Comparison of the hydrodynamic performance of pulsed solvent extraction columns with Tenova pulsed column kinetics Internals (TPC-KI) and standard disc and doughnut Internals for copper extraction using LIX 84 system

Wen Li, Yong Wang, Kathryn A. Mumford, Kathryn H. Smith, Geoffrey W. Stevens*

Department of Chemical and Biomolecular Engineering, The University of Melbourne, Parkville, Victoria 3010,
Australia

Abstract:

The dispersed phase holdup and droplet size are important parameters for understanding the hydrodynamic performance of pulsed solvent extraction columns. This study provides a comparison of the hydrodynamic performance between a Tenova Pulsed Column with Kinetics Internals (TPC-KI) and a pulsed column with standard disc and doughnut internals. The TPC-KI internals are designed to improve the performance of systems with slow reaction kinetics, such as copper extraction with LIX 84 which was the focus of the current study. Holdup and drop size have been measured as a function of pulsation intensity and phase velocities for both types of column internals. Lower holdup and larger drop sizes were obtained with the TPC-KI internals which indicates a higher flood point and a higher throughput. Correlations have been developed to predict the performance of these internals over a range of operating conditions.

1. Introduction

Solvent extraction columns are one of the most effective options for achieving extraction of solutes by pumping two immiscible liquid phases counter-currently through a column. This technique has been used widely in numerous industries including hydrometallurgy, environmental waste purification and to produce relatively pure chemical compounds such as pharmaceuticals ^[1].

The efficiency of industrial solvent extraction columns can be successfully enhanced with the introduction of internals such as the Tenova standard disc and doughnuts which are especially useful when the liquid-liquid system is not limited by kinetics. Tenova standard disc and doughnut internals^[2-4] have been used in solvent extraction columns for applications in the chemical industry and minerals processing (e.g. uranium, nickel, cobalt, zinc and titanium). Recently a novel column internal structure has been designed and named the Tenova Pulsed Column – Kinetics Internals (Tenova kinetics internals, TPC-KI). These new internals are proposed to achieve less back-mixing, higher column holdup and improved mass transfer at the same flux, which is particularly useful for relatively slow kinetics systems.

To evaluate the performance of these new internals it is necessary to measure and compare both the hydrodynamic and mass transfer performance using these new internals and the standard disc and doughnut internals. In this study, the hydrodynamic performance parameters including dispersed phase holdup, characteristic velocity and dispersed phase Sauter-mean diameter were measured with standard disc and doughnut internals and the Tenova kinetics internals. The results were also fitted to empirical correlations to predict performance over a wide range of operating conditions. Experiments were completed using a newly-built 2 m high 76 mm diameter pilot scale pulsed solvent extraction column with copper extraction using LIX 84 extractant which is known to be limited by kinetics^[1].

2. Background

The importance of the hydrodynamic performance of solvent extraction columns, specifically dispersed phase holdup and droplet size, is mainly reflected in their effect on mass transfer performance, which is dependent on the area of contact or the interfacial area (a) between the two immiscible phases. The interfacial area is dependent on dispersed phase holdup x_d and Sauter-mean diameter d_{32} as follows:

$$a = \frac{6x_d}{d_{32}} \quad (1)$$

2.1 Dispersed Phase Holdup

Dispersed phase holdup (x_d) is defined as the volume fraction of the dispersed phase occupied in the active section of the column, and it can be expressed as in equation 2:

$$x_d = \frac{V_{\text{disp}}}{V_{\text{active}}} \quad (2)$$

In which V_{disp} is the volume of the dispersed phase in column, and V_{active} represents the total volume of both dispersed phase and continuous phase in the active section of the column.

Correlations enable prediction of the dispersed phase holdup in solvent extraction columns in either an explicit way or an implicit way. Implicit correlations of holdup are related to the continuous and dispersed phase velocities (v_c and v_d) by a characteristic velocity (v_0)^[5]. By assuming the buoyancy force is proportional to the density difference between the mixed and dispersed phases, the characteristic velocity is expressed as a function of holdup according to Eq. 3:

$$\frac{v_d}{x_d} + \frac{v_c}{1-x_d} = v_0(1 - x_d) \quad (3)$$

Where v_0 represents the mean relative velocity of the droplets extrapolated to zero phase velocities or holdup at a particular frequency and amplitude. This can generally be obtained from experimental data of v_c , v_d and x_d by plotting $v_d + \frac{v_c x_d}{1-x_d}$ versus $x_d(1 - x_d)$ with the slope being equal to v_0 .

Kumar and Hartland^[6] developed an explicit holdup correlation for a pulsed sieve-plate column that was dependent on physical properties, operating conditions and column geometry using 725 data points in the absence of solute transfer, giving:

❖ For mixer-settler region,

$$x_d = 3.91 \times 10^{-3} \left(\frac{A^2 \rho_c g}{\gamma} \right)^{-0.26} \left(\frac{f^4 \gamma}{\rho_c g^3} \right)^{-0.19} \left(\frac{v_d^4 \rho_c}{g \gamma} \right)^{0.28} \left(1 + \frac{v_c}{v_d} \right)^{0.19} \left(\frac{\Delta \rho}{\rho_c} \right)^{-0.81} \left(\frac{\mu_d^4 g}{\rho_c \gamma^3} \right)^{-0.13} \quad (4)$$

❖ For transition region,

$$x_d = 6.91 \left[\frac{(Af)^3 \rho_c^{\frac{1}{4}}}{\beta h \gamma^{\frac{1}{4}} g^{\frac{5}{4}}} \right]^{0.31} \left(\frac{v_d^4 \rho_c}{g \gamma} \right)^{0.30} \left(1 + \frac{v_c}{v_d} \right)^{0.14} \left(\frac{\Delta \rho}{\rho_c} \right)^{-0.79} \left(\frac{\mu_d^4 g}{\rho_c \gamma^3} \right)^{-0.01} \quad (5)$$

❖ For emulsion region,

$$x_d = 3.73 \times 10^{-3} \left[\frac{(Af)^4 \rho_c}{g \gamma} \right]^{0.62} \left(\frac{v_d^4 \rho_c}{g \gamma} \right)^{0.31} \left(1 + \frac{v_c}{v_d} \right)^{0.45} \left(\frac{\Delta \rho}{\rho_c} \right)^{-2.20} \left(\frac{\mu_d^4 g}{\rho_c \gamma^3} \right)^{-0.29} \quad (6)$$

These equations (Eq. 4 - 6) were reported to reproduce the experimental data to within 13%.

Later Kumar and Hartland^[7, 8] presented an empirical correlation for holdup prediction in a pulsed perforated-plate extraction column using 15 different data sources for 23 different liquid-liquid systems with and without solute transfer as follows:

$$x_d = k_1 \exp[k_2 |Af - (Af)_m|] v_d^{0.86} (v_c + v_d)^{0.28} \Delta\rho^{-0.30} \rho_d^{-0.93} \mu_d^{0.77} \varepsilon^{-0.56} h^{-0.56} \quad (7)$$

in which

$$(Af)_m = 9.69 \times 10^{-3} \left(\frac{\gamma \Delta\rho^{1/4} \varepsilon}{\mu_d^{3/4}} \right)^{0.33} \quad (8)$$

The values for k_1 and k_2 in the case of no solute transfer, solute transfer from the continuous phase to the dispersed phase ($c \rightarrow d$) and in the opposite direction ($d \rightarrow c$) are given in the Table 1. The average absolute relative deviation of this correlation was 17.8%.

Table 1 Constants k_1 and k_2 for holdup correlation^[7, 8]

	k_1	k_2
without solute transfer	2.10×10^6	44.53
c $\rightarrow$ d transfer	2.14×10^6	44.53
d $\rightarrow$ c transfer	1.10×10^6	50.56

To extend the operating range to include disc and doughnut column operation under no and low pulsation rates, Wang et al.^[2] modified the correlation (Eq. 7) developed by Kumar and Hartland^[7, 8] using experimental data from a pulsed disc and doughnut solvent extraction column with an Alamine 336 - Shellsol 2046 system to form equation 9:

$$x_d = k_1 \exp[k_2 |Af - (Af)_m|] \left(\frac{v_d^4 \rho_c}{g\gamma} \right)^{0.27} \left(\frac{v_c + v_d}{v_d} \right)^{0.32} \left(\frac{\Delta\rho}{\rho_c} \right)^{-0.98} \left(\frac{\mu_d}{\mu_c} \right)^{0.18} \quad (9)$$

The values for k_1 and k_2 for the condition without mass transfer are 1.577 and 64.11 respectively. The revised dimensionless relationship predicted experimental results with an average absolute relative error for non-pulsing, mixer-settler and emulsion regions of 17.3%, 7.5% and 14.8% respectively.

2.2 Drop size

Sauter-mean diameter (d_{32}), expressed via Equation 10, is determined by relative rates of droplet breakage and coalescence. The initial drop is dependent on the distributor for the dispersed phase, and the droplets pass through the column and change their sizes due to pulsation and column geometry. A dynamic equilibrium Sauter-mean diameter (d_{32}) will be reached at the middle section of the column and d_{32} can be calculated as follows.

$$d_{32} = \frac{\sum_{i=1}^n n_i d_i^3}{\sum_{i=1}^n n_i d_i^2} \quad (10)$$

d_{32} is a key factor for determining the interfacial area for mass transfer performance, and also effects the dispersed phase holdup and residence time. Smaller droplets will result in a higher mass transfer rate, but may also result in an earlier onset of flooding and reduce the allowable throughput of the column.

Kumar and Hartland^[9] presented a unified correlation for droplets Sauter-mean diameter prediction in a range of columns by assuming the disruptive energy due to continuous phase turbulence is determined by the density difference ($\Delta\rho$):

$$\frac{d_{32}}{h} = \frac{C_\psi \varepsilon^{0.32}}{\frac{1}{1.55 \left(\frac{\gamma}{\Delta \rho g h^2} \right)^{1/2}} + \frac{1}{0.42 \left[\left(\frac{\psi}{g} \right) \left(\frac{\Delta \rho}{g \gamma} \right)^{1/4} \right]^{-0.35} \left[h \left(\frac{\Delta \rho g}{\gamma} \right)^{1/2} \right]^{-1.15}}} \quad (11)$$

The values of C_ψ for pulsed and Karr columns are 1, 0.92 and 1.67 for no mass transfer, $c \rightarrow d$ and $d \rightarrow c$ directions of mass transfer, respectively. The average relative deviation of the equation is 16.1%.

Van Delden et al.^[10, 11] modified the correlation of Kumar and Hartland^[12], which was proposed for the prediction of Sauter-mean diameter in pulsed sieve plate extraction columns, by fitting parameters C_1 and C_3 with their experimental data for pulsed disc and doughnut solvent extraction columns.

$$\frac{d_{32}}{\sqrt{\frac{\gamma}{\Delta \rho g}}} = C_1 \varepsilon^{n_1} \left(h \frac{\sqrt{\rho_* g}}{\gamma_*} \right)^{n_2} \left(\frac{\mu_d g^{1/4}}{\rho_*^{1/4} \gamma_*^{3/4}} \right)^{n_3} \left(\frac{\gamma}{\gamma_*} \right)^{n_4} [C_2 + \exp \left\{ C_3 \frac{Af}{\varepsilon \left(\frac{\gamma_* g}{\rho_*} \right)^{1/4}} \right\}] \quad (12)$$

The parameters C_1 , C_2 , C_3 , n_1 , n_2 , n_3 and n_4 in Eq. 12 are 2.84, 0.16 -2.59, 0.30, 0.18, 0.14 and 0.06, respectively. The parameters ρ_* and γ_* are the density and surface tension of water at 20°C. The average absolute relative deviation (AARD) of the correlation is 22.2%, and will become larger for a system with low interfacial tension. The influence of dispersed phase velocity (v_d) was not taken into account for this study.

Torab-Mostaedi et al.^[13] derived a new empirical correlation for d_{32} based on the research of Van Delden et al.^[10, 11] for all operating regions with physical properties of the liquid-liquid systems, column geometrical characteristics and operating parameters. Equation 13 can reproduce the experimental data with the average relative deviation of 7.3%.

$$\frac{d_{32}}{\sqrt{\frac{\gamma}{\Delta\rho g}}} = 33.53 \times 10^{-3} \left(\frac{Af^4 \rho_c}{g\gamma} \right)^{-0.283} \left(\frac{d_a \rho_c \gamma}{\mu_c^2} \right)^{0.29} \left(\frac{\rho_c \gamma^4}{\psi \mu_c} \right)^{-0.13} \left(\frac{\Delta\rho}{\rho_c} \right)^{2.86} \left(\frac{\mu_d}{\mu_c} \right)^{0.085} \left(\frac{h}{d_a} \right)^{-0.734} (1 + L)^{0.34} \quad (13)$$

Wang et al.^[3] developed an empirical correlation based on 244 droplet size experimental data points from operation of a pulsed disc and doughnut extraction column using seven different liquid-liquid systems and pulsation rates which varied from 0 to 0.04 m/s.

$$\frac{d_{32}}{\sqrt{\frac{\gamma}{\Delta\rho g}}} = 31.7 \varepsilon^{1.6} \left(\frac{v_c \mu_c}{\gamma} \right)^{0.116} \left(\frac{v_d}{v_c} \right)^{0.118} \left[0.324 + \exp \left\{ \frac{-6.27 Af}{\varepsilon \left(\frac{\gamma_* g}{\rho_*} \right)^{0.25}} \right\} \right] \quad (14)$$

The regressed parameters for this column are included in equation 14, and the average absolute relative deviation between experimental data and predicted results is 17.8%.

Prediction of holdup and drop size in solvent extraction columns is important for understanding and optimising column performance. The use of novel internals to improve the performance of slow kinetics systems is also important. The aim of this research is to determine the hydrodynamic performance of a new type of column internal designed to enhance the kinetics of relatively slow solvent extraction systems. This will be done by obtaining experimental hydrodynamic data, including holdup and drop size, using a 2 metre high pulsed column with standard and the new internals with a liquid-liquid system with relatively slow kinetics. The data will then be used to update empirical

correlations with regressed parameters in order to predict the performance of these new internals. Comparing the performance of the standard disc and doughnut internals with the Tenova kinetics internals has not been done before and as such is important and novel.

3. Experiment

3.1. Liquid-liquid system

The liquid-liquid system used in this study was tap water as the aqueous phase and 6.4 v/v % LIX 84 (2-hydroxy-5-nonylaceto-phenone oxime), supplied by BASF, diluted with Shellsol 2046, supplied by Shell as the organic phase. In future mass transfer experiments the aqueous phase will contain ~ 2 g/L cupric sulphate. This liquid-liquid system is known to be limited by its reaction rate ^[1] which enables a useful comparison of the column performance between the two different internals. The physical properties of each phase including density, ρ , viscosity, μ , and interfacial tensions $\gamma_{\text{aq-org}}$ were measured and results are shown in Table 2.

Table 2 Physical properties of liquid-liquid system

Aqueous Phase	Organic Phase	ρ_{aq} (kg/m ³)	ρ_{org} (kg/m ³)	μ_{aq} (Pa·s)	μ_{org} (Pa·s)	$\gamma_{\text{aq-org}}$ (N/m)
6.4 v/v % LIX						
Tap water	84 in Shellsol 2046	998	810	1.02 x 10 ⁻³	2.08 x 10 ⁻³	0.022

3.2. Equipment

The 2 m high pilot-scale pulsed solvent extraction column used in this study was designed and built in the Solvent Extraction Pilot Plant in the Department of Chemical and Bio-molecular Engineering at The University of Melbourne. The main column body consists of four QVF precision pipeline components manufactured from borosilicate glass: an unequal tee piece (250 mm long, 76 mm diameter), two precision bore glass tubes (each 1000 mm long, 76 mm diameter) and a concentric reducer (80 mm diameter expanded to 150 mm). All four glassware sections were connected by couplings and appropriate seals. The pulsation unit was assembled with a 1.5 kW three-phase induction motor with

4 pole 90 frame from Airport Electric Motors, a zero-max gearbox purchased from Naismith and a piston unit manufactured in-house. The pulsation unit was connected to the bottom of the main column body to supply pulsing power. Pulsation rates of 15 ~ 180 rpm were achievable and the piston unit can reach a maximum pulsing amplitude of 15 cm, which combined offers a maximum pulsing intensity of 0.45 m/s. Four stainless steel tanks with a volume capacity of 200 L each were connected to the aqueous phase inlet and outlet, and the organic phase inlet and outlet with SS tubing. Two pumps and four rotameters were also arranged before the aqueous and organic phase inlets to the column to enable a range of dispersed and continuous phase flow rates to be studied.

Inside the main column section, two different types of internals (standard disc and doughnut and Tenova kinetics internals) could be studied – refer to schematic drawings and photographs in Figure 1 and Figure 2. The internals were fastened to a stainless steel column cover at the top of the column, in order to prevent the internals from moving up and down due to pulsing of the fluid inside the column. For each set of internals, 77 discs and 76 doughnut plates were arranged alternately and spaced 9.8 mm apart inside the column, resulting in a 19.6 mm compartment height. In addition, the discs were of 68 mm diameter and the doughnuts had apertures of 36 mm diameter, which gave open free area for each plate of 22.4%.

All experiments were operated at room temperature which ranged from 15 ~ 22 °C. The standard disc and doughnut internals and Tenova kinetics internals were both made of PVDF (polyvinylidene fluoride), which is a specialty plastic that gives a high resistance to solvents, acids and bases.

Although both sets of internals were made from PVDF, the wettability of each internal varied due to variation in surface roughness with an advancing contact angle of 69° for the standard internals and 50° for the Tenova kinetics internals with tap water-air-PVDF sheets three-phase system. Contact angles were measured using an Attension Theta optical tensiometer (Biolin Scientific, Sweden).

Figure 1. Schematic drawings for standard disc and doughnut internals (left) and Tenova kinetics internals (right)

Figure 2. Standard disc and doughnut internal plates made of PVDF (top); Tenova kinetics internals made of PVDF (bottom)

3.3. Column Operation

To measure the dispersed phase holdup, the amplitude and frequency on the pulsation unit were first adjusted to their desired values. The continuous phase pump was then started and the continuous phase flowrate adjusted to the desired value using a rotameter until the column section was filled to around 80% with continuous phase liquid. The dispersed phase pump was then turned on and the dispersed phase flowrate was adjusted to its set value using the dispersed phase rotameter. The dispersed phase droplets would then fall down (organic continuous mode) or rise up (aqueous

continuous mode). A clear interface between the aqueous phase and organic phases was maintained at the bottom (organic continuous mode) or top (aqueous continuous mode) of the column. The interface was maintained at the same level in the column by adjusting the flowrate of the aqueous outlet.

The drainage technique proposed by Gayler and Pratt^[5] was used to measure dispersed phase holdup. The column was operated at steady-state conditions and then both the continuous and dispersed phase inlets, aqueous phase outlet and pulsation unit were shut down simultaneously. The original steady-state interface position was noted and the pulsation unit was turned on until all the dispersed phase droplets accumulated at the interface. The pulsation unit was turned off and the final interface position was noted. The dispersed phase volume in the column section was then drained into a volumetric cylinder until the interface was at its original height. The dispersed phase holdup x_d , can then be calculated via Equation (2) with the measured dispersed phase volume and the pre-determined total volume in the column.

The drop size was also measured at steady-state conditions using a photographic technique in the middle section of the column under the desired operating conditions. With the assistance of a Perspex cubical viewing box located at the middle of the column, a Canon G16 camera was used to take digital photographs of dispersed phase droplets and the diameters of the droplets were later measured using Nano Measurer 1.2 software. This data was then used to obtain the Sauter-mean diameter via equation (10). In some cases, the droplets were elliptical, instead of circular, and their equivalent diameters were calculated by the measured minor and major axes (d_1 and d_2) as per Eq. 11,

$$d_e = \sqrt[3]{d_1^2 d_2} \quad (11)$$

4. Results and discussion

4.1. Dispersed phase holdup

In this study, the aqueous phase was used as the dispersed phase and the organic phase was used as the continuous phase. The effect of pulsing intensity on the dispersed phase holdup was studied using both standard disc and doughnut internals and Tenova kinetics internals. The phase flow rates were varied from 6.62×10^{-4} to 1.43×10^{-4} m/s for the organic continuous phase velocity and 7.35×10^{-4} to 1.47×10^{-4} m/s for the aqueous dispersed phase velocity. Figure 3 shows that the dispersed phase holdup increased with dispersed phase velocity at a constant continuous phase velocity of 6.62×10^{-4} m/s (A/O ratio are 1.11, 1.64 and 2.22) for both the standard disc and doughnut internals and the Tenova kinetics internals. For both sets of internals holdup initially decreased until a minimum holdup value was reached at a pulsation intensity of $(Af)_{\min} = 0.018 \pm 0.002$ m/s for the standard disc and doughnut internals and $(Af)_{\min} = 0.012 \pm 0.002$ m/s for the Tenova kinetics internals. Further increasing the pulsation intensity up to 0.03 m/s resulted in increasing holdup to values that were lower than the holdup under the non-pulsing condition. For both sets of internals, increasing the dispersed phase velocity resulted in higher holdup at the same pulsation intensity through the operating range of 0 m/s to 0.03 m/s.

Results from Figure 3 also show that the different column internals result in different dispersed phase holdup under the same operating conditions. Lower dispersed phase holdup was measured with the Tenova kinetics internals particularly at low pulsation intensity from 0 m/s to 0.015 m/s. Under non-pulsing conditions, the dispersed phase holdup with the standard disc and doughnut internals was on average 56% higher than that with the Tenova kinetics internals. Additionally, the pulsation intensity at minimum dispersed phase holdup ($(Af)_{\min}$) is shifted from $0.015 \pm 0.002 \sim 0.02 \pm 0.002$ m/s with the Tenova kinetics internals to $0.01 \pm 0.002 \sim 0.015 \pm 0.002$ m/s with the standard internals for all dispersed phase velocity conditions examined in this study.

Figure 3. Effect of pulsation intensity and dispersed phase velocity on the dispersed phase holdup x_d for different column internals (Open points are standard disc and doughnut internals and closed points are Tenova kinetics internals)

Similar trends can be seen in Figure 4 which plots the change of dispersed phase holdup with varying pulsation intensities and continuous phase velocities at a constant dispersed phase velocity of 7.35×10^{-4} m/s (A/O ratios are 0.51, 0.68 and 1.11) for the two different column internals. The continuous phase velocity had very little impact on the dispersed phase holdup. In conclusion the dispersed phase holdup obtained with the standard disc and doughnut internals was higher than that with the Tenova kinetics internals for all operating conditions investigated in this study.

Figure 4. Effect of pulsation intensity and continuous phase velocity on the dispersed phase holdup x_d for different column internals (Open points are standard disc and doughnut internals and closed points are Tenova kinetics internals)

The holdup results from this study imply that a solvent extraction column with the Tenova kinetics internals has a wider operational work range due to its lower holdup and hence higher flooding point. For the performance of a solvent extraction column, the flood point determines the workable range of operating conditions. When flooding occurs, the dispersed phase droplets will not be able to flow through the column, holdup will increase significantly and phase inversion may occur. To maintain high efficiency in the column, it is necessary to prevent the column from flooding. However in some cases increasing the dispersed phase velocity or the continuous phase velocity is required to improve the yield. Therefore using the Tenova kinetics internals would enable operation at higher phase velocities before flooding conditions are reached.

When operating below the flood point, higher dispersed phase holdup will provide improved separation efficiency due to increased interfacial area for the mass transfer process. Figure 3 and Figure 4 indicate that the standard disc and doughnut internals offer higher holdup. However despite both internals being produced from PVDF, the surface roughness of the PVDF sheets was different for the two types of internals resulting in variation in surface wettability.

With a lower advancing contact angle (measured using water-air-PVDF), the Tenova kinetics internals have a smaller contact angle than the standard disc and doughnut internals, which may lead to water droplet accumulation on the internal surface and result in lower holdup. For the Tenova kinetics internals, the water droplets exiting the distributor tended to accumulate on the surface of the internals and flow along the column wall rather than passing down through the column without interaction with the column internals (see Figure 5). As pulsation is increased this effect is reduced. In Figure 5, the colour of solvent was yellow as a result of a small slight amount of copper remaining in the organic phase after stripping, but there was no observable change on the physical properties.

Figure 5. Dispersed phase droplets inside the column ($v_c = 6.64 \times 10^{-4}$ m/s, $v_d = 7.35 \times 10^{-4}$ m/s, $A_f = 0.005$ m/s); for standard disc and doughnut internals (left) and Tenova kinetics internals (right)

Figure 6. Effect of dispersed phase velocity on dispersed phase holdup at constant continuous phase velocity ($v_c = 6.62 \times 10^{-4}$ m/s) and pulsation intensity ($Af = 0.01$ m/s)

The influences of dispersed phase velocity and continuous phase velocity on holdup are shown in Figure 6 and Figure 7 respectively at constant pulsation intensity. In Figure 6, under the same continuous phase velocity of 6.62×10^{-4} m/s, the dispersed phase holdup for both internals increases linearly with increasing dispersed phase velocity from 3.68×10^{-4} m/s to 1.84×10^{-4} m/s. For the Tenova kinetics internals, holdup increases with dispersed phase velocity at a slower rate than the standard internals. The variation in holdup with continuous phase velocity, shown in Figure 7, shows that there is no noticeable trend when the continuous phase velocity is increased from 2.39×10^{-4} m/s to 1.84×10^{-4} m/s.

Figure 7. Effect of continuous phase velocity on dispersed phase holdup under same dispersed phase velocity ($v_d = 7.35 \times 10^{-4}$ m/s) and pulsation intensity ($A_f = 0.01$ m/s)

4.2. Correlation for dispersed phase holdup

There are a number of correlations in the literature for predicting dispersed phase holdup in solvent extraction columns but these equations are generally only applicable for the geometries and operating conditions for which they were originally developed. Therefore it is important to compare and update these correlations for new column internals for pulsed columns. There are generally two approaches used for predicting holdup: separate equations for the mixer-settler, transition and emulsion regions and a single equation that combines the operating regimes. The empirical correlation for holdup (refer to Eq. 9) developed by Wang et al.^[2] based on the performance of a one meter high pilot-scale pulsed standard disc and doughnut extraction column, used a single equation for the prediction of experimental holdup data through the three operating regimes. The average absolute relative deviation (AARD) between the experimental data in the current study with the predicted holdup from Equation 9 with the parameters provided in the literature is 21.6%. To minimize the AARD Wang et al.'s correlation^[2] was refitted to the pilot-scale experimental

data for both the standard disc and doughnut internals and the Tenova kinetics internals. The correlated relationship is shown as follows:

$$x_d = k_1 \exp [k_2 |Af - (Af)_m| (\frac{v_d^4 \rho_c}{g\gamma})^{0.31} (\frac{v_c + v_d}{v_d})^{0.32} (\frac{\Delta \rho}{\rho_c})^{-0.98} (\frac{\mu_d}{\mu_c})^{0.18}] \quad (15)$$

The refitted constants k_1 and k_2 for each internals are presented in table 3:

Table 3 Regressed parameters for the correlation of dispersed phase holdup

Internals	Operating regime	k_1	k_2	AARD
Standard disc and doughnut internals	mixer-settler	7.34	80.16	5.92%
	transition & emulsion	6.51	43.86	
Tenova kinetics internals	mixer-settler	4.76	40.46	5.60%
	transition & emulsion	6.14	14.40

Figure 8. Comparison of the experimental dispersed phase holdup and predicted holdup with Eq. 15

4.3. Characteristic velocity v_0

The characteristic velocity can be determined by plotting $v_d + \frac{v_c x_d}{1-x_d}$ against $x_d(1-x_d)$. Figure 9 shows the characteristic velocity plots under three pulsation intensities for the standard disc and doughnut internals and the Tenova kinetics internals. For the operational pulsation intensity from 0.005 m/s to 0.03 m/s, the corresponding characteristic velocities were determined and provided in Table 4.

Figure 9. Characteristic velocity plots for different pulsation intensities for the two different internals

Table 4 Characteristic velocities under different pulsation intensities for standard disc and doughnut internals and Tenova kinetics internals

Af (m/s)	0.005	0.01	0.015	0.02	0.025	0.03
v_0 for standard disc and doughnut internals (m/s)	0.008	0.0115	0.014	0.0138	0.0107	0.0095
R^2 for each linear correlation	0.9726	0.9642	0.8997	0.9803	0.9889	0.9886
v_0 for Tenova kinetics internals (m/s)	0.0151	0.0187	0.0175	0.0157	0.0147	0.0141
R^2 for each linear correlation	0.9618	0.9819	0.9925	0.9857	0.9669	0.9408

Additionally, as observed in Figure 10 pulsation intensity has an influence on the characteristic velocity. With increasing pulsation intensity, the characteristic velocity rises to a maximum at the pulsation intensity where the minimum holdup was measured, and then gradually decreases. Overall the characteristic velocities determined for the Tenova kinetics internals were higher than that for standard disc and doughnut internals for all operating pulsation intensities. Higher characteristic velocity indicates lower holdup when operating at defined operating conditions.

Figure 10. Effect of pulsation intensity on the characteristic velocity for the two different internals

4.4. Drop size - Sauter mean diameter d_{32}

Drop size is another key factor for estimating the hydrodynamic and mass transfer performance of a solvent extraction column. The sauter mean diameter, d_{32} , was calculated using photographs taken during column operation that contain 150 - 300 droplets for each operating condition. The range of aqueous to organic phase ratios studied was from 0.51 to 2.22 (the aqueous and organic phase velocities are the same as used in dispersed phase holdup experiments).

In Figure 11, the sauter mean diameter of the dispersed phase droplets is presented for pulsation intensities from 0 to 0.03 m/s using the Tenova kinetics internals (closed points) and the standard disc and doughnut internals (open points). It can be seen that d_{32} decreases as the pulsing rate increases for both sets of internals. The standard disc and doughnut internals, which were more hydrophobic, tended to form smaller droplets. This was most noticeable at lower pulsation intensity. The d_{32} values obtained with the Tenova kinetics internals are slightly larger than those with the standard disc and doughnut internals at low pulsation intensity. However at higher pulsation intensity, the mechanical energy dissipated for pulsing inside the column is strong enough for both internals to reach equivalent d_{32} values. From these results it can be concluded that the structure and surface property of the column internals are important factors for determining droplet diameters at low pulsation intensity, however as pulsation increases above 0.022 ± 0.002 m/s the mechanical energy dissipated to the droplets from the pulsation determines the droplet size. It should also be noted that the droplet size under no pulsing conditions for the Tenova kinetics internals was difficult to determine because the dispersed phase tended to accumulate on the internal plates and form flows instead of single droplets. Figure 11 also shows that there was no noticeable change in the sauter mean diameter with changes in dispersed and continuous phase velocities.

Figure 11. Effect of pulsation intensity on the sauter mean diameter d_{32} with different aqueous to organic phase ratio (Open points are standard disc and doughnut internals and closed points are Tenova kinetics internals)

4.5. Prediction of Sauter mean diameter and cumulative volume distribution

Most correlations in the literature are not suitable for the prediction of Sauter mean diameter under no pulsing conditions, with the exception of the equations presented by Van Delden et al.^[10, 11] and Wang et al.^[3]. Moreover, the correlation developed by Van Delden (refer to equation 16) was modified to fit the experimental data from this study via refitted constants shown in Table 5. Figure 12 provides a comparison between experimental data and predicted results

$$d_{32} = C_1 \varepsilon^{0.3} \left(\frac{\gamma}{\Delta \rho g} \right)^{0.5} \left(h \frac{\sqrt{\rho_* g}}{\gamma_*} \right)^{0.18} \left(\frac{\mu_d g^{0.25}}{\rho_*^{0.25} \gamma_*^{0.75}} \right)^{0.14} \left(\frac{\gamma}{\gamma_*} \right)^{0.06} \left[C_2 + \exp \left\{ C_3 \frac{Af}{\varepsilon \left(\frac{\gamma_* g}{\rho_*} \right)^{0.25}} \right\} \right] \left(\frac{v_d}{v_c} \right)^{0.118} \quad (16)$$

The refitted constants C_1 , C_2 and C_3 in equation 16 are provided in Table 5.

Table 5 Refitted constants for the correlation of sauter mean diameter d_{32}

Constants	C_1	C_2	C_3	AARD
Standard disc and doughnut internals	0.012	0.194	-2.59	13.2%
Tenova kinetics internals	0.023	0.051	-2.59	12.6%

Figure 12. Comparison between the experimental sauter mean diameter and predicted data

The average absolute relative deviation between the predicted Sauter mean diameter and experimental data with refitted constants are 13.2% and 12.6% for the standard disc and doughnut internals and the Tenova kinetics internals respectively.

The Weibull distribution has been used to examine the distribution of drop sizes with the column with each internal. The cumulative probability function (equation 17) with two parameters shown in table 6, a and b, has been used to correlate the large amount of experimental data.

$$F(d_i|a, b) = 1 - e^{-\left(\frac{d_i}{b}\right)^a} \quad (17)$$

Table 6 Regressed parameters a and b for Equation 17

Internals	Operating intensity (m/s)	a	b
Standard disc and doughnut internals	0.01	6.82	3.45
	0.02	6.84	2.45
Tenova kinetics internals	0.01	4.85	5.83
	0.02	4.64	2.98

Figure 13. Prediction of drop size cumulative volume distribution with the cumulative probability function

This equation defines the volume fraction of dispersed phase that is less than or equal to a certain value of droplet size. With correlated parameters, Figure 13 shows that the Weibull distribution equation can predict the experimental drop distribution data well. Additionally, Figure 13 shows that the cumulative drop distribution is becoming narrower as either the pulsation intensity increases or when the standard disc and doughnut internals are used instead of the Tenova kinetics internals.

5. Conclusions

The hydrodynamic performance of a pulsed solvent extraction column with standard disc and doughnut internals and novel Tenova kinetic internals has been investigated in a pilot scale column. Several conclusions can be drawn from this work and have been summarized as follows:

- a. With increasing pulsation intensity, the dispersed phase holdup decreased to a minimum value ($x_{d,min}$ at $(Af)_{min}$) and then increased with higher pulsation intensity for both types of internals. Conversely the characteristic velocity for both sets of internals initially increased until a maximum value ($V_{0,max}$) was reached, at the same values of $(Af)_{min}$.
- b. The dispersed phase holdup increased linearly with increasing dispersed phase velocity but there was no obvious effect of continuous phase velocity for both sets of internals.
- c. The dispersed phase holdup measured with the Tenova kinetics internals is lower than the standard disc and doughnut internals under the same operating conditions (including pulsation intensity, dispersed and continuous phase velocities).
- d. The Sauter mean diameter of the dispersed phase droplets decreased as pulsation intensity increased for both sets of internals, which is due to the breakage of dispersed phase droplets from the higher mechanical power input. At low pulsation rates (below $Af = 0.022$ m/s), the Sauter mean diameter for the Tenova kinetic internals was higher than the standard internals.
- e. There was no noticeable change in Sauter mean diameter with changes in dispersed and continuous phase velocities for both sets of internals.

- f. The Weibull distribution equation was suitable for predicting drop size volume distribution for both sets of internals.
- g. Correlations were presented for dispersed phase holdup, Sauter mean diameter and cumulative volume of dispersed phase. Regressed parameters were determined for the new internals which enabled good prediction of the pilot-scale experimental data. The average absolute relative errors were 6% for holdup and 14% for Sauter mean diameter.
- h. The Tenova pulsed column kinetics internals (TPC-KI) have lower holdup and larger droplet sizes for the same conditions indicates a large potential throughput.

6. Acknowledgements

The authors would like to acknowledge the funding provide by the Australian Research Council through Linkage grant LP130100305 and BHP Billiton, Olympic Dam, for this project, and would also like to thank the Particulate Fluids Processing Centre for the resources provided for this project.

7. Nomenclature

a	interfacial area, m^2
A	pulsation amplitude, m
c	solute concentration in liquid phase, M
d	drop equivalent sphere diameter, m
d_{32}	Sauter-mean drop diameter, m
d_e	equivalent drop diameter for elliptical drops, m
f	pulsation frequency, Hz
g	gravity acceleration, m/s^2
h	column height, m
L	flow ratio, viz velocity ratio of dispersed phase to continuous phase, dimensionless
V	Volume of the column, m^3
V_{disp}	dispersed phase volume in column, L

V_{active}	active section volume in column, L
v_d/v_c	dispersed phase velocity/continuous phase velocity, m/s
v_s	slip velocity, m/s
v_0	characteristic velocity, m/s
x_d	volume fraction holdup of dispersed phase, dimensionless
$x_{d,f}$	volume fraction holdup of dispersed phase at flooding, dimensionless
ε	the fractional free area, dimensionless
ϵ	drop distortion parameter, dimensionless
γ	interfacial tension, N/m
μ_d/μ_c	viscosity of dispersed phase/continuous phase, Pa·s
ρ_d/ρ_c	density of dispersed phase/continuous phase, kg/m ³
$\Delta\rho$	density difference between phases, kg/m ³
ψ	mechanical power dissipation per unit mass, W/kg
Subscripts or superscripts	
c	continuous phase
d	dispersed phase
in	an inlet to column
outlet	an outlet from column
*	equilibrium value

7. References

1. Rydberg, J., *Solvent extraction principles and practice, revised and expanded*. 2004: CRC Press.
2. Wang, Y., et al., *Dispersed-Phase Holdup and Characteristic Velocity in a Pulsed and Nonpulsed Disk-and-Doughnut Solvent Extraction Column*. Industrial & Engineering Chemistry Research, 2016. **55**(3): p. 714-721.
3. Wang, Y., et al., *Prediction of drop size in a pulsed and non-pulsed disc and doughnut solvent extraction column*. Chemical Engineering Research and Design, 2016. **109**: p. 667-674.
4. Wang, Y., et al., *Performance of an Industrial Pulsed Disc-and-Doughnut Extraction Column*. Solvent Extraction and Ion Exchange, 2016. **34**(2): p. 161-171.
5. Gayler, R. and H.R.C. Pratt, *Symposium on liquid-liquid extraction. Part II. Hold-up and pressure drop in packed columns*. Transactions of the Institution of Chemical Engineers, 1951. **29**: p. 110-125.

6. Kumar, A. and S. Hartland, *Correlations for dispersed phase hold-up in pulsed sieve-plate liquid-liquid extraction columns*. Chemical Engineering Research and Design, 1983. **61**: p. 248-52.
7. Kumar, A. and S. Hartland, *Prediction of dispersed phase hold-up in pulsed perforated-plate extraction columns*. Chemical Engineering and Processing: Process Intensification, 1988. **23**(1): p. 41-59.
8. Kumar, A. and S. Hartland, *Prediction of dispersed-phase holdup and flooding velocities in Karr reciprocating-plate extraction columns*. Industrial & engineering chemistry research, 1988. **27**(1): p. 131-138.
9. Kumar, A. and S. Hartland, *Unified Correlations for the Prediction of Drop Size in Liquid– Liquid Extraction Columns*. Industrial & engineering chemistry research, 1996. **35**(8): p. 2682-2695.
10. Van Delden, M., N. Kuipers, and A. De Haan, *Extraction of caprolactam with toluene in a pulsed disc and doughnut column—Part I: Recommendation of a model for hydraulic characteristics*. Solvent extraction and ion exchange, 2006. **24**(4): p. 499-517.
11. Van Delden, M., et al., *Extraction of caprolactam with toluene in a pulsed disc and doughnut column—part II: experimental evaluation of the hydraulic characteristics*. Solvent extraction and ion exchange, 2006. **24**(4): p. 519-538.
12. Kumar, A. and S. Hartland, *Prediction of drop size in pulsed perforated-plate extraction columns*. Chemical engineering communications, 1986. **44**(1-6): p. 163-182.
13. Torab-Mostaedi, M., A. Ghaemi, and M. Asadollahzadeh, *Flooding and drop size in a pulsed disc and doughnut extraction column*. Chemical Engineering Research and Design, 2011. **89**(12): p. 2742-2751.

TOC Graphic:

Minerva Access is the Institutional Repository of The University of Melbourne

Author/s:

Li, W;Wang, Y;Mumford, KA;Smith, KH;Stevens, GW

Title:

Comparison of the Hydrodynamic Performance of Pulsed Solvent Extraction Columns with Tenova Pulsed Column Kinetics Internals and Standard Disc and Doughnut Internals for Copper Extraction Using the LIX 84 System

Date:

2017-01-01

Citation:

Li, W., Wang, Y., Mumford, K. A., Smith, K. H. & Stevens, G. W. (2017). Comparison of the Hydrodynamic Performance of Pulsed Solvent Extraction Columns with Tenova Pulsed Column Kinetics Internals and Standard Disc and Doughnut Internals for Copper Extraction Using the LIX 84 System. SOLVENT EXTRACTION AND ION EXCHANGE, 35 (5), pp.303-320. <https://doi.org/10.1080/07366299.2017.1344487>.

Persistent Link:

<http://hdl.handle.net/11343/241466>