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Numerical simulation on micromixing of viscous fluids in a stirred-tank reactor

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ABSTRACT

Micromixing of viscous systems in a stirred-tank reactor with Rushton turbine is investigated numerically. It is characterized by the product selectivity of parallel competitive reactions. Flow fields inside the reactor are determined by the Reynolds Stress Model (RSM). A computational fluid dynamics (CFD) method combining the standard E model and Finite-Rate/Eddy-Dissipation (FR/ED) model is implemented and is validated using experimental data in the literature. The simulations show that a higher agitation speed, a lower fluid viscosity and/or a feeding location closer to the discharge area of the impeller favor micromixing and the reaction rate. The trajectory of the reaction plume also influences the micromixing performance. The value of the FR/ED model parameter determined by lab-scale experiments decreases with increasing fluid viscosity. However, it is little affected by the agitation speed. These results provide useful guidelines for the scale-up of industrial reactors with complex chemical reactions.

1. Introduction

Mixing processes in industrial reactors with high-viscosity media may have a significant effect on the product yield and distribution, especially molecular weight distributions for polymers (Baldyga and Bourne, 1998; Bourne et al., 1989, 1995; Gholap et al., 1994; Verschuren et al., 2001). Macromixing describes the process for fluid elements to break up and advect into a space of interest. Micromixing takes place below Kolmogorov and Bachelor scales, promoting molecular diffusion and therefore influencing the product distribution.

The last decades have seen many studies on developing methods to characterize micromixing in aqueous and low-viscosity solutions. Consecutive reaction systems and competitive reaction ones are widely accepted (Akiti et al., 2005; Baldyga and Bourne, 1984; Bourne and Gholap, 1995; Bourne, 2003; Ottino et al., 1979; Vicum et al., 2004), because their product selectivity depends very much on micromixing (Brucato et al., 2000; Fournier et al., 1996). There are also studies on the effect of multiscale mixing by considering fine-scale mixing together with mesomixing (Akiti and Armenante, 2004; Baldyga et al., 1997).

Micromixing performance is a function of reactor geometry, chemical and physical characteristics of reacting system, operating conditions and feeding method of reagents. In the case of an unbaffled tank, feeding reactants at the upper trailing vortex of a Rushton turbine (Assirelli et al., 2002) with a rotating feed pipe (Assirelli et al., 2005, 2008) show good micromixing. In semi-batch tanks, the fluid viscosity affects the characteristics of the fine-scale, a scale near Kolmogorov microscale, and therefore micromixing (Bourne et al., 1995; Gholap et al., 1994; Guichardon et al., 1997). Atibeni (2005) and Cong et al. (2006) found that an increase in the fluid viscosity increased the selectivity of the side product. Kunowa et al. (2007) discussed the effect of micromixing in polymerization reactors and found that mixing performance decreased rapidly with increasing viscosity of reaction media. Yang et al. (2009) studied the micromixing performance of viscous media in a microreactor and found that the segregation index increased with increasing viscosity. A screw extruder is a special type of reactor for highly viscous systems for reactive extrusion processes (Cartier and Hu, 1998; Chen et al., 1995; Feng and Hu, 2004; Hu and Kadri, 1998; Hu et al., 1998; Hu and Cartier, 1999; Hu et al., 2003) in which micromixing may play a crucial role. Li et al. (2010) studied the micromixing performance of highly viscous polymer melts in a twin-screw extruder upon developing a new parallel competitive system consisting of two macromolecular reactions.

With the development of high performance computers, computational fluid dynamics (CFD) methods have been widely used as a simulation tool to investigate macromixing and turbulence of non-reactive fluid systems. Nevertheless, it is very challenging when it comes to a chemical reactor with interactions between micromixing and complex chemical reactions.

The commonly used micromixing models are multi-environment (ME), engulfment (Eng), engulfment–deformation–diffusion (EDD), interaction by exchange with the mean (IEM), and direct

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quadrature method of moments interaction by exchange with the mean (DQMOM-IEM). (Öncül et al., 2009) The formulation of the implemented micromixing models can be based on the probability density function (PDF) approaches. The full PDF models are considered to be more powerful to solve micromixing and complex chemical reactions than the presumed PDF ones. (Baldyga and Makowski, 2004; Kolhapure and Fox, 1999; Marchisio and Barresi, 2003; Tsai and Fox, 1996; Wang and Fox, 2004) Nevertheless, the computational difficulty is further aggravated.

The simplified EDD models, i.e., the standard or modified Engulfment model (E model) proposed by Baldyga and Bourne (1989a, 1989b) have been widely used for ideal reactors such as CSTR and plug flow reactors owing to their simplicity and computational performance (Baldyga and Bourne, 1990; Gholap et al., 1994; Kougoulos et al., 2006; Vicum et al., 2004). Akiti and Armenante (2004) incorporated the E models into CFD to track the reaction zone by means of the Volume of Fluid (VOF) model upon taking detailed hydrodynamics into account. They assumed that there was little diffusion between different species. Although the VOF could show a distinct reaction zone interface, it has to treat the homogeneous volumetric reactions as heterogeneous interfacial ones, which is an inherent drawback.

This work aims at studying the micromixing effect in viscous fluids in terms of the final product selectivity of a competitive parallel reaction system in a semi-batch stirred-tank reactor with a Rushton turbine. The standard E model and the Finite-Rate/Eddy-Dissipation (FR/ED) model are used to describe volumetric reactions. The effects of the agitation speed, feeding location and fluid viscosity on micromixing are simulated, revealing the different impacts of micromixing on different types of reactions and positions in the reactor. The numerical models are validated using experimental results in the literature (Atibeni, 2005).

2. Numerical models and simulation methods

A micromixing model (standard E model) coupled with the turbulence reaction model (FR/ED model) and local hydrodynamics is developed to investigate the micromixing effect of viscous fluids in a stirred-tank reactor.

2.1. Reaction system

The reaction system used is composed of parallel competitive reactions, of which one reaction is much faster than mixing while the characteristic time of the other is comparable with that of micromixing (Brucato et al., 2000):

$$\mathrm{NaOH(A)} + 0.5\mathrm{CuSO_4(B)} \xrightarrow{k_1} 0.5\mathrm{Cu(OH)_2(P)}\downarrow + 0.5\mathrm{Na_2SO_4(R)} \quad (1)$$

$$\mathrm{NaOH(A)} + \mathrm{CH_2ClOOC_2H_5(C)} \xrightarrow{k_2} \mathrm{CH_2ClOONa(Q)} + \mathrm{C_2H_5OH(S)} \quad (2)$$

where k_1 and k_2 are the reaction kinetic constants and are 10^7 and 0.023 $m^3/(mol.s)$, respectively. For convenience, Eq. (1) is called fast reaction and Eq. (2) slow reaction, although it is not slow actually. Q and S stand for the side products while P and R the primary ones.

The selectivity of side product Q, X_Q, as a quantitative index for describing the micromixing effect or performance, is sensitive to both fluid mixing and reaction kinetics. If micromixing is perfect, which refers to complete mixing at the molecular scale, X_Q is solely dictated by the kinetic constants of two reactions and is given by Eq. (3) and tends to zero as k_1 is very large compared to k_2.

$$X_Q = \frac{k_2 C_C}{k_1 C_B + k_2 C_C} \approx 0 \quad (3)$$

On the contrary, complete segregation refers to cases where no micromixing occurs. X_Q is then not dictated by the reaction kinetics but by the initial concentrations of reactants. X_Q is given by Eq. (4). The maximum value of X_Q is 0.667 because the initial concentration of C, C_{Co}, is twice that of B, C_{Bo}.

$$X_Q = \frac{C_Q}{C_P + C_Q} = \frac{C_{Co}}{C_{Co} + C_{Bo}} \approx 0.667 \quad (4)$$

In practice the value of X_Q is between these two limits and is a measure of the interaction between fluid mixing and reaction kinetics when the initial concentrations of reactants are fixed.

2.2. Simulation conditions

Simulations are carried out in a fully baffled (four baffles) stirred tank, whose inner diameter and liquid height are equal and are 0.476 m. The reactor is equipped with a six-bladed Rushton turbine whose diameter is 0.190 m and whose blade width is 0.038 m. The clearance between the impeller and the bottom is 0.159 m. Fig. 1 shows the details of the apparatus.

There are two feeding points. They are located at a vertical mid-plane between two adjacent baffles. One (L1) is 0.100 m below the liquid surface and the other (L2) close to the discharge area of the Rushton turbine and 0.159 m above the bottom of the

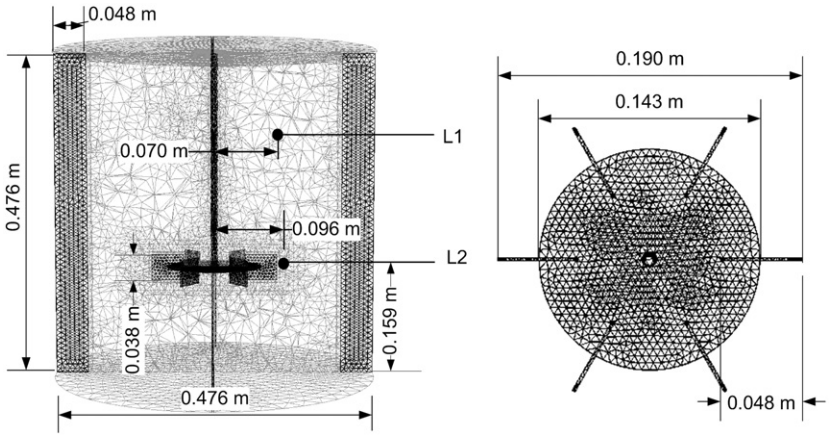

Fig. 1. Schematic presentation and computational grid of the stirred tank reactor with Rushton turbine (Left: Tank; Right: Impeller details).