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Simulation of an industrial adiabatic multi-bed catalytic reactor for sulfur dioxide oxidation using the Maxwell–Stefan model

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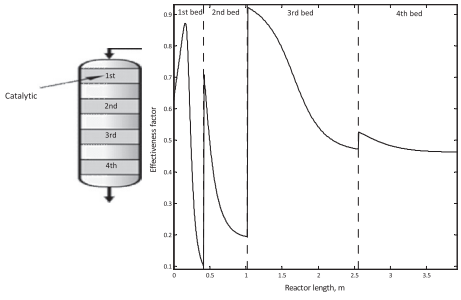
HIGHLIGHTS

- A rigorous model was proposed for the analysis of a SO_2 oxidation catalytic reactor.
- The Maxwell–Stefan diffusional model was justified from thermodynamic correction factor matrix.
- The model allowed evaluating the reactor behavior with errors lower than 3%.

GRAPHICAL ABSTRACT

Catalytic performance – Effectiveness factor profiles in the multi-bed reactor.



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ABSTRACT

In this work, a rigorous heterogeneous model for the analysis of an industrial adiabatic multi-bed catalytic reactor for sulfur dioxide oxidation was developed. It was based on the Maxwell–Stefan diffusional model, which was selected from the analysis of the Maxwell–Stefan diffusivity and the thermodynamic correction factor matrix. The reactor model, implemented in MatLab®, allowed evaluating truthfully the behavior of each catalytic bed (e.g., concentration, conversion, pressure and temperature profiles) with errors lower than 3%. Additionally, the effectiveness factor variation along the reactor was estimated. Thus, a better understanding of the effect of diffusional resistances on reactor performance was possible.
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1. Introduction

The manufacture of sulfuric acid from sulfur involves three important steps: (1) the formation of sulfur dioxide (SO_2), (2) its catalytic oxidation to sulfur trioxide, SO_3 (a reversible and highly exothermic reaction – ca. 96,000 kJ/kmol of SO_3 formed) and (3) the reaction of sulfur trioxide with H_2O in absorption towers. Sulfuric acid is recognized as very important commodity chemical. Only in 2012, its world production was about 220 million tonnes. Most of it is used for the production of phosphatate fertilizers, calcium dihydrogen phosphate, and ammonium phosphates [1].

In order to start the catalytic oxidation reaction, the temperature of the feeding gas to the converter reactor must be high

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Nomenclature

Latin letters

c_{pi}	specific heat capacity of component i, kJ/(kmol K)
c_i	molar concentration of component i, kmol/m^3
$D_{ij,ef}$	effective molecular diffusivity of component i binary with j, m^2/h
$\underline{\underline{D}}_{ij,ef}$	Maxwell–Stefan diffusivity, m^2/h
$D_{i,Kn,ef}$	Knudsen diffusivity for component i, m^2/h
D_p	diameter of particle in the bed, m
G_o	superficial mass velocity, kg/(m^2 h)
J_i	diffusion flux of species i relative to the molar average reference velocity, kmol/(m^2 h)
k_i	reaction rate constant
K_p	equilibrium constant
L	reactor length, m
M_i	molecular mass of component i, kg/mol
N_i	molar flux of species i in a stationary coordinate frame of reference, kmol/(m^2 h)
N_t	total molar flux, kmol/(m^2 h)
P	total pressure, bar
p_i	partial pressure of component i, bar
R	universal gas constant, J/(mol K)
r_{SO_2}	intrinsic SO_2 rate of reaction, kmol SO_2/(kg cat h)
r_{js}	rate of reaction of component j at the catalytic surface, kmol/(kg h)
T	temperature of the reactor, K
X_{SO_2}	conversion of SO_2, dimensionless
x_i	mole fraction of component i, dimensionless
z_p	half-thickness of the catalyst particle, m

Greek letters

∇	gradient
η	effectiveness factor
ΔH_{Rxn}	heat of reaction, kJ/kmol
ε	porosity, m^3 (gas)/m^3 (cat)
ε_B	bed voidage fraction, dimensionless
Γ	thermodynamic correction factor matrix
μ_G	viscosity of the gas mixture, kg/(m h)
ρ_G	density of the gas mixture, kg/m^3
ρ_B	bulk density of the catalyst bed, kg/m^3
τ	tortuosity, dimensionless

enough to activate the catalyst (e.g., 688–700 K for vanadium pentoxide based catalyst) and next to guarantee a high reaction rate. As the gas passes through the reactor, the exothermic reaction proceeds and the gas temperature increases. The reaction rate is dependent on catalyst activity and approach to equilibrium. The reaction extent depends on the reaction rate and the residence time of the gas in contact with the catalyst in the reactor. Several papers on modeling and simulation of SO_3 oxidation have been published in the open literature. Some relevant characteristics related to both catalyst pellet and reactor modeling can be summarized as follows:

- **Catalytic pellet:** In an early attempt, Olson et al. [2] have studied experimentally the importance of diffusion in the catalytic oxidation process. Later, Livbjerg and Villadsen [3] analyzed the kinetics and effectiveness factor for the industrial vanadium catalyst. They restricted their study to a very narrow range of experimental data (727–757 K) and to an intraparticle effective diffusivity model. Some inadequacy of this model was pointed out by Davis et al. [4]. They improved the analysis using the dusty-gas model and a single value for the tortuosity factor. In fact, other authors also discourage the use of the Fickian type models because they do not represent qualitatively the diffusion process in multicomponent systems [5]. However, effective diffusion coefficient approach, though it fails in the mechanistic sense, is nevertheless adequate in several situations and could be safely used for reactor simulation studies in such cases. Nevertheless, the basic problem is how to identify these *a priori*. In the present work, it was done using the procedure of Reddy and Murty [6].
- **Packed-bed oxidation reactors modeling:** Young and Finlayson [7] and Rosendall and Finlayson [8] proposed a model including radial and axial dispersion, heterogeneity, and density variations. Some criteria were developed to predict *a priori* which effects should be included. More recently, Nodehi and Mousavian [9] used a model for the simulation and optimization of a multi-bed adiabatic reactor, including a heterogeneous plug flow model (expressing the mass transfer by Fick's law with an effective diffusivity coefficient). A dynamic model was also proposed by Kiss et al. [1]. They include the catalytic reactor (five pass converter), heat exchangers, mixers, splitters and reactive absorption columns. The kinetic parameters were fitted to the real plant data.

In this work, a rigorous heterogeneous model for the simulation of an industrial adiabatic multi-bed catalytic reactor for sulfur dioxide oxidation is developed. The reactor comprises one cylindrical vessel which acts as a fixed bed reactor with four separate beds of catalyst. For a satisfactory yield of sulfur trioxide (above 99.5% conversion is needed), the applied temperature is as low as economically possible. Thus, the heat is removed and recovered from the gas leaving each bed using heat exchangers. The produced sulfur trioxide is removed from the reactor between its third and fourth bed and passed to the next stage, where the sulfur trioxide is converted into sulfuric acid. However, a low amount of sulfur dioxide remains without conversion and is returned to the reactor through the fourth bed of catalyst and then the resulting gases, mainly sulfur trioxide, flow to the next stage of the process. In the view of the state of the art of previously proposed models, the following advances of this work can be pointed out: (i) the suitable model for diffusion and reaction in the catalyst pellet (vanadium (V) oxide supported on silica) was justified basing on the methodology proposed by Reddy and Murty [6] and confirmed by a comparison of the results obtained using the dusty-gas, the Maxwell–Stefan, and the effective diffusivity (Fick) models; (ii) the catalyst effectiveness factor was calculated and evaluated for each catalytic bed; (iii) model validation was done comparing the simulation results with plant data.

2. Mathematical model

2.1. Reaction and rate expression

The exothermic, gas phase, reversible reaction under study can be expressed as follows:

$$SO_{2,(g)} + 1/2O_{2,(g)} \rightleftarrows SO_{3,(g)} \quad (1)$$

The following factors can increase the formation of SO_3: high pressure (this is not economical due to the high energy cost to compress the gas and the high capital cost to build vessels strong

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