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Modelling and experimental investigation of micromixing of single-feed semi-batch precipitation in a liquid–liquid stirred reactor

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HIGHLIGHTS

- Micromixing effect in liquid–liquid stirred tanks is quantified by experiment.
- Liquid–liquid flow is simulated with explicit algebraic stress model and k–ε model.
- Flow prediction is validated by experimental macromixing homogenization curves.
- Finite-mode probability density function model is adopted to model precipitation.
- Agreement between model predictions and the experimental data is satisfactory.

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ABSTRACT

A finite-mode probability density function (FM-PDF) model is adopted to simulate the semi-batch precipitation process of barium sulfate from aqueous solutions of barium chloride and sodium sulfate in the presence of kerosene drops in a turbulent liquid–liquid stirred reactor driven by a Rushton turbine. The turbulent kerosene–water flow field is calculated with an anisotropic two-phase explicit algebraic stress model (EASM) and the standard k–ε model based on an Eulerian–Eulerian approach using in-house codes. The flow predictions are validated against macromixing homogenization curves measured from tracer mixing experiments. The particle size distribution has been calculated by solving five moments of the particle size distribution coupled with both nucleation and growth kinetics. The effects of impeller speed and inert dispersed oil volume fraction have been investigated. Agreement between micromixing model predictions and experimental data is satisfactory, which suggests that the time-dependent finite-mode probability density function model is a feasible approach for numerical prediction of the fast precipitation process in turbulent two-phase stirred reactors.

1. Introduction

Mixing plays a central role in operating stirred immiscible liquid–liquid dispersions, which are extensively encountered in chemical and pharmaceutical industries. Turbulent mixing is a multiscale process, which is usually divided into three typical scales, namely, macromixing, mesomixing and micromixing. The fast or instantaneous chemical reactions are prone to be bottlenecked by the micromixing process.

Reactive precipitation involving mixing-limited instantaneous or fast reaction, particle nucleation and growth, is a key unit operation in production of many solid materials such as catalysts, fine chemicals and pharmaceutical products. The final quality of these products is enormously affected by particle morphology and particle size distribution (PSD), which is a result of competition between micromixing and rapid reactive precipitation. Numerous studies have been focused on the interactions of mixing and fast precipitation processes in single phase systems. However, the reactive precipitation in multiphase systems is actually encountered more frequently in industrial processes and demands more attention.

Computational fluid dynamics (CFD) has increasingly become a powerful tool for rational design and scale-up of such multiphase stirred reactors with complex flow and chemical reactions. Direct numerical simulation (DNS) and large eddy simulation (LES) are still very time-consuming and infeasible when it comes to industrial-scale applications. Furthermore, the agitated liquid–liquid flow simulations in stirred vessels by means of LES have not yet been quantitatively validated. Coroneo et al. [1] concluded that

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Nomenclature

b	impeller blade length, mm
c	tracer concentration, g/m^3
$\bar{c}$	normalized tracer concentration
c_c	solid concentration, kg/m^3
c_∞	homogeneous tracer concentration, g/m^3
C	off-bottom distance, mm
c_{A0}, c_{B0}	actual initial concentrations of A and B, mol/L
$\overline{c_{A0}}$, $\overline{c_{B0}}$	mean initial concentrations of A and B, mol/L
d	impeller disc diameter, mm
d_{32}	Sauter mean diameter, μm
d_{43}	volume-averaged particle size, μm
D	impeller diameter, mm
f_ϕ	joint composition PDF
G	particle grow rate, m/s
H	liquid height, mm
J_μ	constant
k	turbulent kinetic energy, m^2/s^2
k_d	mass transfer coefficient, (m/s)(m^3/kmol)
k_g	growth rate constant, m/s
k_v	particle volume shape factor
k_s	solubility product for barium sulphate
N	impeller agitation speed, rpm
N_e, N_s	total number of environments and scalars
m	moment of particle number density
M	molecular weight, kg/mol
p_n	probability of environment n
Q	constant
r	radial coordinate, mm
r_0	radial position of the center of the circulation loop
R	radius of the stirred tank, mm
R_N	nucleation rate, number/(m^3 s)
s_n	weighted mixture fraction of environment n
t	time, s
S	supersaturation
$\bar{S}_{\text{vol}}$	volume-averaged supersaturation
T	tank diameter, mm
u	velocity component, m/s
w	impeller blade width, mm
Y	reaction progress variable
z	vertical distance from tank bottom, mm

Greek letters

α	dispersed phase holdup
β_v	volume ratio
ε	turbulent kinetic energy dissipation rate, m^2/s^3
φ_R	composition of scalar R
μ	dynamic viscosity, Pa s
μ_t	turbulent dynamic viscosity, Pa s
ν	kinetic viscosity, m^2/s
γ	micromixing rate, 1/s
γ_s	spurious dissipation rate, 1/s
$\gamma\pm$	activity coefficient
χ	constant
ρ	density, kg/m^3
$\langle\xi\rangle$	local mean mixture fraction
$\langle\xi'\rangle^2$	mixture fraction variance
ξ_n	mixture fraction of environment n
ξ_s	stoichiometric mixture fraction
η	stoichiometric ratio of reactants
Γ_t	turbulent mass diffusivity coefficient, m^2/s
θ	azimuthal coordinate, ^{o}
ψ	composition vector

Subscripts

av	averaged
c	continuous phase
d	dispersed phase
eff	effective
j	radial, tangential or axial directions
r	radial direction
t	turbulent
z	axial direction
θ	tangential direction

Reynolds averaging of the convection–diffusion equation was an acceptable approximation after performing a systematic and stringent evaluation of the contribution of numerical issues to the accuracy of the most widespread Reynolds-averaged Navier–Stokes (RANS) simulation based on the k–ε model closure for stirred vessels. The computationally efficient RANS approach with appropriate turbulence models thus might be considered as the main tool in practical industrial applications so far.

In the RANS simulations, the macromixing process can be resolved in terms of convection and turbulent diffusion, whereas micromixing must be modelled additionally. The significance of micromixing model in numerical simulation of the fast precipitation process has been identified by many researchers [2,3]. Marchisio [2] investigated the role of micromixing model in RANS simulations of fast reactions in a Taylor–Couette reactor working in semi-batch conditions, and unveiled that the predicted total particle number density was overestimated by more than 200% and the simulated mean particle size was underestimated by about 50% if micromixing was neglected, suggesting the significant importance of micromixing models.

Various CFD-oriented micromixing models have been developed, and many of them have yielded fruitful results. Probability density function (PDF) method is a popular one due to the fact that the chemical reaction term can be treated exactly, whereas many of the other models require a closure assumption. PDF methods are usually classified as full and presumed ones. Tsai and Fox [4] used a joint-composition PDF model to study the effect of the scalar mixing rate on a series–parallel reaction in a single-jet tubular reactor. The PDF simulations were performed using a set of notional particles moving in the computational domain. Pipino and Fox [5] used a full PDF method to simulate the process of homogeneous nucleation of $BaSO_4$ in a tubular reactor, but they did not consider particle growth. Piton et al. [6] applied the 4 environment generalized micromixing (4-EGM) model to describe the turbulent mixing and precipitation of barium sulfate in a tubular reactor, and the 4-EGM model results were shown to compare favorably to the predictions by the presumed beta-PDF model. Marchisio et al. [7] and Marchisio and Barresi [8] used a finite-mode probability density function (FM-PDF) method to model the semi-batch precipitation of barium sulfate and competitive reaction systems in Taylor–Couette reactors. Moreover, Falk and Schaer [9] calculated the evolution of the particle size distribution based on the full PDF model coupled with the IEM model in a coaxial mixing jet reactor.

Comparisons were made between presumed (e.g., finite-mode, beta) and full PDF models for turbulent precipitation in a tubular reactor [10] and in an ideal, perfectly mixed batch reactor [11]. Wang and Fox [12] compared the multienvironment-presumed