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Liquid–liquid dispersion in co-current disc and doughnut pulsed column effect of the operating conditions, physical properties and materials parameters

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HIGHLIGHTS

- Co-current disc and doughnut pulsed column.
- High volume dispersed phase fraction.
- Effect of hydrodynamic and materials.
- Modelling through dimensionless number.

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ABSTRACT

Pulsed columns are traditionally used to perform liquid–liquid extractions at counter-current. They are currently seen as a potential tubular reactor to perform co-current continuous processes. In this paper the case of liquid–liquid dispersion is considered. Various parameters are investigated such as the dispersed phase holdup, the flowrate and pulsation conditions but also some process parameters such as the packing material or the physical properties such as the type of surfactant. The energy dissipation rate is estimated according to correlations found in literature. Breakage and coalescence rate models are applied in order to understand the various observed phenomena. Finally, a correlation to predict the mean droplet size is proposed depending on various dimensionless numbers characterizing the flow, the internal and the physico-chemistry of the phase systems.

1. Introduction

Among the available continuous technologies, the pulsed column has already been largely studied in the past, mainly for counter-current liquid–liquid extraction processes [1]. It consists in a column packed with discs and doughnuts equally spaced or perforated sieved plates.

Today, this kind of tubular plug flow reactors could be used for continuous crystallization, for polymerization [2,3] or for biodiesel production [4] but with a co-current flow.

In this paper, we focus on the liquid–liquid dispersion mechanism which supports various processes, most of the time batch-wise. A lot of processes require a control of the initial mean droplet size and droplet size distribution to provide the correct properties of the final product.

Moreover, in order to avoid the solvent removal and to increase productivity, it is interesting to produce liquid–liquid dispersion at a high dispersed phase fraction. Indeed, it is expected to decrease the cost, but also to facilitate the separation steps in order to obtain the final product and to reduce the effluent post-treatment operations.

Nowadays, the industry is interested in developing continuous processes to replace the classical batch reactors. In stirred tanks the emulsification duration is generally the time needed to reach stabilized mean droplets sizes. This characteristic duration ranges from few minutes to hours [5–9].

Continuous processes can significantly reduce this duration which corresponds to the residence time. Besides, continuous processes offer a higher level of control on the different process parameters such as droplet size and exhibit plug flow behavior. They also enhance temperature management compared to batch process. Calabrese and Pissavini [10] present the benefits of continuous processes in terms of cost, safety and quality.

The study of various designs in literature confirms the ability of pulsed flows to perform a good mixing [11,12] and transport of liquid–liquid dispersion and liquid–solid suspension [13]. The continuous oscillatory baffled reactor (COBR, Nitech®), a special

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design composed of rings equally spaced has been extensively studied. Heat transfer [14], mixing performance [15,16], and liquid–liquid dispersion [17–19] have been particularly detailed but often in batch process.

In this paper, our goal is to study the performance of a co-current pulsed upflow disc and doughnut column performing a liquid–liquid dispersion with a controlled mean diameter. Two model systems are investigated: Water/PVA/Toluene and Water/SDS/Toluene. PVA is partially hydrolyzed polyvinyl acetate and SDS is sodium dodecyl sulphate. The aim of this study is to analyze the effect of the operating conditions (pulsation and flowrate), design parameters (material) and physical properties (surfactant and dispersed phase holdup) on mean droplet size and on the droplet size distribution. Identification of the relevant parameters affecting the liquid–liquid dispersion characteristics (droplets size distributions, mean droplets size, etc.) leads to propose a correlation to predict the Sauter mean drop diameter.

2. Materials and methods

2.1. Fluids used

In this study two water/oil/surfactant systems are investigated varying only by the surfactant type (anionic or non ionic). Toluene (95%) was purchased from Gaches Chimie. PVA is partially hydrolyzed polyvinyl acetate and is a non ionic surfactant. It was provided by Nippon Gohsei. Sodium dodecyl sulphate (SDS) was provided by Panreac and is an anionic surfactant. The liquid–liquid dispersion created consists in oil in water (O/W) dispersion.

The properties of the different systems are summarized in Table 1. The interfacial tension is measured by the pendant drop method (Krüss DSA 100). Viscosity measurements are carried out using an AR 2000 rheometer (TA Instruments).

The aqueous phase is composed of distilled water and surfactant and the dispersed phase is only composed of toluene.

2.2. Experimental rig

The experimental set up consists of a 3 m long column of 50 mm internal diameter D_c packed with immobile discs and doughnuts equally alternated and spaced ($H = 24$ mm) made of stainless steel (316 L) or PTFE. The inner diameter of the ring aperture is D_a and the disc diameter is D_d. Both diameters are fitted in order to obtain the same transparency factor T (ratio between inserts area to cross section) for the disc or the doughnuts. The characteristics of the different inserts (disc and doughnut) are given in Fig. 1. Whatever the insert's materials, the open free area T (transparency factor) is of 26%. They are mounted horizontally and centered with respect to the column axis. One level represents either two doughnuts (or rings) separated by a disc or two discs separated by a doughnut (Fig. 1). The outer edge of the rings is extended to the column wall. The flow follows two routes: a central one through the ring aperture and a peripheral one between the disc edge and the column wall.

Table 1
Physicochemical properties of the two systems.

	Water/PVA/toluene	Water/SDS/toluene
ρ_c (kg m^{-3})	997	998
ρ_d (kg m^{-3})	870	870
μ_c (Pa s)	0.0059	0.0059
μ_d (Pa s)	0.001	0.001
Surfactant concentration	0.07% mass/kg toluene	2.3% mass/kg toluene
σ_e (mN m^{-1})	3.5	3.5

The aqueous phase is prepared in a 220 L tank in which the water and the surfactant are mixed. The toluene is stored in a 200 L tank. Each tank is under constant weighting to check the flowrates. The two flows are pumped through membrane pumps to the bottom of the column. The feed lines are equipped with anti-pulsatory balloons which absorb the pulsation due to the volumetric pumps and then ensure a constant flowrate. The pulsation is imposed via a pump without check valves allowing the control of the oscillation amplitude, A, via the pump vernier and of the oscillation frequency *f*, thanks to the variable frequency regulator. This device induces reciprocal up and down movements of the fluids inside the column. Different samples are collected all along the column: at the basis (generation of the dispersion at the first doughnuts) and after every stage to measure the droplet size distribution.

The amplitude A describes the total displacement of the flow. In our case, the minimum amplitude is equal to the length between a disc and a doughnut $H_{\min}$ and the maximum amplitude to the difference between two discs or two doughnuts, $H_{\max}$.

The dispersed phase holdup in volume Φ (hold-up) is defined thanks to the respective phases flowrates as follows:

$$\Phi = \frac{Q_d}{Q_d + Q_c} = \frac{Q_d}{Q_{tot}} \qquad (1)$$

where Q_d, Q_c and Q_{tot} are respectively the dispersed phase, continuous phase and total volume flowrates.

It implicitly suggests that there is no slip velocity.

2.3. Analytical aspects

2.3.1. Interfacial tension measurement

Due to the presence of surfactants in the aqueous phase, the interfacial tension between the toluene and the aqueous phase evolves dynamically until it reaches its equilibrium value. In a continuous process, the interfacial tension evolves all along the equipment due to the constant breakup-coalescence processes and the rearrangement of the surfactant molecules at the interface. In both studied systems, the surfactant concentration in the aqueous phase is above the critical micellar concentration (concentration mentioned in Table 1).

The transient interfacial tension is measured for each system via the pendant drop method (Krüss DSA100). It consists in generating a drop at a tip of a needle. A CCD camera allows the recording of the drop shape. Then, thanks to the droplet image analysis software, the interfacial tension is calculated via the Young–Laplace equation. The method is well-known and has been largely commented in different publications [20].

2.3.2. Contact angle measurement

At the inserts surface, three phases are in contact. As previously described, a first interface composed by the two liquid phases is created: the dispersed phase and the continuous phase containing the surfactant. The internal represents the third phase (solid). Depending on the internal type (made of stainless steel or PTFE), it is expected that the liquid phases interact differently resulting in different behaviors of the phase systems.

To analyze the interaction, the contact angle of a single drop is measured via the Krüss DSA100 by using the captive bubble method.

A piece of the internal is maintained in the aqueous phase and thanks to a curved needle, a toluene droplet is posed on the surface. The complementary part of the contact angle is then measured. The inserts used for measurement are part of the column inserts. Table 2 reports the values of the different contact angles.

As it was expected, stainless steel surface is more hydrophilic than the PTFE internal. It seems that the PTFE is not preferentially