Heterogeneous reaction kinetics and transport modeling in catalytic foam

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Highlights

- A heterogeneous model of foam unit cell was prepared using BCC (Body Centered Cubic structure) geometry.
- Reaction and diffusion phenomenon in the foam unit cell was studied for LHHW (Langmuir Hinshelwood Hougen Watson) and general order kinetics.
- The cases of isothermal and non-isothermal catalyst wall were studied using FEM (Finite Element Method) based simulations.
- Effectiveness factor vs Thiele modulus correlation was arrived at for foam based catalyst and compared with correlations published in the literature for spherical catalyst pellets.

1. Introduction

A heterogeneous model of the foam unit cell was prepared using Body Centered Cubic Structure (BCC) geometry to study reaction and diffusion in foam based catalysts. Kinetic expressions like general order kinetics with an order of 0.5 and Langmuir-Hinshelwood-Hougen-Watson (LHHW) kinetics for isothermal and non-isothermal catalyst wall cases were studied using finite element method based simulations. The effectiveness factor vs. the Thiele modulus relationships for LHHW and general order kinetics are developed for foam catalysts based on Rajadhyaksha et al.'s[1] approach for spherical catalyst pellets in both isothermal and non-isothermal catalytic wall cases.

2. Methods

Foam is different from typical catalyst pellets in that the diffusion of reactants is outward from hollow space filled with fluid (compared to inward diffusion of reactants in conventional catalyst pellets). The BCC geometry was used to represent the foam unit cell [2] for modelling the reaction vs. diffusion phenomena in the catalyst wall (Figure 1). The concentration and temperature profiles from the 3D unit cell model can be used to calculate the effectiveness factor. [3]

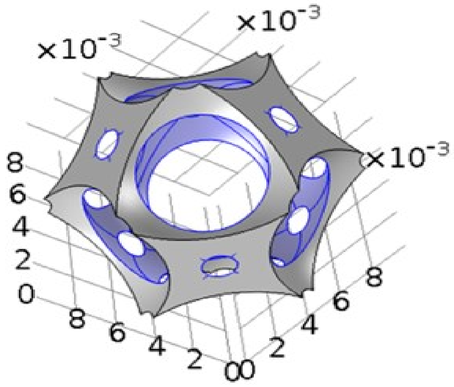

Figure 1. BCC geometry for foam unit cell

Mole and heat balance equations used in the model are given below.

$$\nabla.(-D_i \nabla C_i) + u.\nabla C_i = R_i \qquad (1)$$

For any species i, C_i is the concentration of species i, R_i is the reaction rate for species i, D_i is the diffusion coefficient for species i and u is the velocity of the bulk flow.

$$\rho C_p u.\nabla T + \nabla.q = Q \qquad (2)$$

Where, q = -k ∇T $\qquad$ (3)

Where, Q represents the heat source or sink, denoted appropriately by a positive or negative sign, ρ is the density of the fluid, C_p is specific heat and T is the temperature.

3. Results and discussion

The effectiveness factor vs. Thiele modulus relationship for LHHW and General order kinetics for foam follows Aris' correlation[4], if Thiele modulus, ∧ in Aris' correlation is replaced by modified Thiele modulus ∧′ as defined by Rajadhyaksha et al.[1]

To put this explanation in equation form,

$$\eta = \frac{\tan h\wedge'}{\wedge'} \qquad (4)$$

Figures 2 (a) and (b) show the comparison of the effectiveness factor vs. modified Thiele modulus from simulations vs. calculations (equation (4)) with a small deviation at $\wedge' > 2$.

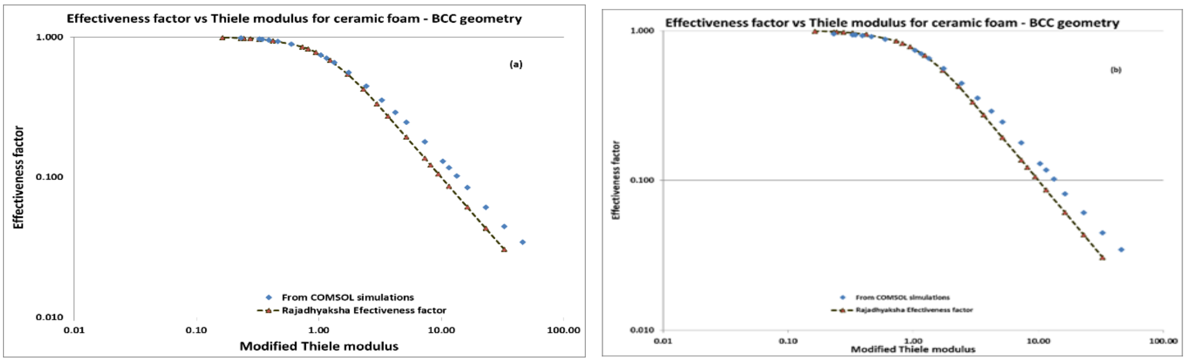

Figure 2. Simulated vs. calculated effectiveness factors for BCC geometry for general order (n=0.5) reaction at (a) -80 kJ/mol heat of reaction and (b) +800 kJ/mol heat of reaction].

4. Conclusions

It was observed that by using foam diffusion length from Makhania and Upadhyayula's work[3] and replacing the Thiele modulus with a modified Thiele modulus as defined by Rajadhyaksha et al.[1], the effectiveness factor vs. Thiele modulus relationship assumes a form similar to that of Aris' relationship. The thermal conductivity of the foam based catalysts is high, such that the temperature gradient in the foam structure is small even for the reactions with large heat of reaction. Because of this, the non-isothermal correction factor [1]does not have a significant impact on the effectiveness factor vs. the modified Thiele modulus relationship.

References

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Keywords

Foam; Effectiveness factor; Modified Thiele modulus; LHHW and general order kinetics