A non-isothermal, transient model of a non-catalytic reaction in a packed bed with a multi-component high-temperature gas mixture

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Abstract

A mathematical model has been developed to simulate heterogeneous reactions with a complex set of physico-chemical and thermal phenomena in a packed bed. These include the chemical reaction itself, the transport of gaseous species in the pores, the generation or consumption of heat by the reactions, and the transport of heat in the porous domain. Solution of complicated sets of governing equations is based upon the concept of a finite volume fully implicit approach. The model has then been applied to analyze the reduction of a hematite pellet in a moving packed bed reactor.

Keywords: Gas–solid reaction; Porous media; Finite volume; Moving packed bed; Heat transfer

1. Introduction

The modeling of high-temperature flows in porous media is a challenging task, involving fluid mechanics, heat, and mass transfer together with chemical reactions. Such flows are observed in chemical and metallurgical reactors. Relying largely on the computational effort, much of the work in this field has been based on relatively simple formulations such as pseudo-steady state, isothermal, single component, and non-reactive [1–3]. The aim of the present paper is to demonstrate a general model with a lower simplicity of heterogeneous gas–solid reactions in a continuum porous media as a pellet. External transfer of gaseous species to the surface of the pellet, gaseous transport within the pores, the heterogeneous exo- and endothermic reactions, and the heat-transfer process inside the porous domain are formulated and included in the set of equations. This model could, therefore, be described as a transient, non-isothermal, multi-component, and reactional flow in a porous media of a pellet.

2. Concepts and mathematical formulation

The system under study is a porous specimen pellet of a packed bed that is generally made up of a number of grains of different sizes separated by pores. A virtual depiction of the pellet is shown in Fig. 1. The heterogeneous reactions of type Eq. (1) take place inside a particle of this sort immersed in a multi-component gas mixture, where A and P are solid reactant and product, respectively; B_i and Q_i are gaseous reactants and products, respectively, and a, b_i, p, and q_i are stoichiometry coefficients.

$$aA_{(s)} + b_iB_i(g) \rightarrow pP_{(s)} + q_iQ_{i(g)} \tag{1}$$

The basic assumptions of the model are:

- The pellet considered is spherical and there is no change in diameter.
- The reactions proceed independently without interaction between the gaseous species.
- The porosity of pellet will not be changed during the reaction.

2.1. Reaction modeling

A special grain model has been used in order to deal with heterogeneous reactions in porous pellets. It is further assumed that the chemical reaction and gaseous diffusion proceed simultaneously. Therefore, the reaction proceeds nearly homogeneously in a macroscopic sense and proceeds topochemically in microscopic view. Hence, the reaction rates are usually expressed as [3]:

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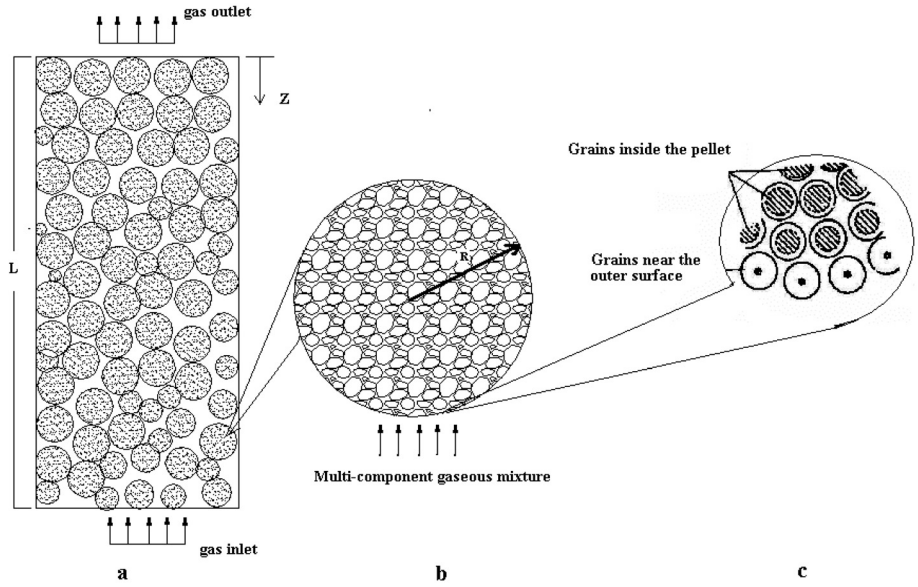

Fig. 1. Schematic configuration of a packed bed, pellet, and grain model of a porous pellet.

$$\dot{R}_i = \frac{3k_{r,i}}{r_g}\left(1+\frac{1}{Ke_i}\right)(1-f)^{2/3}(C^n_{B_i}-\frac{C^l_{Q_i}}{Ke_i}) \quad (2)$$

$$k_{r,i} = k_{o,i}Exp\left(\frac{-E_{a,i}}{\bar{R}T}\right), f = 1-\left(\frac{r_j}{r_g}\right)^3, \frac{l}{q_i}=\frac{n}{b_i} \quad (3)$$

$$\frac{\partial f}{\partial t} = \sum_i \left(\frac{3k_{r,i}}{\rho_A r_g}\right)\left(1+\frac{1}{Ke_i}\right)(1-f)^{2/3}(C^n_{B_i}-\frac{C^l_{Q_i}}{Ke_i}) \quad (4)$$

where C is the concentration of the reactant, Ea is the activation energy, f is the local conversion, k_r is the reaction rate constant, k_o is the frequency factor, Ke is an equilibrium constant, $\bar{R}$ is a gas constant, $\dot{R}$ is the overall rate of chemical reactions, r_g and r_j are the radii of a grain in a pellet and reaction interface within the grain, respectively, ρ is the molar density of solid reactant, t is time, and T is temperature.

2.2. Conservation equations

The heat equation including conductive and radiant parts is written as follows:

$$(\rho C_P)_{eff}\frac{\partial T}{\partial t} = \nabla.(\lambda_{eff}\nabla T) + (1-\varepsilon)\sum_i(-\dot{R}_i)\Delta H_i \quad (5)$$

The mass equation for each gaseous species as reactant B_i or product Q_i is obtained as follows:

$$\varepsilon\frac{\partial y_i}{\partial t} = \nabla.(D_{eff,i}\nabla y_i) + \frac{(1-\varepsilon)(-b_i\dot{R}_i)}{C_t}$$
$$\left(or\ \frac{(1-\varepsilon)(q_i\dot{R}_i)}{C_t}\ for\ products\right) \quad (6)$$

where $(\rho C_p)_{eff}$ and λ_{eff} are the effective heat capacity and conductivity for porous media, respectively, ΔH is the heat of reaction, ϵ is the porosity of the specimen pellet, y is the mole fraction of gaseous species, D_{eff} is the effective diffusivity of gaseous reactants, and C_t is the total concentration of the gaseous mixture.

For the surface of the pellet, $r = R$ continuity of heat and mass flux were used as boundary conditions, At the center of the pellet, $r = 0$ spherical symmetry condition was used. At initial conditions, all variables will be equal to zero.

3. Application

The equations are discretized and solved by using the finite volume fully implicit approach [4]. The model has then been validated and is then applied to simulate the reduction of hematite within the moving packed bed by using syngas (a mixture of H_2, CO, CO_2, and H_2O) based on the following reactions:

$$Fe_2O_{3(s)} + 3H_2\ (\text{or CO})_{(g)} \rightarrow 2Fe_{(s)} + 3H_2O\ (\text{or } CO_2)_{(g)} \quad (7)$$

Fig. 2 shows a comparison between the experimental results for a scale model packed-bed furnace [1] and

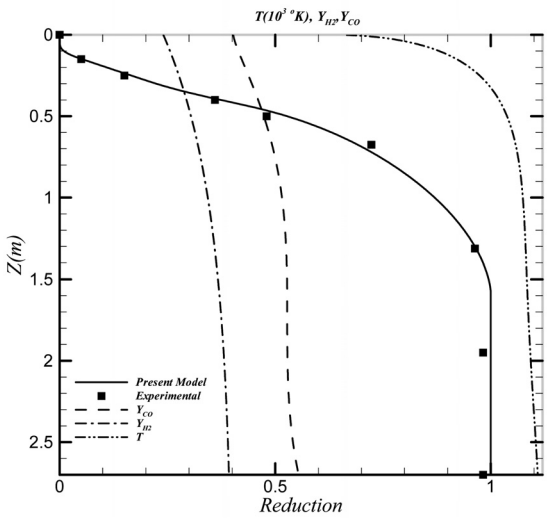

Fig. 2. Comparison of model estimation with experimental data reported by Takenaka et al. [1].

predicted conversion of hematite with syngas alongside the bed. It is observed that there is reasonable agreement between the measurement and the model results.

4. Conclusion

A mathematical model has been developed for investigating the reactional treatment of a packed bed made up of porous pellets immersed in a mixture of multicomponent gas. A special grain model is used to depict the heterogeneous reactions that take place in the solid interfaces. The implicit finite volume formulation has been used for solving the governing equation. The results of the model are found to be in agreement with those of the experiment.

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