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Prediction of drop sizes for liquid–liquid systems in stirred slim reactors—Part I: Single stage impellers

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ABSTRACT

Although investigations in the field of stirred liquid/liquid dispersions have a long history, new questions are still emerging in dealing with the different aspects of industrial applications, such as suspension polymerizations. In this study the influence of physical parameters on drop size and power consumption, like liquid level, stirrer speed, stirrer height and baffle length, were experimentally analyzed. The results were used to determine modeling approaches which are capable of displaying the influence of the named parameters. It was shown that the energy law ($d_p \sim \varepsilon^{-0.4}$; Shinnar, 1961 [1]) using the average energy dissipation only roughly predicts the Sauter mean diameter. The population balance equation (PBE) used with a one-zone modeling approach is slightly better in its prediction of results. Very satisfying predictions were obtained by using the PBE with a two-zone model. The overall deviations between calculated and predicted Sauter mean diameter was less than 10% using this approach. Only the successful prediction of the influence of the baffle length remained unattainable, even with the PBE two-zone model.

1. Introduction

Suspension polymerization is commonly used in the chemical industry for producing a wide variety of commercially important polymers, e.g. polystyrene, poly methylacrylate or poly vinyl chloride. In the initial phase of this polymerization the insoluble monomer is dispersed in an aqueous phase which contains a protective colloid or an inorganic suspension additive. This produces small liquid drops of monomer with a size range 1 μm to 3 mm. The drop size distribution (DSD) in this initial phase generally determines the final particle size distribution after the exothermic polymerization [2]. To control this distribution, a deeper understanding of the influencing parameters and an accurate prediction of the initial DSD is of major importance. Due to the economy of scale, reactor sizes are increasing. Only accurate models facilitate a precise scale-up for the reactor growth by increasing filling level.

In this study, an example production process of PVC is examined, with final mean diameters of solid particles ranging from 50 to 500 μm. PVC is one of the most important chemical products according to revenue. For example, the German PVC industry generates sales of 20 billion € per year. Eighty percent of PVC is produced by suspension polymerization worldwide. Growing markets and growing economies lead to higher PVC production rates. To achieve this more efficiently, newly built production reactors are increasing in height, while the diameter is fixed due to limits of space and issues of transportation. As a consequence, the ratio of liquid level height H vs. tank diameter T of such apparatus is enhanced; a ratio of 2.5 or higher is common and values of four are expected in the future. Predictive models for stirred vessels with a reactor height vs. diameter ratio of 1.0 are widely represented in the literature. The understanding of dispersion processes in slim reactors is incomplete and differs compared to the standard system and therefore extra difficulties are expected. Thus, the scale-up of a slim reactor from pilot plant to industrial scale remains a process where much empiricism, as well as expensive and time-consuming experimental programs, is usually required [3]. Only accurate predictions of system behavior will change this situation.

During the production process of PVC, stirring serves two purposes. Firstly, during the mixing of the organic phase of vinyl chloride with the aqueous phase, stirring facilitates effective dispersion. In the second step, stirring provides a homogeneous energy dissipation to control the agglomeration under polymerization and the cooling of the exothermic reaction.

In this work the task of dispersing two immiscible liquids was of major interest. In the experiments, the Sauter mean diameter ($d_{32} = \sum d_i^3 / \sum d_i^2$) and the DSD were analyzed. To predict these values the power numbers have been examined for various set-ups.

Abbreviations: CFD, computational fluid dynamics; DSD, drop size distribution; PBE, population balance equation; PVA, poly vinyl alcohol; PVC, poly vinyl chloride; RCI, retreat curve impeller.

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Nomenclature

Nomenclature

a, b, c, C_i	numerical or prediction constants
d	stirrer diameter, drop diameter [m]
d_{32}	Sauter mean diameter [m]
d_{max}	maximum drop diameter [m]
h	bottom clearance, stirrer height [m]
H	liquid level [m]
l_B	baffle length [m]
n	stirrer speed [rpm]
M	moment [Nm]
N	number of drops [#]
Ne	power number
P	power [W]
Re	Reynolds number
t	time [s]
T	tank diameter [m]
w'^2	fluctuation velocity [m/s]
We	Weber number

Greek letters

γ	interfacial tension [mN/m]
ε	P/V—energy dissipation rate [W/m^3]
ε	P/M—energy dissipation rate [m^3/s^2]
η	dynamic viscosity [kg/(ms)]
λ	Kolmogoroff scale [m]
ν	kinematic viscosity [m^2/s]
ρ	density [kg/m^3]
σ	standard deviation
φ	dispersed phase fraction

The DSD were then simulated using empirical equations and the Population Balance Equation (PBE). The purpose of this work is to systematically compare experimental DSDs and Sauter diameters with predicted values for different set-ups of slim reactors.

2. State of the art: drop size prediction

2.1. Empirical and half empirical equations

In literature a great number of equations for drop size prediction are available. Most of them are based on the descriptions of Shinnar [1]. He explains that drop breakage is caused by micro-turbulences and that the emerging drops are about the same size as the micro-turbulence eddies [1]. These micro-turbulences were first described by Kolmogoroff [4] and are characterized by the micro scale λ (Kolmogoroff-Scale) which depends on the kinematic viscosity ν and the average energy dissipation rate ε:

$$\lambda = \left(\frac{\nu^3}{\varepsilon}\right)^{1/4} \tag{1}$$

This equation is based on estimations of the Sauter mean diameter d_{32} [4]. A key figure is the Weber number We which puts deforming forces in the agitated system in relation to the stabilizing interfacial energy:

$$\mathrm{We} = \frac{\rho n^2 d^3}{\gamma} \tag{2}$$

When the Weber number reaches a critical value, drop breakage occurs. With the average fluctuation velocity w' and the continuous phase density ρ, the critical Weber number is reached when interfacial tension γ equals the external deforming forces $\rho w'^2 d_{max}$. Under the assumption of constant energy input in the whole vessel and homogenous isotropic turbulences, the size of these micro-turbulences d_{max} can be calculated depending on the stirrer diameter d and We:

$$\frac{d_{max}}{d} = C_1 \left(\frac{n^2 d^3 \rho}{\gamma}\right)^{-3/5} = C_1 \mathrm{We}^{-0.6} \tag{3}$$

Because of the linear dependency between d_{max} and d_{32} [5], the equation for the Sauter mean diameter is presented in literature as the following:

$$\frac{d_{32}}{d} = C_1 C_2 \mathrm{We}^{-0.6} \tag{4}$$

The value of the constant C_1 depends on vessel geometry and the stirrer type, and has to be evaluated experimentally. In the literature the values for C_2 are between 0.38 and 0.7 [6]. Depending on the system, C_2 can be altered according to the following factor:

$$C_2 = C_4(1 + C_3\varphi), \tag{5}$$

with C_4 as a constant for the type of stirrer and C_3 reflecting the coalescence characteristic of the medium. The summarized equation also includes the volume phase fraction φ:

$$\frac{d_{32}}{d} = C_4(1 + C_3\varphi)\mathrm{We}^{-0.6} \tag{6}$$

Literature shows values for C_3 from 3 to 20 [7]. This equation assumes a linear correlation between φ and d_{32} that could not be confirmed by the experimental studies of Kraume et al. [8] and Angle et al. [9,10].

Furthermore, the validity of C_3 is questioned because of its interconnection with the drop size.

Eq. (3) with $\mathrm{We}^{-0.6}$ is also commonly expressed as a function of the specific power input with $\varepsilon^{-0.4}$. The Weber number is not affected by changes in the filling level and thus cannot effectively predict changes in the corresponding power input. Variations are introduced into the constant C_1 and need to be known. Therefore, the discussions about drop sizes in this study concerning the energy dissipation rate are always based on $C_1\varepsilon^{-0.4}$ and not on $C_1\mathrm{We}^{-0.6}$.

A broad and detailed overview of existing empirical and half empirical equations for the prediction of the Sauter mean diameter is given by Zerfa and Brooks [6] as well as by Angle and Hamza [10].

2.2. Population balance equations (PBE)

All presented drop size prediction equations assume an ideally mixed tank meaning the dependency on space is neglected. Such a simplification is too restrictive as the flow field, which highly influences drop breakage, coalescence, and the therefore the drop sizes, is usually very inhomogeneous throughout the stirred tank [11,12]. However, a detailed simulation using Computational Fluid Dynamics (CFD), which includes the coupled phenomena of the interactions of turbulent eddies in the flow field and the drops, requires great computational power and thus is very time consuming. A reliable compromise between detailed modeling and savings in computational time is offered by the compartment model approach. Various studies have shown promising improvements using a compartment model when compared to calculations using an average energy dissipation rate [13–16].

In this work the vessel is separated into two well-mixed regimes; one standing for the impeller region and the other for the remaining reactor volume. The size of these flow compartments is based on CFD simulations which were carried out using the software STAR-CCM+®. Furthermore, the unknown parameters such as the ratio of compartment volumes, energy dissipation rates and their exchange flow rates are computed. Those parameters are then implemented into a two-compartment model which calculates the drop size distributions for each regime based on the population