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# Population balance of droplets in a pulsed disc and doughnut column with wettable internals

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#### HIGHLIGHTS

• Three new source terms considering wettability of PDDC internals were introduced into PBE.

• Functions in the new source term of the PBE were experimentally measured.

• Correlations were proposed and applied in a simplified PBM calculation.

• Experimental results provided base data for the study of droplet population balance.

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### $A \hspace{0.1in} B \hspace{0.1in} S \hspace{0.1in} T \hspace{0.1in} R \hspace{0.1in} A \hspace{0.1in} C \hspace{0.1in} T$

Population balance of droplets was studied in a square-sectioned pulsed disc and doughnut column with wettable internals. It was found that a liquid film of the dispersed phase was formed on the internals. To describe the interaction between the droplets and the liquid film, three more terms need to be added into the source term of the classic population balance equation (PBE). To determine the source term of the PBE, functions including the droplet-layer coalescence frequency, residual droplet size distribution, droplet dispersion frequency from the liquid layer, droplet breakup frequency, and daughter droplet size distribution were measured experimentally. The droplet-layer coalescence frequency increases as the pulsation intensity decreases because of the longer contacting time. The residual droplet size is around half of the original coalescing droplet and affected little by the pulsation. The droplet dispersion frequency gets higher with the increasing of the pulsation intensity or the thickness of the liquid layer. The droplet breakup frequency is positively related to the energy input and droplet size. The daughter droplet size distribution changes from inverted U-shaped to M-shaped as the pulsation intensity rises. Based on the experimental results, empirical correlations were proposed to calculate these functions. Furthermore, the correlation equations were applied in a simplified population balance model to calculate the droplet number density in the column. The predicted results agree well with the experimental data, which proved the feasibility of the correlations.

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## 1. Introduction

Droplet breakup and coalescence are common phenomena in the nature and the industrial production, such as formation of rain drops in clouds, emulsion preparation, micro-fluidics, polymer synthesis, pharmaceutical industries, and solvent extraction (Luo and Svendsen, 1996; Zhao et al., 2006; Gross et al., 2013). Especially in solvent extraction, droplet breakup and coalescence is important in the enhancement of mass transfer performance by increasing and refreshing contact area of liquid-liquid two-phase flow. To pro-

\* Corresponding author. E-mail address: lsw@tsinghua.edu.cn (S. Li). mote droplet breakup, researchers have developed many types of extraction equipment with different ways of energy input, such as pulsation (Billerbeck et al., 1956), stirring (Srilatha et al., 2010), oscillation (Noh and Baird, 2004) and rotation (Wardle et al., 2008). Pulsed disc and doughnut column (PDDC) is one of these types, which enhances droplet breakup by pulsation.

PDDC is a relatively newly developed extraction column which has the advantages of stronger dirt unloading capacity and weaker channeling effect comparing to the traditional pulsed sieve column. Thus, it has been applied extensively in reprocessing of the spent nuclear fuel in France and Japan (Angelov et al., 1990; Iseki et al., 2007; Nitta and Matsuda, 2005). Though researchers have done lots of work on PDDC (Torab-Mostaedi et al., 2011a,b,c,d;







#### Nomenclature

Α	ampl	itude of	the pu	lsation,	mm
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Af	pulsation intensity, mm s <sup>-1</sup>

$B^{\mathrm{B}}(D,t)$	birth rate of droplets due to droplet breakup, $m^{-4} s^{-1}$
$B^{C}(D,t)$	birth rate of droplets due to droplet-droplet coales-
	cence, $m^{-4} s^{-1}$

- $\bar{B}^{C}(D, t)$  birth rate of residual droplets due to droplet-layer coalescence, m<sup>-4</sup> s<sup>-1</sup>
- $B^{D}(D,t)$  birth rate of droplets due to droplet dispersion, m<sup>-4</sup> s<sup>-1</sup> D diameter of the droplet, mm
- $D^{\rm B}(D,t)$  death rate of droplets due to droplet breakup, m<sup>-4</sup> s<sup>-1</sup>
- $D^{C}(D, t)$  death rate of droplets due to droplet-droplet coalescence, m<sup>-4</sup> s<sup>-1</sup>
- $\bar{D}^{C}(D,t)$  death rate of droplets due to droplet-layer coalescence,  $m^{-4} s^{-1}$
- $F_{\beta}(f_V)$  daughter droplet size distribution function, –
- ffrequency of the pulsation, Hz $f_V$ volume ratio of the daughter droplet to the mother droplet, –(D)P(D)
- $\begin{array}{ll} G(D) & \text{droplet-layer coalescence frequency function, m s}^{-1} \\ H(D,D') & \text{residual droplet size distribution function, m}^{-1} \\ h(\alpha) & \text{transformed residual droplet size distribution} \\ & \text{function.} \end{array}$

- *l* length of the plate edge, m
- $n(\overline{x}; D, t)$  number density function of the droplets, m<sup>-4</sup>
- Q(D,D') droplet-droplet coalescence frequency function, m<sup>3</sup> s<sup>-1</sup>
- S area, m<sup>2</sup>

t time, ms

V volume, m<sup>3</sup>

#### Greek letters

- $\alpha$  diameter ratio of the residual droplet to the coalesced droplet, –
- $\beta(D,D')$  daughter droplet size distribution function, m<sup>-1</sup>
- $\Gamma$  (D) breakup frequency function, s<sup>-1</sup>
- $\Lambda$  (*D*,  $\delta$ ) droplet dispersion frequency function, m<sup>-2</sup> s<sup>-1</sup>
- $\delta$  thickness of the liquid layer, mm

#### Abbreviations

PBE	population balance equation
PBM	population balance model
PDDC	pulsed disc and doughnut column

Kumar et al., 2013), it is still an ongoing research issue to predict the droplet size distribution in the column, which is very important for determining interfacial areas and mass-transfer rates. The most used method to predict the droplet size is the population balance model (PBM), which considers the effect of both droplet breakup and coalescence.

One of the key problems of PBM is to determine the breakup and coalescence frequency, which has been researched for more than fifty years. The models most often used for droplet breakup and coalescence frequency are proposed by Coulaloglou and Tavlarides (1977) and Luo and Svendsen (1996). These models are still used in recent works to simulate the two-phase flow in extraction columns (Drumm et al., 2009, 2010). The PBM calculation in these reports and most other relevant literature involves four source terms describing the effect of droplet breakup and coalescence. Though the four-source-term PBM is feasible for most two-phase dispersion system, it is not valid to calculate the flow in PDDC when the internals are wettable to the dispersed phase. For example, the co-decontamination columns of the PUREX process in the commercial nuclear fuel reprocessing plants in France and Japan are such type of devices. The stainless steel internals are wettable to the dispersed aqueous phase in these columns, thus an aqueous liquid layer is formed on the discs and doughnuts. The droplets colliding with the liquid layer may coalesce into the layer while new droplets are torn off from the liquid layer at the edge of the internals due to the shear effect of the continuous phase. To the best of our knowledge, these two factors have never been included in the PBM calculation in the literature. New terms must be added to the source of the PBM equation to describe the droplet-layer coalescence and dispersion. To determine these new terms by measuring the droplet coalescence and dispersion frequency is of great significant in the study of liquid-liquid two-phase flow in the PDDC.

In this work, an improved PBM was firstly established to describe the liquid-liquid two-phase flow in the PDDC. A highspeed online camera was then used to investigate the droplet behavior in a square-sectioned PDDC in order to experimentally determine the source term in the PBM. The frequencies of the droplet coalescence into and dispersion off the liquid layer were counted from the captured video. Meanwhile, the breakup frequency and daughter size distribution of free droplets were also measured from the video, similar to our previously reported work (Liu et al., 2016). The effects of the pulsation and two phase flowing conditions were analyzed. Correlations were then established to describe the measured results. The correlations were used in a simplified model to perform a PBM calculation. The calculated droplet number density agrees well with the experimental results. The experimental results provided important base data for the population balance study of droplets.

#### 2. Theory

When dealing with the hydrodynamic problem of liquid-liquid two-phase flow, a PBM defines how the number density of droplets with specific size develops over time. The population balance equation (PBE) in this case is a conservation form of the number density function, as expressed by Eq. (1).

$$\frac{\partial n(D; \vec{x}, t)}{\partial t} + \vec{u} \cdot \nabla n(D; \vec{x}, t) - \nabla \cdot (\Gamma_t \nabla n(D; \vec{x}, t)) = S(D, t)$$
(1)

where  $n(D; \vec{x}, t)$  is the number density function of the droplets and thus  $n(D; \vec{x}, t)dD$  is the number of droplets with size range from *D* to D + dD in unit volume.  $\Gamma_t$  is the effective diffusion coefficient of the number density, which is mainly caused by the turbulence of the flow. The source term S(D, t) includes four parts as shown in Eq. (2).

$$S(D,t) = B^{C}(D,t) - D^{C}(D,t) + B^{B}(D,t) - D^{B}(D,t)$$
(2)

where  $B^{C}(D, t)$  and  $D^{C}(D, t)$  represent the rate of birth and death of droplets of size *D* respectively due to coalescence.  $B^{B}(D, t)$  and  $D^{B}(D, t)$  represent the rate of birth and death of droplets of diameter *D* respectively due to breakup. Here the four terms of S(D, t) can be written as follows when only binary breakup and coalescence are considered.

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