentration profiles for a multi-lobe particle shape whose lobes are described by a rounded step function are shown in Figure 2a. In this case, the Wilke model is used to describe the diffusional fluxes. Here, the particle surface is exposed to a constant bulk concentration and temperature. A cross section of the temperature profiles inside the particle at the particle mid-section is shown in Figure 2b. The results show that diffusional limitations exist. The lobe regions represent zones having the greatest concentration and temperature gradients, although the lobes serve to reduce pressure drop.

The particle model provides the starting basis for coupling local particle behavior to the external particle field equations, which is an essential component of SO_2 oxidation reactor modeling. Concepts for reduction of emissions by particle optimization will be described.
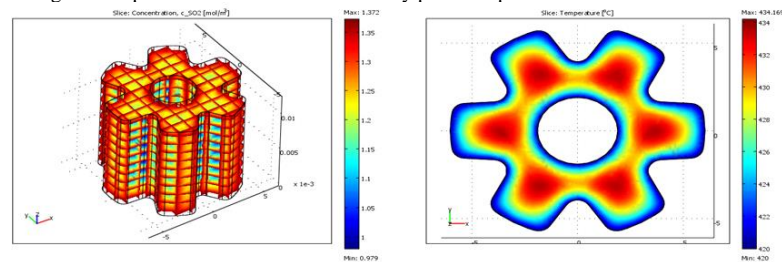

Figure 2a. SO_2 Concentration profiles for a novel shape. Figure 2b-Catalyst temperature profiles at the mid-section.

References

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