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ATALANTE 2012  
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Experimental and Modelling Studies of Mass Transfer in  
Centrifugal Contactors

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**Abstract**

Experimental trials have been undertaken to determine interfacial area for mass transfer in 5cm and 25cm diameter annular centrifugal contactors. All trials used 30% TBP in Exxsol D80 diluent, with extraction of HNO<sub>3</sub> being studied at varying flowrates and volumetric flow ratios. Droplet size was determined using a laser reflectance probe.

Results from the 5 cm rig showed two different regions of operation and the value of the dimensionless constant (K) in the Kolmogoroff expression was estimated to be 0.28 for O/A ratios below 2.5 and 0.30 above 2.5. From the 25cm rig trials K was estimated to be 0.26.

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Keywords: Annular Centrifugal Conactor; Area; Droplet Size; Mass Transfer; Solvent Extraction

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## 1. Introduction

It is anticipated that future nuclear reprocessing facilities will utilise centrifugal contactors for many of the solvent extraction operations. Optimisation of the design process will require reliable models able to predict contactor performance under a variety of operating conditions.

In order to predict the rate of mass transfer in a solvent extraction system it is necessary to have a knowledge of the mass transfer coefficient, distribution ratios of solutes between the two phases and of interfacial area. The distribution ratios are not specific to the type of contactor used and can be predicted from existing expressions. Correlations are also available for mass transfer coefficients under various conditions. However, the interfacial area for mass transfer is dependent on the power dissipated through mixing the phases. This is dependent on the type of contactor and in practice this must be obtained from experimental data which can be used to fit empirical correlations relating power density, fluid properties and droplet size. To provide the required data, trials on 5 and 25cm contactors were undertaken in the Netherlands at the University of Groningen and by CINC Solutions B.V at Denekamp, to measure the dispersed phase droplet size under different conditions.

### Nomenclature

$\Delta r_c$	width of contactor annulus (m)
$d_{32}$	Sauter mean diameter of droplet sizes (m)
$H_c$	height of liquor in annulus (m)
$j$	Reynolds number factor
$K$	dimensionless constant
$N$	rotor speed (revs/s)
$P$	energy dissipated in fluid (W)
$Re$	Reynolds number
$r_{Ro}$	outside radius of contactor rotor (m)
$\beta$	exponent in relative viscosity correlation
$\epsilon_d$	volumetric fraction of dispersed phase in annulus
$\Psi$	power dissipation per unit volume in the annulus ( $W/m^3$ )
$\rho$	liquid density ( $kg/m^3$ )
$\mu$	liquid (c:continuous phase, d: dispersed phase) viscosity, (Pa.s)
$\sigma$	interfacial tension (N/m)
$\omega$	rotational speed (radians/s)

## 2. Previous Work

The interfacial area for a mixed phase system can be calculated from the Sauter mean diameter of the dispersed phase droplets. Baird [1] proposed a Kolmogoroff type expression relating Sauter mean diameter to the power dissipated per unit volume through mixing in several different types of solvent extraction contactor.

$$d_{32} = \frac{0.6 K \sigma}{\Psi^{0.4} \rho^{0.2}} \quad (1)$$

Baird reported that the dimensionless constant (K) was likely to be in the range of 0.18 - 0.36 regardless of the means of agitation.

The annular centrifugal contactor supplied by CINC Solutions B.V was developed at the Argonne and Oak Ridge National Laboratories in the USA. During its development, a model was derived by Arafat et al [2] which predicted the power dissipated in the annulus of the contactor. This value can then be divided by the liquor volume in the annulus to give the power dissipated per unit volume term which is used in equation (1).

$$P = 0.0261 H_c r_{Ro}^{3.75} (j\omega)^{2.75} \rho^{0.75} \left( \frac{\mu}{\Delta r_c} \right)^{0.25} \quad (2)$$

where

$$j = 0.0554 (\log_{10} Re) + 1.368 \quad (3)$$

The fluid density in equations (1) and (2) refers to an average value of the dispersed and continuous phases and the same approach can be used for fluid viscosity, though section 4.1 describes the use of a mixture viscosity which can be used as an alternative to average viscosity. Hence the measurements of droplet size from the current trials can be introduced to the Kolmogoroff expression (1) along with the calculated power dissipation to derive a value for the dimensionless constant (K).

Recent work has proposed alternative correlations for power dissipation and droplet size. Kadam et al [6] have conducted trials where effective interfacial area was calculated using a chemical method involving the hydrolysis of dodecyl formate with sodium hydroxide. Assuming spherical droplets the expression for Sauter mean diameter is given as:

$$d_{32} = \frac{0.0085 \sigma^{0.6}}{\Psi^{0.21} \rho^{0.17}} \left( \frac{\mu_d}{\mu_c} \right)^{0.1} (1 + 8.5 \varepsilon_d) \quad (4)$$

while the power dissipated in the annular zone is given as:

$$\left( \frac{P}{N^3 r_{Ro}^5 \rho} \right) = 41.55 \left( \frac{N r_{Ro}^2 \rho}{\mu} \right)^{-0.5} \left( \frac{\Delta r_c}{r_{Ro}} \right)^{-0.066} \left( \frac{H_c}{r_{Ro}} \right)^{0.84} \quad (5)$$

The power dissipation trials were conducted in a custom-built rig designed to represent an annular centrifugal contactor with an acrylic solid rod rotating in an acrylic stationary cylinder. A similar expression for Sauter mean diameter was also proposed [6], where the model was based on droplet size measurements made with a Coulter-counter. However the expression in equation 4 shows a better approximation to the current data and is compared in section 4 with values predicted by equation (1).

### 3. Experimental set-up and procedure

Two different rigs were used to measure the droplet size using the same technique. The aqueous feed stream used was 4M nitric acid and the organic stream was 30% v/v TBP in odourless kerosene. The flow diagram of the rig for the 5cm contactor is shown in figure 1 below showing the vessels pumps and measurements along with the CS50 contactor.

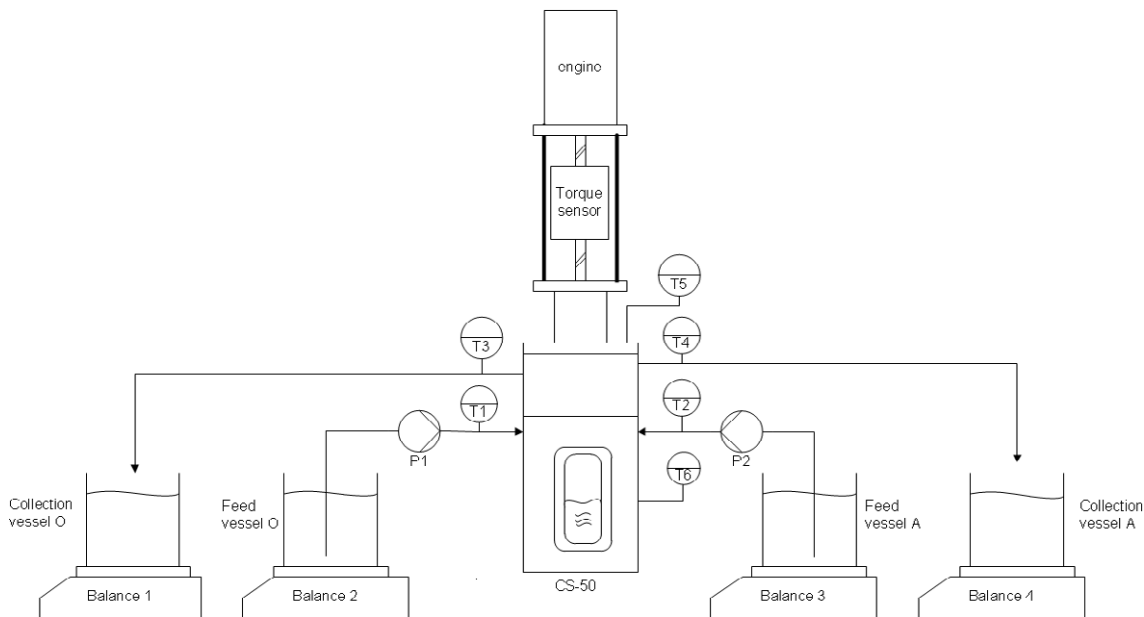


Fig. 1. Flow Diagram of 5cm Centrifugal Contactor Rig

The annular centrifugal contactors were manufactured by CINC Solutions with a transparent window and a number of different ports in the front of the contactor so that a laser reflectance probe could be used to measure the droplet size distribution in the annulus. A number of runs were carried out under organic:aqueous flow ratios. Each run was allowed to progress until the Sauter mean diameter had reached steady state, the steady state values were then used in the model for parameter estimation. In addition to droplet size, on the 5cm rig measurements were taken of power input, annular liquor volume, temperature and of the acidity of the aqueous feed and outlet streams.

The trials were conducted at ambient temperatures with no temperature control, though the temperatures of the liquor streams, mixing zone, contactor housing and room were all monitored during the 5cm trials to ensure no significant temperature changes over the course of each experiment. The mixing zone temperature on the 5cm trials was within the range of 21-23.5°C taken as an average over the course of each run.

The outlet acidity measurements were taken to compare with the model predictions. Since the rate of mass transfer is dependent on droplet size, the comparison would provide some degree of validation of the model.

#### 4. Results & Discussion

Data from the two rigs were applied to a model of the contactors developed at the National Nuclear Laboratory (NNL) in the UK. The model was parameterized for each rig by using the appropriate physical dimensions.

##### 4.1. 5cm rig

Trials were conducted on the 5cm contactor running at 2860 rpm with total flowrates of 500, 750, and 1000 ml/min and a variety of phase ratios for each flowrate. The results are shown in figure 2 below:

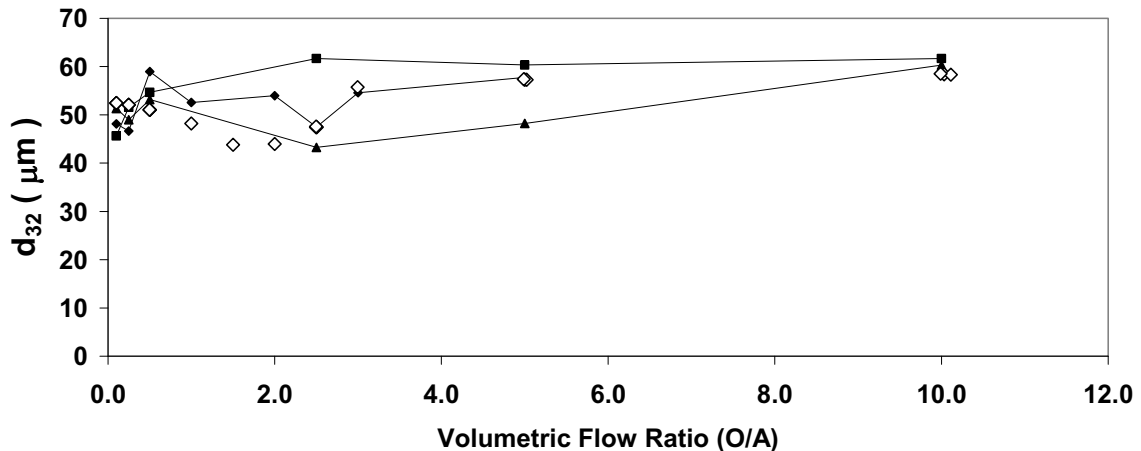


Fig. 2. Effect of volumetric flow ratio on Sauter mean diameter for rig and model with model fit on two different regimes.  
 ◆ Rig 750ml/min ■ Rig 500ml/min ▲ Rig 1000ml/min ◇ Model (results were independent of total flowrate)

From figure 2 it appears that there is a change in rig behaviour with increasing O/A ratio. Sensitivity of droplet size to total flow rate increases as the O/A ratio is increased towards a value of 2.5 thereafter decreasing as O/A ratio is further increased to 10. Results at low O/A ratios show a greater amount of ‘scatter’. It is assumed that this difference in behaviour is due to a change in the continuous phase, with the aqueous phase being continuous below an O/A ratio of 2.5 and the organic phase being continuous above 2.5. Hence the initial approach was to fit two different values of the dimensionless constant (K in equation 1), with a value of  $0.28 \pm 0.02$  for ratios below 2.5 and  $0.30 \pm 0.03$  for ratios above 2.5. When parameter estimation was carried out on the whole data set, the value of  $0.29 \pm 0.06$  was obtained for the dimensionless constant. The results in figure 2 correspond to the values of  $K=0.28$  and  $K=0.30$  for O/A ratios less than and greater than 2.5 respectively. These are considered the best fit as the range of uncertainty corresponding to the 95% confidence limits is less than that in the fit to the whole data set. It should be noted however, that the range of uncertainty spans the values of K for both regimes, hence one value of K might be appropriate to represent the whole data set if variation in the droplet size data could be reduced.

There was also a different method used for calculation of the fluid viscosity in the different operating regimes as used by Padiyal-Collins et al [3] based on the approach of Cheng & Law [4]. Above an O/A ratio of 2.5 and below an O/A ratio of 1 the viscosity of the fluid is calculated as an effective viscosity using an exponential function of the phase volume fractions. However in the region of  $1 < O/A < 2.5$  the same exponential term is used for effective viscosity but the dispersed phase volume fraction is ‘capped’ at 0.486 (OA=1.058) which was found to give the best fit to the data.

Use of the effective viscosity of the mixture incorporates an effective viscosity term ( $\mu_{eff}$ ) derived from relative viscosity ( $\mu_r$ ) and continuous phase viscosity ( $\mu_c$ ). The value of  $\beta$  used to fit the experimental data was 0.95.

$$\mu_{mixture} = \varepsilon_{aq} \mu_{eff}^{aq} + \varepsilon_{org} \mu_{eff}^{org} \quad (6)$$

$$\mu_r = \frac{\mu_{eff}}{\mu_c} \quad (7)$$

$$\mu_r = \exp\left[\left(\frac{2.5}{\beta}\right)\left(\frac{1}{(1 - \varepsilon_d)^\beta} - 1\right)\right] \quad (8)$$

Use of the effective viscosity prediction in equation 2 gives much higher viscosities around the point where phase inversion is assumed to occur and gives a better fit to the data. There is some empirical evidence that this phenomenon has been observed previously on small scale contactors with Leonard et al. [5] reporting an increase in viscosity on approaching the phase inversion point on 2 cm annular centrifugal contactors.

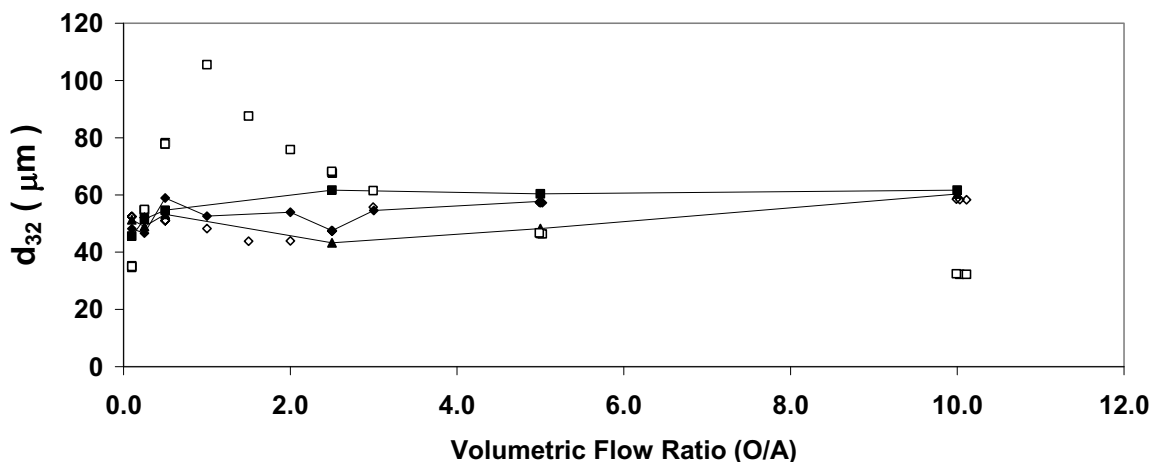


Fig. 3. Comparison of the current model with the model developed by Kadam et al .

◆ Rig 750ml/min ■ Rig 500ml/min ▲ Rig 1000ml/min □ Kadam et al ◇ model-current work

The predicted droplet size from figure 2 is compared in figure 3 to the values predicted by the model developed by Kadam et al [6]. Figure 3 shows that there is much more variation with changing volumetric flow ratio in the model of Kadam et al. than that observed in the current model. This is due to the inclusion of the fractional volumetric holdup term in equation (4). It should be noted however, that the model of Kadam et al shows 2 different operating regimes with a transition at the phase inversion point (in this case assumed to occur where O/A =1), and this is without using the mixture viscosity term defined in equation 6. It is around the phase inversion point that the predictions deviate most from the current data, and since this is the region where the mixture viscosity is most relevant, consideration of the mixture viscosity in the derivation of equations (4) and (5) may make the model developed by Kadam et al more relevant to the current data. Equation (5) for power dissipated in the annulus was developed using a single phase system and the high mixture viscosities developed around the phase inversion point are outside its range of validity (25mPa.s for maximum mixture viscosity against 10mPa.s for upper limit of correlation).

The model predictions of acidity of the aqueous outlet stream are compared against the rig measurements and figure 4 below shows that the model predicts higher acidities than the rig data. The calculated acidities assuming an equilibrium contactor are also shown on the plot and it is evident that the extent of mass transfer required to obtain the acidities shown in the rig data would be greater than that represented by the equilibrium values. Hence there are obviously some significant errors in the rig acidity data and it is not possible to use it for validation of the model. The model predicts stage efficiencies ranging between 79 - 94% for the runs on the 5cm contactor.

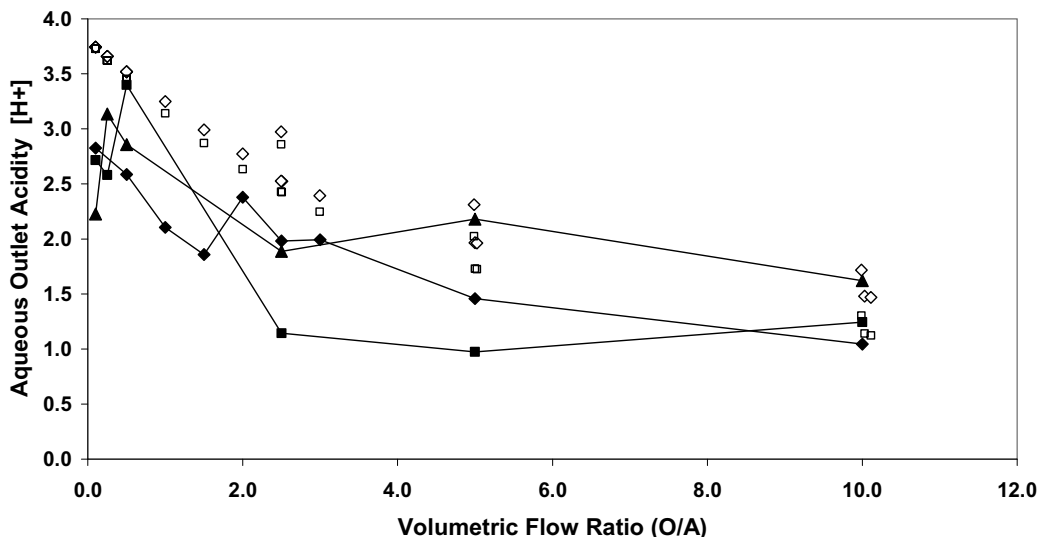


Fig. 4. Acidity of Aqueous Outlet Stream on 5cm Rig Compared with Model Predictions

◆ 750ml ■ 500ml ▲ 1000ml ◇ Model all flows □ Equilibrium

#### 4.2. 25cm rig

The increase in scale from the 5cm to the 25 cm contactor imposed restrictions on the volume of reagents available for testing. Hence it was not possible to repeat the same run schedule as used on the 5cm contactor. The rig runs in figure 5 were carried out at a rotational speed of 1080 rpm with organic/aqueous ratios greater than 2. Hence it was assumed that the solvent formed the continuous phase. The total volumetric flow ranged from 15-50 litres/min, changing the total flow with each change in flow ratio, so it is evident that the change in total flow did not produce the same variation in droplet size as observed on the 5cm rig. The dimensionless constant obtained from the 5cm rig over-estimated the droplet size on the 25cm rig and so it was necessary to re-fit the model. This resulted in a dimensionless constant of  $0.262 \pm 0.008$ . The stage efficiencies predicted by the model for the 25cm rig ranged between 85-93%.

On the 5 cm rig it was assumed that an O/A ratio of 2.5 was in the transition region between aqueous continuous and organic continuous and the fluid viscosity was calculated accordingly. On the 25 cm rig there are two data points at an O/A ratio of two which appear to be consistent with other data points in the organic continuous region, hence their viscosity is calculated on this basis.

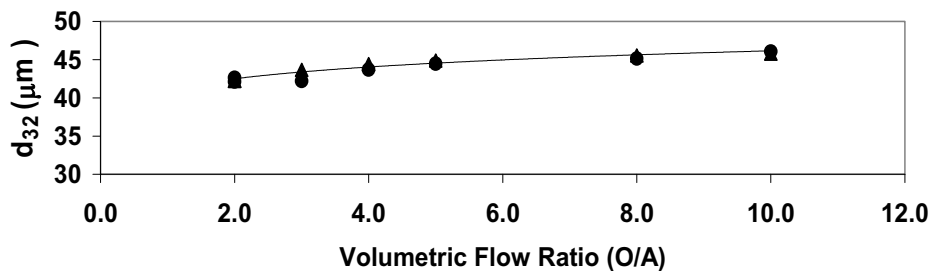


Fig. 5. Effect of Volumetric Flow Ratio on Sauter mean diameter for Rig (▲) and Model (●)



The fact that variation in total volumetric flow did not change the droplet size on the 25cm rig is encouraging since equations (1) & (2) do not predict any relationship to total flow. This is evident in figure 2 where the model predictions of droplet size on the 5cm rig at different total flowrates (organic flow + aqueous flow) appear as one point and vary only with changing flow ratio, where the rig data shows significant variation with total flowrate. It is likely that the relationship between droplet size and total flow observed on the 5cm rig is due to variation in the droplet size and flow measurements associated with the smaller annular volume.

## 5. Conclusions

Values of the dimensionless constant (K) in the Kolmogoroff expression for droplet size ranged between 0.26 and 0.30 for the annular centrifugal contactors used in the trials, dependent on size of contactor and which phase was continuous.

Scale-up of interfacial area from the 5cm to the 25cm annular centrifugal contactors is now feasible using the model developed over the course of this work.

Further validation of the model is required to establish confidence in the predicted rate of mass-transfer, this should include extraction trials with sampling to calculate stage efficiency.

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