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Author/s:

Grabin, T;Smith, KH;Mumford, KA;Wang, Y;Stevens, GW

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# **Effect of Plate Wettability on Dispersed Phase Holdup in a Pulsed Disc and Doughnut Solvent Extraction Column**

Teobaldo Grabin, Kathryn H. Smith, Kathryn A. Mumford, Yong Wang, Geoffrey W. Stevens\*

Particulate Fluids Processing Centre, Department of Chemical Engineering, The University of Melbourne, Parkville, Victoria 3010, Australia.

\*Corresponding author:

Name: Professor Geoffrey Stevens

E-mail: [gstevens@unimelb.edu.au](mailto:gstevens@unimelb.edu.au)

Postal Address: Department of Chemical Engineering

The University of Melbourne

VIC 3010 Australia

Tel.: +61 3 8344 6621

Fax: +61 3 8344 8824

## **Abstract**

The effect of plate wettability on the dispersed phase holdup in a pulsed disc and doughnut solvent extraction column is presented. Teflon, Nylon and Stainless Steel plates have been used to simulate a change in the wetting characteristics of the plate material that can occur in an operating column due to ageing or deposits accumulating on the plate. Experimental holdup data has been measured over a range of operating conditions using a 1.0 m long glass column with an internal diameter of 72.5 mm containing alternating discs and doughnuts. The liquid-liquid system studied was tri-n-octylamine (TOA)-kerosene-water with sulphuric acid as the solute. Results show that there are noticeable changes in the characteristic velocity (determined from measured holdup) and operational regimes for the different plate materials, particularly at low pulsation intensities, when operating under dispersed aqueous conditions. Experimental holdup data from this study has also been compared to correlations from literature for predicting holdup. As none of these correlations for holdup incorporate plate wettability, a new correlation for predicting holdup has been proposed that incorporates the contact angle of the plate material to allow for changes in the wettability of the plate surface.

**Keywords:** solvent extraction, pulsed disc and doughnut column, plate wettability, contact angle, characteristic velocity, holdup

## **1. Introduction**

Solvent extraction is used as a separation and purification process in a large number of chemical and metallurgical industries.<sup>(1)</sup> Several types of contactors have been developed with different agitation systems and different internal geometries such as mixer settlers, columns and centrifugal extractors. In each of these equipment options, the aqueous and organic phases are mixed to allow effective mass transfer of the solute before separation of the aqueous and organic phases. In order to promote contact between both phases van Dijk<sup>(2)</sup> introduced mechanical agitation to columns, these became known as Karr reciprocating plate columns and pulsed perforated plate columns (PPPC). Each of these columns contained perforated plates in order to increase dispersion. In the Karr column the plates were reciprocated up and down and in the PPPC the liquid was pulsed through the stationary plates.

The pulsed disc and doughnut column (PDDC) was developed more recently and can be seen as a variation of the PPPC as it contains a series of alternating stationary discs and doughnuts through which the liquids are pulsed. The discs are circular and smaller than the column diameter to provide an annular open area while the doughnuts have an external diameter ideally identical to the column diameter and an internal diameter whose area is equivalent to the disc's annular open area. Both are placed axially within the column. Aqueous and organic phases can be continuous or dispersed depending on where the interface is maintained and the phases are pulsed sinusoidally via a piston type unit or compressed air. The PDDC has been used in the French nuclear industry and metallurgical industries<sup>(3)</sup> and has gained attention due to its simple design, compact nature and lack of internal moving parts leading to safety and economic benefits.

When designing columns such as the PDDC which contains internals such as plates, the internals are commonly chosen such that they are preferentially wetted by the continuous phase. Little attention has been given to how this choice of material affects the overall performance of the column. Additionally the wetting characteristics of the plate material may change over time due to aging of the material, adsorption of processing materials or corrosion, leading to a change in the hydrophobicity or hydrophilicity of the plate material and subsequent change in column performance.

When operating a solvent extraction column such as the PDDC, the dispersed phase will undergo a series of breakage and coalescence events until equilibrium is reached. This equilibrium is maintained along the column, generating a droplet size distribution assumed to be constant. Such distribution is an important parameter since it will determine the interfacial area for mass transfer. The volumetric fraction of dispersed phase in the active section of the column is called the dispersed phase holdup and is another important parameter for understanding the performance of a solvent extraction column since it relates to the residence time, droplet size distribution and interfacial area for mass transfer.

This study aims to investigate how plates with different wetting properties affect the dispersed phase holdup performance of a PDDC. The wetting behaviour of the plates is changed by using different plate materials, namely Teflon, Nylon and Stainless Steel (SS). Teflon can be considered wetting (when organic dispersed) or non-wetting (when aqueous dispersed) while Stainless Steel follows the opposite trends. Nylon exhibits intermediate wetting properties under both conditions. Hydrodynamic performance is measured via holdup and the characteristic velocity is determined by linear regression. The effect of plate wettability on the characteristic velocity is assessed over a range of conditions. The models for predicting dispersed phase holdup from literature are compared to experimental data and a new model is presented that considers the plate wetting characteristics in terms of the contact angle measured in the liquid-liquid-solid system.

## 2. Background

Numerous studies have reported on the performance of a range of solvent extraction column geometries for a wide range of systems but little attention has been given to plate wettability and most importantly how it affects the overall performance of the column. Several authors<sup>(4-7)</sup> have used correlations developed for the PPC or pulsed sieve plate column (PSPC) to refit the correlation for a PDDC but these do not consider the effects of plate wettability in a PDDC.

### 2.1. Holdup correlations

Jeong and Kim<sup>(8)</sup> developed a correlation for predicting holdup in a disc and doughnut column using kerosene and water system in the absence of mass transfer:

$$x_d = 341.14h_c^{-0.44}Af^{1.28}V_d^{0.93} \quad (1)$$

Where  $h_c$  is the compartment height,  $A$  peak to peak amplitude,  $f$  frequency and  $V_d$  the dispersed phase velocity.

Hartland and Kumar<sup>(9)</sup> developed a correlation for a PPC based on a total of 725 data points, with organic as the dispersed phase, in three different regimes: mixer-settler, dispersion and emulsion. The boundary between dispersion and emulsion was defined by the rate of energy dissipation per unit mass of the mixed phases. Later the same authors<sup>(10)</sup> developed a correlation

for a PPPC based on 1574 data points in the absence of mass transfer as shown:

$$x_d = k_1 \exp[k_2 |Af - (Af)_m|] V_d^{0.86} (V_c + V_d)^{0.28} \Delta\rho^{-0.30} \rho_d^{-0.93} \mu_d^{0.77} \varepsilon^{-0.56} h_c^{-0.56} \quad (2)$$

Where  $k_1$  and  $k_2$  are constants ( $1.99 \times 10^6$  and 44.31, respectively),  $V_c$  is the continuous phase velocity,  $\Delta\rho$  is the density difference between phases,  $\rho_d$ ,  $\mu_d$  are the density and viscosity of dispersed phase respectively,  $\gamma$  is the interfacial tension and  $\varepsilon$  is the fractional free area:

$$(Af)_m = 9.69 \times 10^{-3} \left( \frac{\gamma \Delta\rho^{1/4} \varepsilon}{\mu_d^{3/4}} \right)^{0.33} \quad (3)$$

Kumar and Hartland<sup>(11)</sup>, later used a large bank of published data including eight different types of solvent extraction columns (which did not include the PDDC) to develop a unified correlation for holdup in terms of mechanical power dissipation ( $\Pi$ ), flow rates ( $\Phi$ ), physical properties ( $\Psi$ ) and column geometry ( $\Gamma$ ) as follows:

$$x_d = \Pi\Phi\Psi\Gamma \quad (4)$$

where:

$$\Pi = C_\Pi + \left[ \frac{\Phi}{g} \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right]^{n_1} \quad (5)$$

$$\Phi = \left[ V_d \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right]^{n_2} \exp \left[ n_3 V_c \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right] \quad (6)$$

$$\Psi = C_\Psi \left( \frac{\Delta\rho}{\rho_c} \right)^{n_4} \left( \frac{\mu_d}{\mu_w} \right)^{n_5} \quad (7)$$

$$\Gamma = C_\Gamma e^{n_6} \left[ h_c \left( \frac{\rho_c g}{\gamma} \right)^{1/2} \right]^{n_7} \quad (8)$$

Further work was completed by Kumar and Hartland<sup>(11)</sup> to develop a general correlation for predicting holdup in eight different liquid-liquid extraction columns that was independent of column geometry. The correlation was based on data from 7,198 measurements however it did not include data from a PDDC:

$$x_d = \left[ 0.27 + \left\{ \frac{\varphi}{g} \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right\}^{0.78} \right] \left[ V_d \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right]^{0.87} \exp \left[ 3.34 V_c \left( \frac{\rho_c}{g\gamma} \right)^{1/4} \right] \times \left( \frac{\Delta\rho}{\rho_c} \right)^{-0.58} \left( \frac{\mu_d}{\mu_w} \right)^{0.18} C_{\Gamma} e^{n_6} \left[ h_c \left( \frac{\rho_c g}{\gamma} \right)^{1/2} \right]^{-0.39} \quad (9)$$

This equation reproduced the entire set of experimental data reported in the study with an average absolute value of the relative error of 29.5%. Comparing this with the original correlation specific to each type of the column using equation (4), it was noted that the geometries of each type of column are very important when predicting holdup. The droplet motion and continuous-phase flow patterns are significantly influenced by the type of agitation and geometrical characteristics and therefore the use of a generalized correlation for predicting holdup in a PDDC is not recommended.

Van Delden et al.<sup>(12)</sup> and Ranjish et al.<sup>(4)</sup> have recently applied equation (4) to predict holdup data collected from a PDDC. A range of constants had to be refitted in both studies in order to reasonably predict holdup in the PDDC. The most extensive study on holdup in a PDDC has been reported by Jahya<sup>(6)</sup> who made use of an existing correlation for a PPPC and refitted it to experimental data from the PDDC. Most recently Torab-Mostaedi et al.<sup>(7)</sup> reported holdup data using a PDDC with three different systems – kerosene-water, toluene-water, and n-butyl acetate-water – with SS plates in the absence of mass transfer. For transition and emulsion regimes the correlation for holdup was refitted to the equation presented by Hartland and Kumar<sup>(9)</sup> as follows:

$$x_d = 3.73 \times 10^{-3} \left( \frac{(Af)^4 \rho_c}{g\gamma} \right)^{0.62} \left( \frac{V_d^4 \rho_c}{g\gamma} \right)^{0.31} \left( 1 + \frac{V_c}{V_d} \right)^{0.45} \left( \frac{\Delta\rho}{\rho_c} \right)^{-2.20} \left( \frac{\mu_d^4 g}{\rho_c \gamma^3} \right)^{-0.29} \quad (10)$$

## 2.2. Effect of plate material

Garner et al.<sup>(13)</sup> were one of the first to report on the effect of plate material on PPPC performance. In summary they recommend that hydrophobic materials be used for dispersed aqueous and organic continuous systems. Ikeda and Suzuki<sup>(14)</sup> analyzed the wetting effects on the hydrodynamics in a PPPC. They concluded that SS nozzle plates were best suited for organic continuous, despite their experiments being carried out in the absence of mass transfer, and silica

for aqueous continuous. They noticed that when the liquid on the plate changes from the continuous phase to the dispersed phase, the dispersion of the droplets became insufficient in the column, which resulted in the dispersed phase coalescing on the plate surfaces. They called this a “wettability change” and they correlated the required pulse velocity to avoid the wetting change as the maximum pulse power of a single pulse (left side of equation (11)) with the work of adhesion (right side of (11)):

$$0.25\rho_c d^2 A f^2 \pi^3 \geq \gamma(1 + \cos \theta) \quad (11)$$

where  $\rho_c$  is the continuous phase density,  $d$  is the column diameter,  $A$  is the peak-to-peak amplitude,  $f$  is the frequency,  $\gamma$  is the interfacial tension and  $\theta$  is the contact angle measured using a “preferential wetting method” proposed by the authors. The dispersed phase holdup was also correlated with the work of adhesion,  $W$ , derived from an air-liquid-solid system, presented by Neumann<sup>(15)</sup>. They used a system of 30% TBP in kerosene and water and three different plate materials – SS, Alumina and Silica to develop the following correlation for holdup in a PPC:

$$x_d = 26.8 \left[ \left( \frac{(Af)^3 \mu_d}{\beta W V_d^2} \right)^{1/3} \left( \frac{V_d^2}{\gamma \Delta \rho h_c} \right)^{1/4} \right]^{0.21} \quad (12)$$

where  $\beta = \frac{\varepsilon^2}{(1-\varepsilon)(1-\varepsilon^2)}$  for  $Af \leq 0.1$  and  $W = \gamma(1 + \cos \theta)$

Mate et al.<sup>(16)</sup> studied the wetting effects in transient state (start up) with organic dispersed phase in a PDDC. The time delay was measured until the dispersed phase outlet equaled the inlet and compared the results of SS and Teflon plates. Similarly to Ikeda and Suzuki<sup>(14)</sup>, the Teflon formed a film (thereby changing the wettability) and by subtracting the outlet flow rates of dispersed phase at identical conditions, they were able to quantify the volume of film retained within the column.

Arunachalam<sup>(17)</sup> studied the effect of plate material on the performance of a Karr reciprocating plate column and found equation (4) to be the most promising model for predicting holdup in a Karr column with different plate materials. In this study the constant  $C_\Psi$  was adjusted to fit the experimental data for each plate material via contact angle resulting in a power relation as follows:

$$C_{\Psi} = 1.406(1 + \cos \theta)^{-0.1373} \quad (13)$$

### 2.3. Characteristic velocity

The dispersed phase holdup can be related to the flow rates of the dispersed and continuous phases by a slip velocity,  $V_s$ , as follows<sup>(18)</sup>:

$$V_s = \frac{V_d}{x_d} + \frac{V_c}{(1 - x_d)} \quad (14)$$

Thornton<sup>(19)</sup> expressed the slip velocity as a function of a characteristic velocity,  $\bar{v}_0$ , as follows:

$$V_s = \bar{v}_0(1 - x_d) \quad (15)$$

Combining equations (14) and (15) gives the following:

$$V_d + \frac{V_c x_d}{1 - x_d} = \bar{v}_0 x_d (1 - x_d) \quad (16)$$

The holdup can be calculated based on the characteristic velocity by solving the cubic equation (16).

The first correlation for predicting the characteristic velocity was proposed by Thornton<sup>(19)</sup> for emulsion regime:

$$\bar{v}_0 = 0.6 \left( \frac{\gamma}{\mu_c} \right) \left( \frac{\rho_c \gamma^4}{\varphi \mu_c^5} \right)^{0.24} \left( \frac{d_a \rho_c \gamma}{\mu_c^2} \right)^{0.9} \left( \frac{\mu_c^4 g}{\Delta \rho \gamma^3} \right)^{1.01} \left( \frac{\Delta \rho}{\rho_c} \right)^{1.8} \left( \frac{\mu_d}{\mu_c} \right)^{0.3} \quad (17)$$

The correlation was later refitted by Groenier<sup>(20)</sup> for a wide range of PPPC geometries and chemical systems. Jahya<sup>(6)</sup> further modified Groenier's correlation by adding dimensionless groups which provided good prediction results for the PDDC and applied it to emulsion and transition regime data:

$$\begin{aligned} \bar{v}_0 = 6.889 \times 10^{-4} & \left( \frac{d}{d_a} \right)^{1.59} \left( \frac{A^2 \rho_c g}{\gamma} \right)^{0.13} \left( \frac{\gamma}{\mu_c} \right) \left( \frac{\rho_c \gamma^4}{\varphi \mu_c^5} \right)^{0.32} \\ & \times \left( \frac{d_a \rho_c \gamma}{\mu_c^2} \right)^{0.47} \left( \frac{\mu_c^4 g}{\Delta \rho \gamma^3} \right)^{0.81} \left( \frac{\Delta \rho}{\rho_c} \right)^{0.98} \left( \frac{\mu_d}{\mu_c} \right)^{0.12} \end{aligned} \quad (18)$$

## 3. Materials and Methods

### 3.1. Chemicals

The organic phase used in this study was 3 vol% Alamine 336<sup>®</sup> – Tri-n-octylamine (0.07 mol/L), supplied by BASF and 1 vol% isodecanol (0.054 mol/L), supplied by ExxonMobil in Shellsoll

2046<sup>®</sup>, supplied by Shell. The aqueous phase used was tap water. Sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) AR from Sigma Aldrich was used as the solute for mass transfer. H<sub>2</sub>SO<sub>4</sub> (0.4 wt%) and organic solution free of sulfuric acid was used for mass transfer from dispersed aqueous to organic.

The densities, viscosities and interfacial tension for this system were obtained from Jahya<sup>(6)</sup>. The interfacial tension was periodically measured using a FTÅ 200 supplied by First Ten Ångstroms and results agreed well with those published by Jahya<sup>(6)</sup>. The physical properties can be found in Table 1.

**Table 1: Physical properties of the aqueous and organic phases<sup>(6)</sup>**

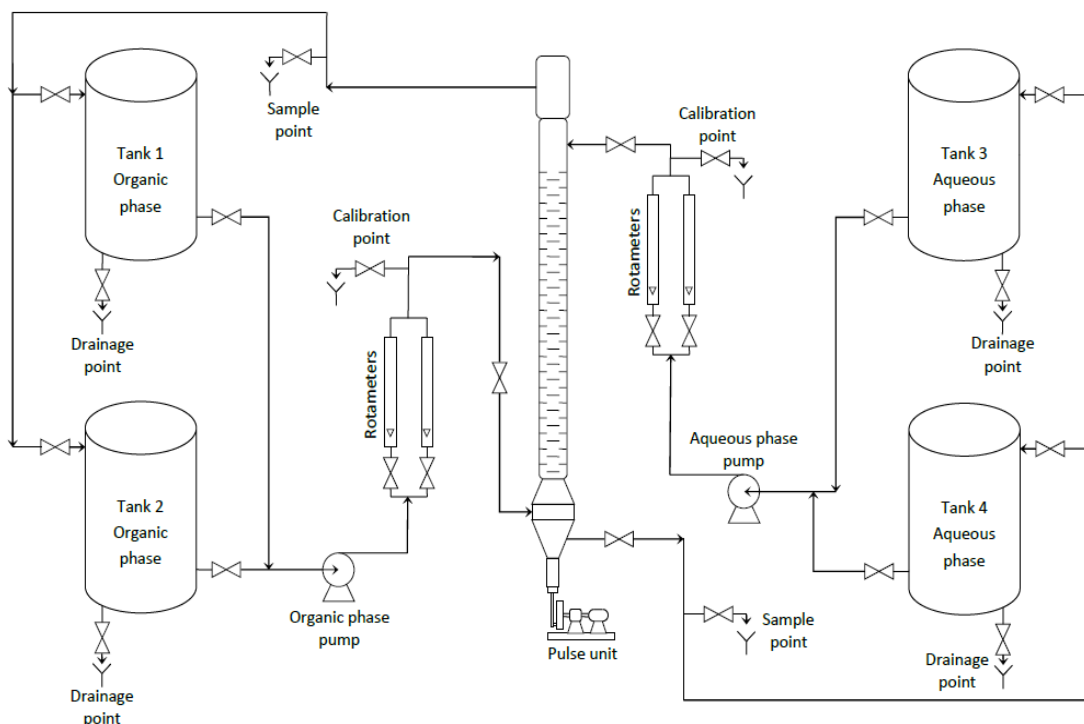
	Aqueous	Organic
Density (kg/m <sup>3</sup> )	998	805
Viscosity (Pa s)	1 x 10 <sup>-3</sup>	1.85 x 10 <sup>-3</sup>
Interfacial tension (N/m)	0.011	

### 3.2. Equipment

The pilot scale PDDC used in this study consists of a 1.0 m long QVF<sup>®</sup> precision bore glass column with an internal diameter of 72.5 mm. A t-piece is placed on top of the main column section to act as the organic phase outlet. Below the main column there is an expanded glass section which increases the column internal diameter to 100 mm and encloses the Stainless Steel (SS) distributor. It is supported by a SS conical adapter which reduces the diameter to 50 mm, and is supported by the piston-type pulsing unit. The piston-type unit consists of a motor, a variable speed unit (controls the frequency), a variable eccentric drive head (controls the stroke/amplitude), a crank arm and a plunger equipped with a Nylon cylinder and two rubber seals in a SS cylinder. The Plunger is used to provide a sinusoidal motion to the fluids in the column.

The continuous and dispersed phases are pumped into the column via distributors from their SS storage tanks using two 0.19 kW magnetically driven March pumps with flame proof electric motors. Two rotameters with SS floats, are installed in the inlet lines in parallel for each phase

to control the flow rates. All piping is made from 12.5 mm od SS tube connected with Swagelok Compression fittings. Two tanks of 100 L capacity are used to store the organic phase while two tanks of 150 L are used for the aqueous phase. All experiments were done at room temperature which for the pilot plant facility ranged 14°C to 21°C. Refer to Figure 1 for a process flow diagram of the column setup.



**Figure 1: Process Flow Diagram of Column Setup**

Three different plate materials were used in this study (Teflon, Nylon and Stainless Steel) to assess the change in column performance with different plate wetting characteristics. The geometric characteristics of each material are listed in Table 2.

**Table 2: Geometric characteristics of column internals: Teflon, Nylon and SS**

	Teflon	Nylon	Stainless Steel
Effective column height, $H_c$	815 mm	791 mm	790 mm
Effective column volume, $v_e$	3,365 cm <sup>3</sup>	3,265 cm <sup>3</sup>	3,261 cm <sup>3</sup>
Number of pairs (of discs & doughnuts), $N$	32	32	29
Plate thickness, $E$	3 mm	3 mm	1.6 mm
Disc diameter, $d_d$	62.4 mm	62.4 mm	63.5 mm
Doughnut aperture, $d_a$	36.9 mm	36.9 mm	35.0 mm
Plate spacing, $h$	9.73 mm	9.36 mm	12.02 mm
Compartment height, $h_c$	25.47 mm	24.72 mm	27.24 mm
Plate free area, $\varepsilon$	25.9 %	25.9 %	23.3 %

### 3.3. Procedures

#### 3.3.1. Column Operation

The stroke (amplitude peak to peak) was determined by measuring the liquid-liquid interface displacement when the piston was placed manually at the bottom and at the top positions in the column. The frequency was measured by a portable non-contact and reflective type digital tachometer. To start the column, the continuous phase pump is turned on and the continuous velocity is set at desired value using a rotameter. The dispersed phase pump is then turned on and the flow rate is gradually increased until the desired value is achieved. The height of the interface is manually controlled via a valve on the aqueous outlet. Steady state conditions were obtained when there was no change in the solute concentration in two consecutive samples. The column was operated over a range of operating conditions as shown in Table 3.

**Table 3: Operating Conditions**

	Teflon & Nylon	Stainless Steel
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Frequency, $f$	1.7, 2.0 & 2.3 Hz	1.5, 1.8 & 2.1 Hz
Amplitude, $A$	0.9 cm	1.2 cm
Pulsation rate, $Af$	1.5, 1.8 & 2.1 cm/s	1.8, 2.2 & 2.5 cm/s
Dispersed phase velocity, $V_d$	0.05 – 0.1 cm/s	
Continuous phase velocity, $V_c$	0.04 – 0.05 cm/s	
Phase flow ratio, $V_c/V_d$	0.37 – 1.06	
Temperature	Room temperature: 14 – 21 °C	

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### *Dispersed Phase Holdup*

Holdup was measured by the drainage technique as described by Gayler and Pratt<sup>(21)</sup>. Once steady state conditions were reached and the interface was at its desired position, the aqueous and organic flows into and out of the column were stopped by closing the inlet and outlet valves simultaneously. For aqueous dispersed, the droplets were allowed to settle and the column was drained to bring the interface back to its original height. For organic dispersed, the change in height of the interface was measured. The volume of dispersed phase ( $v_d$ ) was then used to calculate holdup,  $x_d$ , as follows:

$$x_d = \frac{v_d}{v_e} \quad (19)$$

Where  $v_d$  is the dispersed phase volume and  $v_e$  the total volume of the active section of the column.

### **3.3.2. Contact Angle**

The contact angles between the aqueous and organic phase on the different plate materials were measured with a FTÅ 200 supplied by First Ten Ångstroms using the captive needle approach. The dispersed phase was displaced/aspirated at rate of 0.189  $\mu\text{l/s}$ . A combined plastic and fused silica (hydrophobic) needle 34G (100  $\mu\text{m}$  ID, 164  $\mu\text{m}$  OD) was used to minimize distortion in the angle formed. The software captures one picture every second during the droplet displacement. The effects of roughness and heterogeneity of the plates have been neglected.

## 4. Results and Discussion

### 4.1. Contact Angle

The wetting characteristic of a solid material reflects its ability to maintain contact with a liquid. This is usually measured by contact angle. The advancing contact angles,  $\theta_a$ , of each plate material were measured with aqueous phase droplets to indicate the wettability of the surface of the disc and doughnuts inside the column (refer to results in Table 4). Teflon showed super-hydrophobic wetting characteristic in the liquid-liquid-solid system used in this study. Nylon showed intermediate wetting characteristics with an advancing contact angle of  $119^\circ$  for an aqueous droplet. Stainless Steel was the most hydrophilic plate material studied.

**Table 4: Contact angles measured with aqueous droplet for different plate materials**

Plate material	Wetting classification	Contact angle, $\theta_a$ (aqueous drop)
Teflon	Hydrophobic	$179^\circ$
Nylon	Intermediate	$119^\circ$
Stainless Steel	Hydrophilic	$95^\circ$

Teflon and Nylon  $A = 0.9cm$ ; Stainless Steel  $A = 1.2cm$

### 4.2. Operational Regimes

Operational regimes were observed over a range of pulsation frequencies with constant amplitude equal to plate spacing ( $h$ ). The organic and aqueous phase flow rates were set, frequency was adjusted and the system was allowed to stabilize. The system showed three operating regimes: mixer-settler, transition and emulsion. As the frequency was increased the three different operating regimes for the dispersed aqueous phase were observed as shown in Table 5. The dispersed phase flow rate also influences the operating regime e.g. Teflon at 1.5Hz may be classified as emulsion at high dispersed phase flow rate or mixer-settler at low flow rates.

**Table 5: Operating regimes - Dispersed aqueous phase**

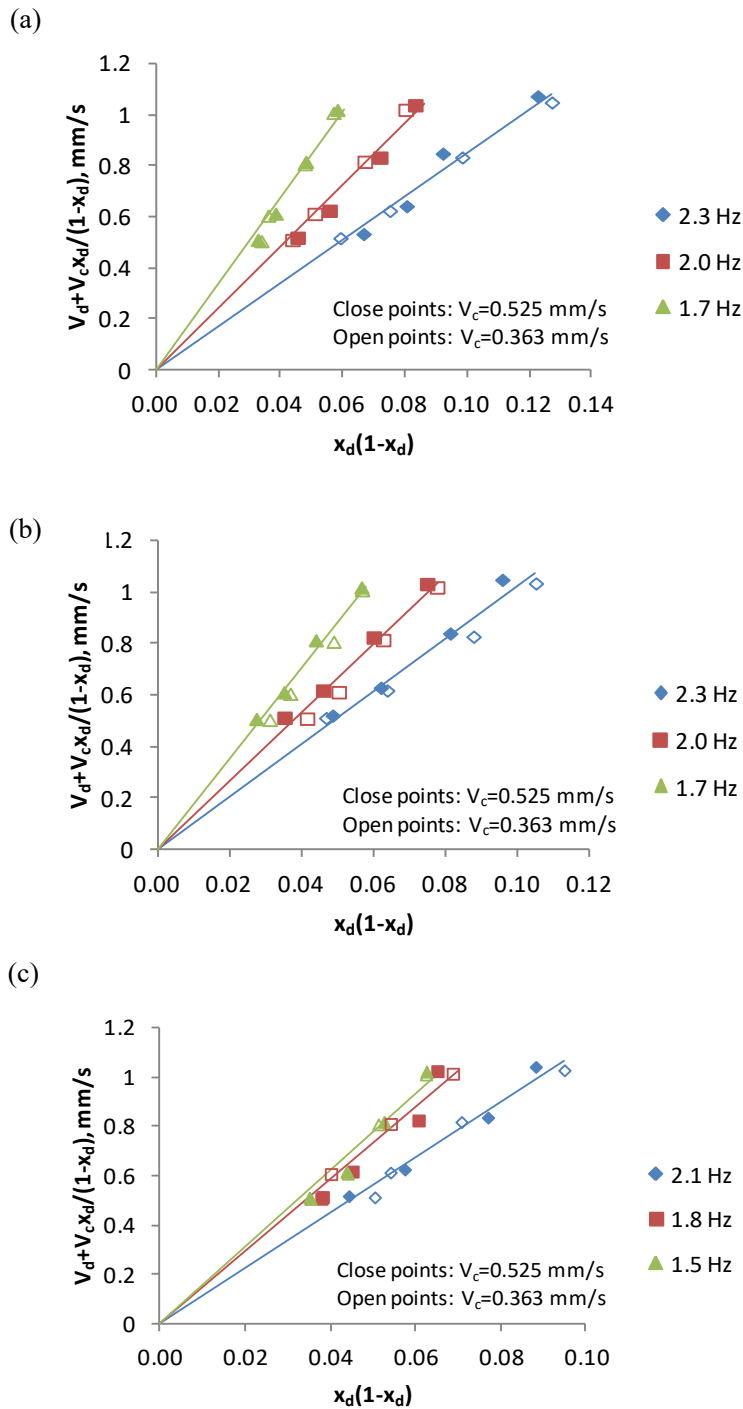
Frequency (Hz)	1	1.25	1.5	1.75	2.0
Teflon	Mixer-settler	Mixer-settler	Transition	Emulsion	Emulsion
Nylon	Mixer-settler	Mixer-settler	Transition	Emulsion	Emulsion
Stainless Steel	Mixer-settler	Transition	Emulsion	Emulsion	Emulsion

Teflon and Nylon  $A = 0.9cm$ ; Stainless Steel  $A = 1.2cm$

### 4.3. Characteristic Velocity

The characteristic velocity was determined by the slope of the plot  $V_d + V_c x_d / (1 - x_d)$  against  $x_d(1 - x_d)$ . The characteristic velocity is independent of the phase flow rates but dependent on the column geometry and physical properties of the system<sup>(22)</sup>.

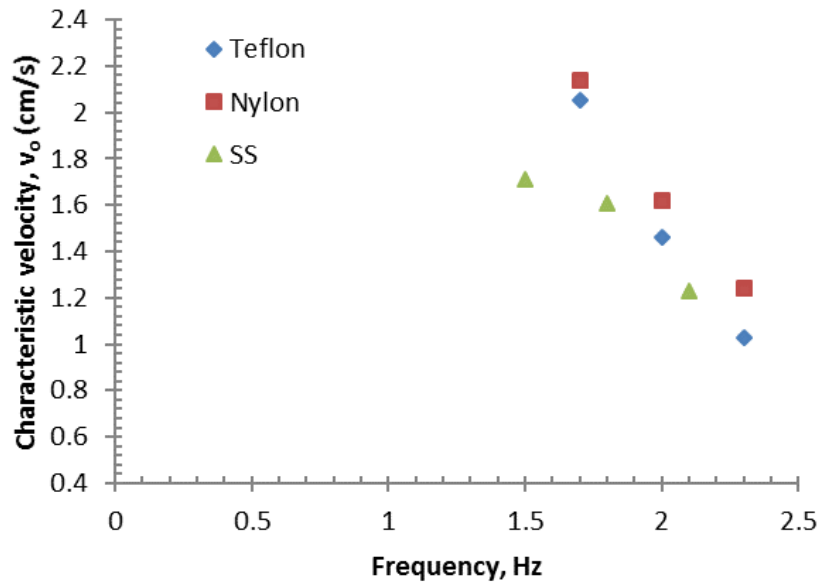
The characteristic velocity plots for Teflon, Nylon and Stainless Steel for mass transfer from dispersed aqueous to continuous organic can be found in Figure 2. From Figure 2 and Figure 3 it can be seen that the characteristic velocity decreased as pulsation rate increased for all plate materials.



**Figure 2: Characteristic Velocity plot for mass transfer from dispersed aqueous to continuous organic for (a) Teflon, (b) Nylon and (c) Stainless Steel**

By plotting the characteristic velocity for each plate material against pulsation rate (Figure 3) it can be seen that when the aqueous phase is dispersed both Teflon (non-wetting) and Nylon (semi-wetting) showed similar linear trends of decreasing characteristic velocity with increasing pulsation frequency. This trend is in agreement with other studies using pulsed columns such as Yi et al.<sup>(23)</sup>. However for the SS plates (wetting) this trend was not observed and

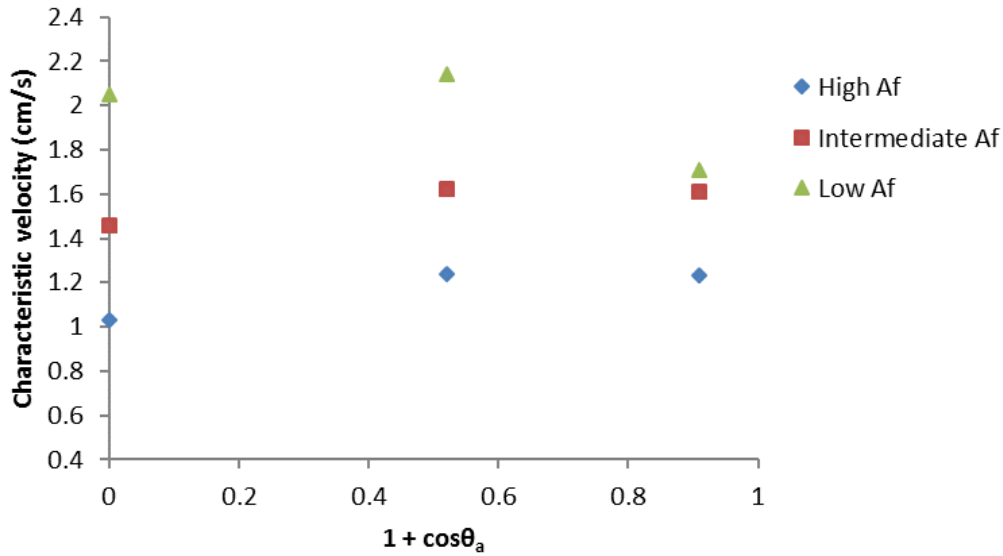
only a small change in characteristic velocity was observed at lower pulsation frequencies.



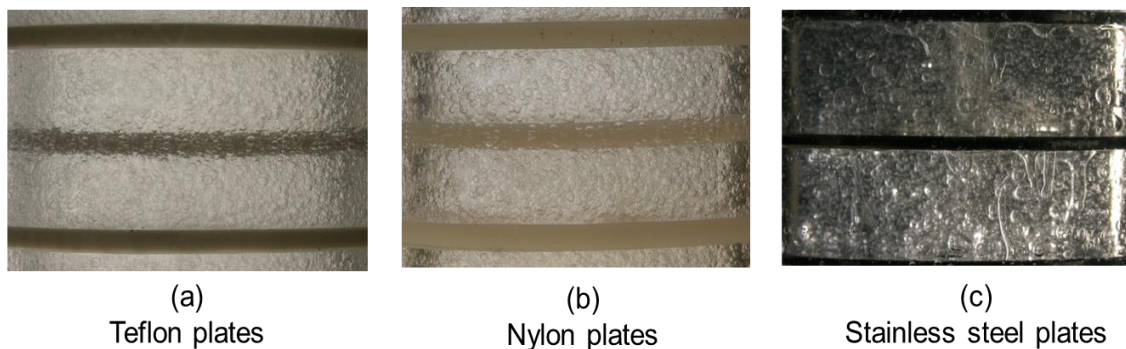
**Figure 3: Characteristic velocity as a function of pulsation frequency for dispersed aqueous phase and three different plate materials**

From Figure 3 it can be seen that when the plates are wetted by the continuous organic phase increasing pulsation intensity results in a linear decrease in  $\bar{v}_0$  and hence increase in holdup. Additionally from Figure 4 it can be seen that for high and intermediate pulsation rates, the characteristic velocity increases (holdup decreases) as the hydrophilicity of the plate material increases (as indicated by the value  $1 + \cos \theta_a$ ) for aqueous dispersed. However for low pulsation rates, aqueous films formed around the stainless steel plates resulting in an increase in holdup (and lower characteristic velocity). This result can also be seen from

Figure 5 which shows the dispersed aqueous phase clinging to the stainless steel plates in photo (c) in contrast to the even distribution of droplets in photos (a) and (b) for Teflon and Nylon plates.



**Figure 4: Effect of plate wettability on the characteristic velocity for dispersed aqueous at different agitation intensities:**



**Figure 5: Effect of plate wettability on droplet behavior for aqueous dispersed at a pulsation rate of 1.8 cm/s (all other conditions constant)**

From these results it can be concluded that plate wettability has an impact on dispersed phase holdup in a PDDC particularly at low pulsation rates. This is in agreement with the results from Christo et al.<sup>(22)</sup> who found that plate material had a larger impact on characteristic velocity in Karr reciprocating plate solvent extraction columns at lower agitation rates.

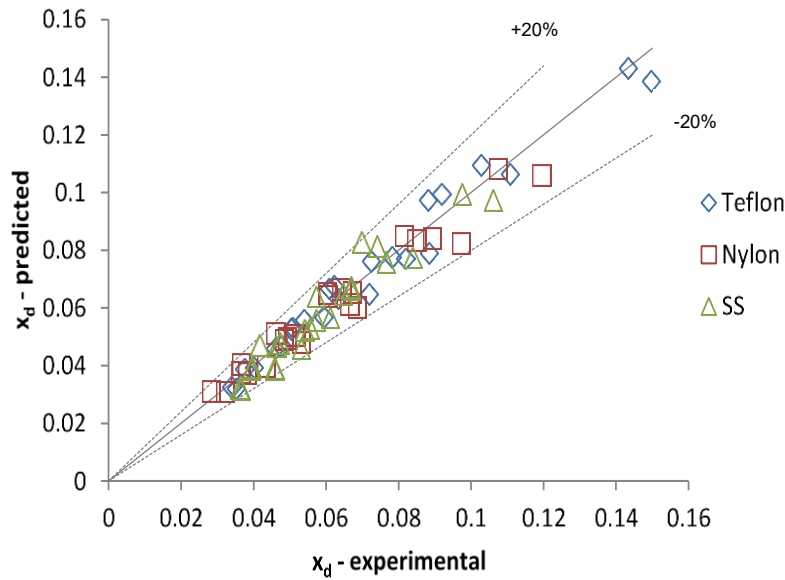
#### 4.4. Holdup Correlation

There are a number of correlations available in the literature for predicting dispersed phase holdup in solvent extraction columns as described in section 2. However there are no

correlations for a PDDC that incorporate the effect of plate wettability on dispersed phase holdup. A number of existing correlations from the literature were applied as published to the current data but none were suitable as indicated by large average absolute relative deviations (AARDs)<sup>(24)</sup>. The correlation presented by Jahya<sup>(6)</sup> for predicting characteristics velocity in PDDC, refer to equation (18), was most relevant to refit the current data. In order to incorporate plate wettability, linear regression of the experimental holdup data was completed to relate the coefficient from equation (18) to the advancing contact angle. Additionally the index that accounts for the effect of agitation intensity was set at 0.548 for this study. Thus equation (18) can be rewritten as follows:

For a PDDC with dispersed aqueous:

$$\begin{aligned} \bar{v}_0 = & (1.85 \cos \theta_a + 4.87) \times 10^{-6} \left(\frac{d}{d_a}\right)^{1.59} \left(\frac{A^2 \rho_c g}{\gamma}\right)^{0.13} \left(\frac{\gamma}{\mu_c}\right) \left(\frac{\rho_c \gamma^4}{\phi \mu_c^5}\right)^{0.548} \\ & \times \left(\frac{d_a \rho_c \gamma}{\mu_c^2}\right)^{0.47} \left(\frac{\mu_c^4 g}{\Delta \rho \gamma^3}\right)^{0.81} \left(\frac{\Delta \rho}{\rho_c}\right)^{0.98} \left(\frac{\mu_d}{\mu_c}\right)^{0.12} \end{aligned} \quad (20)$$



**Figure 6: Comparison between experimental and predicted holdup (dispersed aqueous) using the refitted Jahya characteristic velocity correlation, equation (20)**

As shown in Figure 6 this refitted correlation provided a good fit to the experimental data with

dispersed aqueous (overall AARD = 10.5 %). It should be noted that mass transfer will also influence the interfacial properties and wetting characteristics which will affect the drop size and holdup. Thus further work is needed to quantify these effects.

## 5. Conclusions

Dispersed phase holdup performance in a pulsed disc and doughnut solvent extraction column has been presented over a range of operating conditions for three different plate materials: hydrophobic (Teflon), intermediate wetting (Nylon) and hydrophilic (Stainless Steel). The characteristic velocity was found to increase with an increase in hydrophobicity of the plate material at high and intermediate pulsation rates for dispersed aqueous. However at low pulsation rates, aqueous films formed around the stainless steel plates resulting in an increase in holdup (and lower characteristic velocity) when operating under dispersed aqueous conditions. From this study it can be concluded that the operational regime and holdup are influenced by the wettability of the plates, especially at low pulsation rates. Data from this study was used to develop a modified version of Jahya characteristic velocity correlation<sup>(6)</sup> for a PDDC that incorporated the effects of plate wettability via advancing contact angle. This correlation was able to predict the holdup data from this study to within 10.5% for aqueous dispersed.

## 6. Nomenclature

$A$	Peak to peak amplitude, m
$C_0$	Discharge coefficient for flow through hole, dimensionless
$C_{\square}, C_{\Psi}, C_{\Gamma}$	Parameters in ,equations (5), (7) and (8), dimensionless
$d$	Column diameter, m
$d_a$	Doughnut aperture, m
$d_d$	Disc diameter, m
$E$	Plate thickness, m
$f$	Frequency, Hz
$g$	Acceleration due to gravity, $m/s^2$
$H_c$	Effective column height, m

$h$	Plate spacing, m
$h_c$	Compartment height, m
$k_1, k_2$	Parameter in eq (2), dimensionless
$N$	Number of pairs of disc and doughnuts
$n_1$ to $n_7$	Indexes eqs (5) to (8), dimensionless
$V$	Superficial velocity, m/s
$v_d$	Dispersed phase volume, L
$v_e$	Effective column volume, L
$\bar{v}_0$	Characteristics velocity, m/s
$V_s$	Slip velocity, m/s
$W$	Work of adhesion, N/m
$x_d$	Dispersed phase holdup, dimensionless

#### *Greek symbols*

$\gamma$	Interfacial tension, N/m
$\Delta\rho$	Density difference between phases, kg/m <sup>3</sup>
$\varepsilon$	Fractional free area, dimensionless
$\theta$	Contact angle
$\theta_a$	Advancing contact angle
$\mu$	Viscosity, Pa s
$\mu_w$	Viscosity of water and 20 °C, Pa s
$\rho$	Density, kg/m <sup>3</sup>
$\varphi$	Frictional power dissipation per unit mass of mixed phases: $\frac{2\pi^2(1-\varepsilon^2)}{3\varepsilon^2 C_0^2 h_c} (Af)^3$ in equations (5) and (9) and $\frac{\pi^2(1-\varepsilon^2)}{2\varepsilon^2 C_0^2 h_c} (Af)^3$ in equations (17) and (18), W/kg

#### *Subscripts*

$c$	Continuous phase
$d$	Dispersed phase

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