Mass Transfer Performance of a Rotating Spiral: Comparison with Conventional Contacting Methods

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Abstract **Previous work by the authors develops a novel technology for counter-current contacting of immiscible fluid phases based on a rotating spiral channel. The work has established theoretical modelling allowing prediction of mass transfer rates for physical absorption and desorption of dilute solute species and has verified the main hydrodynamic elements of the theory, with experimental measurements over a wide range of flow rates and liquid viscosity. A key feature of rotating spiral contacting is that the relative thicknesses of the phase layers is constant throughout the contacting process and, along with the relative phase flow rates, can be varied regardless of phase and solute properties to produce optimum conditions for the contacting. It has been shown that there are two criteria that determine the optimum. First, the flow rate ratio of the phases is matched to the equilibrium distribution of the solute between the two phases, so that sufficient solvent phase flows to allow full removal of solute from the processed stream. Second, the relative layer thicknesses should maximise specific throughput, i.e. the flow rate of the processed stream per device volume. These ideas of optimum contacting guide a comparison here of recent mass transfer data for the spiral with corresponding literature data for the packed column and rotating packed bed.**

Index Terms: **rotating spiral; absorption; desorption; mass transfer; optimum; counter-current.**

I. INTRODUCTION

Recent work [1-3] has established the fundamentals of a new technology for immiscible fluid phase contacting using a rotating spiral channel. The rotation introduces centrifugal body force to drive the fluids and to maintain segregation of the phases so they flow in parallel layers in the spiral channel. The spiral enables the driving body force to be maintained constant with radius, since as radial position increases the angle of the channel relative to the azimuthal direction decreases such that the component of body force along the channel remains constant.

Appropriate selection of pressure gradient allows the phases to flow counter-currently, which is essential for effective mass transfer. Changing rotation rate and pressure gradient enable continuous adjustment of the ratios of phase flow rates and layer thicknesses. These are arguably the two most critical parameters determining contacting effectiveness. Thus, the rotating spiral may be applied to solute transfer from one fluid phase to another to achieve optimum contacting for the full range of separation processes involving phase contacting: distillation, absorption, desorption and liquid extraction.

The rotating spiral clearly has advantages over conventional fluid phase contacting approaches and the purpose of the present work is to consider how comparisons between different approaches should be made and to make preliminary comparisons. Attention is restricted to the gas-liquid contacting operations of absorption and desorption. Two conventional contacting methods are considered in the comparisons: packed columns and rotating packed beds. The packed column is in widespread use and the rotating packed bed has received considerable attention in the past several decades [4, 5]. Both approaches use body force to drive the fluid phases counter-currently through a packing that breaks one phase into small elements travelling through the other phase. This increases mass transfer effectiveness but brings unwanted problems, including entrainment of small droplets by the gas phase, uneven distribution of the phases, flooding and the requirement of phase separation when contacting is complete. The spiral avoids all of these and, uniquely, allows operation at optimum effectiveness over the full range of possible phase and solute properties [2] that may be encountered in practice.

Comparison of different approaches requires careful consideration so that observed differences are not due to the particular conditions of operation of compared results. So the first task here is to develop the framework to be used. Data for the rotating spiral comes from current work that builds on the hydrodynamic study of MacInnes and Zambri [3] in a 1.5 mm channel by introducing mass transfer measurements for a range of different solute molecules desorbing at dilute concentrations from water into air. These data are as yet unpublished, but some of the data for acetone and for ethanol from that study are used here. Evidence of the effect of channel size is available from the binary stripping distillation results for a 0.32 mm rotating spiral channel in MacInnes et al. [1], using the computed values

Received 31 December 2016; revised 24 January 2017; accepted 25 January 2017.

Available online 27 January 2017.

of mass transfer coefficients in Ortiz-Osorio et al. [6]. Data for the packed column comes from the absorption experiments with acetone and ethanol of Houston and Walker [7] and that for the rotating packed bed comes from the experiments for absorption of ethanol of Chiang et al. [8]. Comparison of absorption and desorption results is possible by avoiding the usual reference to 'gas' and 'liquid' phases in favour of the phase being 'cleaned' of solute and the 'solvent' phase used to do the cleaning.

II. BASIS FOR COMPARISON

The analysis now presented gives the derivation of the relations for overall mass transfer coefficient and flow rate per passage volume (specific throughput) that can be used to compare contacting for different devices and under different conditions. Each one depends on the mole flow rate ratio of the phases, q_S, and the slope of the equilibrium curve, f_S', defined in each case as the ratio of the solvent phase quantity to that of the cleaned phase:

$$q_S = \frac{n_S u_{SB}(1-\xi_C)}{n_C u_{CB} \xi_C} \tag{1}$$

$$f_S' = \frac{Y_S}{Y_C} \tag{2}$$

The mole flow rate ratio, which is a negative quantity for counter-current flow, is expressed in terms of the mole density, n, and the bulk velocity, u_B, in each phase, where subscripts '*S*' and '*C*' signify the solvent and the cleaned phase, respectively. ξ_C is the fraction of the flow passage occupied by the cleaned phase. The slope of the equilibrium curve for dilute solute (Eq. 2) is just the ratio of the mole fractions in the solvent and cleaned phases, Y_S and Y_C. These two parameters the most important ones for understanding phase contacting.

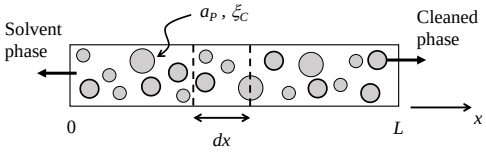

Figure 1. Counter-Current Contacting Arrangement Where Solute Transfers From the 'Cleaned' Phase to the 'Solvent' Phase.

With contacting in the x direction as shown in Figure. 1, the differential equation for solute conservation in the cleaned phase and the balance equation enforcing overall solute concentration are

$$n_C u_{CB} \xi_C dY_{CB} = K_C a_P \left(Y_{CB}^* - Y_{CB}\right) dx \tag{3}$$

$$Y_{SB}(x) = Y_{SB}(0) - \frac{Y_{CB}(x) - Y_{CB}(0)}{q_S} \tag{4}$$

K_C is the overall mass transfer coefficient based on the cleaned phase variables and $Y_{CB}^* = Y_{SB} / f_S'$. The specific interface surface area, a_P, is per unit volume of passage. That is, it does not include the volume of the solid packing and the usual values based on container volume, a, must be adjusted by the packing void fraction, ε, using $a_P = a/\varepsilon$. If Figure. 1 depicts a dispersed liquid phase then this is desorption. If absorption was occurring with the circumstances otherwise unchanged, then the gas would become the cleaned phase with the liquid the solvent and the x coordinate axis would be to the left (in the direction of cleaned phase flow). Eq. 4 can be used to determine a critical ratio of flow rates of the phases below which complete separation is not possible and Eq. 3 can then be used to demonstrate the relationship between purity of separation and non-dimensional independent parameters. First, evaluating Eq. 4 at $x = L$ and placing the result in terms of the purity of the outlet cleaned stream, c_C, and of the solvent inlet stream, c_S, leads to the relation:

$$-q_S > \frac{1-c_C}{f' - c_S} \tag{5}$$

The purity parameters, c_C and c_S are ratios of inlet and outlet solute mole fractions:

$$c_C = Y_{CB}(L)/Y_{CB}(0) \; c_S = Y_{SB}(L)/Y_{CB}(0) \tag{6}$$

When the inlet solvent stream is pure ($c_S = 0$) and the cleaned stream exits with only a minor amount of the initial solute ($c_S \to 0$) Eq. 5 takes a simple form,

$$-f_S' q_S > 1 \tag{7}$$

For a given phase and solute system, f_S' is fixed and from Eq. 7 the flow rate ratio must be greater than the value $-q_S = 1/f_S'$ for effective separation to be possible. If it is not large enough the solvent stream flow will be insufficient, even were equilibrium reached, to hold all of the solvent in the inlet flow. The situation is further clarified by integrating Eq. 3 over the length of contacting to form a relation for the purification, c_C, that is achieved. Variables may be separated and K_C, a_P, n_C, u_{CB} and ξ_C are constant to excellent approximation for packed columns and rotating spiral channels. Using Eqs. 1, 2, 4 and 6 allows the result to be expressed as:

$$c_C = \frac{q_S c_S + (1 - q_S c_S + f_S' q_S)(-f_S' q_S)^{-\Phi_C t_{mC}}}{f_S' q_S + (-f_S' q_S)^{-\Phi_C t_{mC}}} \tag{8}$$

This is the purification of the cleaned stream as a function the flow rate ratio, the slope of the equilibrium curve, the purity of the inlet solvent and the two further parameters which emerge from the solution:

$$\Phi_C = \frac{K_C a_P}{n_C} \frac{\left(1+ f_S' q_S\right)}{f_S' q_S \ln\left(- f_S' q_S\right)} \tag{9}$$

$$t_{mC} = \frac{L}{u_{CB}\xi_C} \tag{10}$$

The first parameter is the specific throughput based on the passage volume required to achieve one equilibrium stage, i.e. the volume flow rate of the cleaned phase per volume of passage corresponding the equilibrium length, ℓ_e:

$$\ell_e = \frac{n_C u_{CB}\xi_C}{K_C a_P} \frac{f_S' q_S \ln\left(- f_S' q_S\right)}{\left(1+ f_S' q_S\right)} \tag{11}$$

The dependence on the overall mass transfer coefficient and specific interface area in Eq. 9 makes it clear that this parameter is a measure of the effectiveness of contacting. The second parameter (Eq. 10) is a mean residence time for the cleaned phase and represents the effect on purification of longer contact time. Eq. 8 is plotted in Figure. 2 for different values of $\Phi_C t_{mC}$, in the case where the solvent contains no solute at inlet. The horizontal axis is the remaining parameter in the equation which may be thought of as the relative flow rate ratio, $- f_S' q_S$, i.e. the flow rate ratio relative to the slope of the solute phase equilibrium curve, f_S'.

Since $c_C = 0$ corresponds to complete removal of the solute from the cleaned phase, it is clear that $- f_S' q_S$ should be greater than unity as Eq. 7 prescribes. Further, the improving purification with increasing $\Phi_C t_{mC}$, by either increasing specific throughput or increasing residence time, is as expected. The shaded zone represents the approximate region where economically optimum operation is likely to be found. Relative flow rate ratios greater than 2.5 must be considered exceedingly wasteful of solvent, values of purification greater than $c_C = 0.2$ are in any case probably not difficult and purification penalty is severe below $- f_S' q_S$ equal to unity.

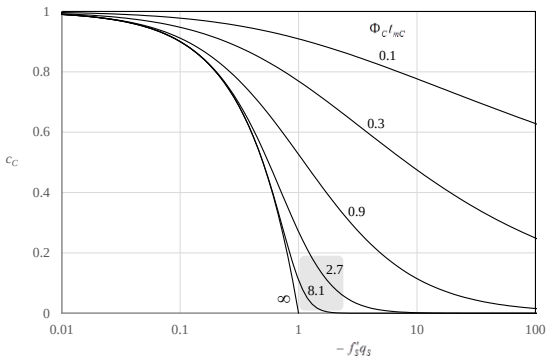

Figure 2. The General Purification Function for Different Values of the Contacting Effectiveness, $\Phi_C t_{mC}$. The Shaded are Shows the Region where Economic Optimum is Likely to Fall.

III. DATA SOURCES AND ANALYSIS

Data for absorption or desorption of acetone or ethanol in the water-air phase system is found in the literature for both packed columns and rotating packed beds and this allows comparison with desorption data available for the rotating spiral channel for these same solutes.

Houston and Walker [7] give overall mass transfer coefficient data for both acetone and ethanol absorption in a column packed with 1" Raschig rings. Chiang et al. [8] give the same for ethanol absorption in a rotating packed bed with 0.22 mm diameter wire mesh packing. The determination of the mass transfer coefficient values uses the conventional relation, which is just Eq. 8 rearranged to show $K_C a_P$ as an explicit function of the measured inlet and outlet solute mole fractions of the phases:

$$\frac{K_C a_P L}{n_C u_{CB}\xi_C} = \frac{Y_{CB}(L) - Y_{CB}(0)}{\overline{\Delta Y_{CB}}} \tag{12}$$

where

$$\overline{\Delta Y_{CB}} = \frac{\Delta Y_{CB}(L) - \Delta Y_{CB}(0)}{\ln\left[\Delta Y_{CB}(L)/\Delta Y_{CB}(0)\right]} \tag{13}$$

with

$$\Delta Y_{CB} = Y_{CB}^* - Y_{CB} \tag{14}$$

Where $K_S a_P$ is reported, $K_C a_P$ can be determined using the identity $K_S = K_C / f_S'$. For rotating packed beds the flow section area varies in the contacting directions and the equations must be modified. Calculated values of mass transfer from [8] take the area change into account and Eq. 9 is otherwise unchanged.

Data for the rotating spiral channel contacting comes from the apparatus described in MacInnes and Zambri [3] for desorption of acetone and ethanol at 3200 rpm. The channel section is 1.5 mm by 4.0 mm with phase layers flowing parallel to the longer side. A further set of data is available for the spiral from the stripping distillation experiment of MacInnes et al. [1]. The channel in that case is much smaller, 0.32 mm by 0.11 mm (layers parallel to the shorter side). These data allow the h^{-2} [2] scaling of specific throughput (where h is the channel dimension perpendicular to the layers) to be demonstrated. The value of f_S' is taken as the average value and the mass transfer coefficient at the average mole fraction for the range of bulk mole fraction in the stripping distillation. Values of $K_C a$ are converted to $K_C a_P$ using $a_P = a/\varepsilon$. Table 1 summarises the data sets and lists some relevant parameters. The specific area values are for packing surface area for columns and beds but are for true phase interface area for the rotating spiral channels.

IV. RESULTS

To allow both absorption and desorption to be compared on a common basis, two adjustments need to be made. First, the large density difference between gas phase and liquid phase means the volume flow rate of cleaned phase in the specific throughput, Φ_C, will be far larger for absorption than for desorption. This can be adjusted by multiplication by the mole density of the cleaned phase, so specific throughput is on a mole flow rate basis. Secondly, when comparing absorption and desorption the constraint on flow rate ratio, Eq. 7, shows that the absorption specific throughput will be greater than that for desorption when f'_S is less then unity and the reverse when greater than unity. This can be adjusted by multiplying the specific throughput by f'_S in cases of absorption to place it on the same basis as desorption results. Thus, for absorption $f'_S n_C \Phi_C$ is plotted and for desorption $n_C \Phi_C$.

Figure. 3 shows the results of specific throughput for acetone transfer with the different contactors, where data is available. As expected, the rotating spiral gives highest specific throughput since a larger specific surface area is achieved (Table 1), by about a factor of 10 in the useful range of $f'_S q_S$. The data for the packed column at values of $-f'_S q_S < 1$ will not allow significant absorption of the acetone (Figure. 2). Data from all three contactor types is available for ethanol transfer and these are collected in Figure. 4. Again, the rotating spiral gives about a factor of 10 larger specific throughputs.

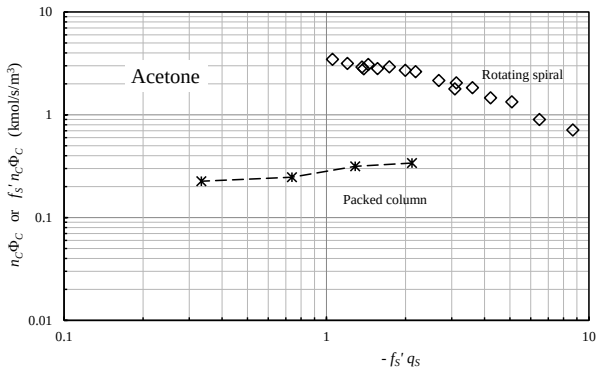

Figure 3. Comparison of Specific Throughput for Contacting with a Rotating Spiral and with a Packed Column. Absorption/Desorption of Acetone for the Water-air System.

Table 1. Data Sets and Essential Parameter Values

Contactor	a_P	ε	Mode	Solute	f'
Packed column [7]	260 m^{-1}	0.72	Absorption	Acetone Ethanol	2.50 0.33
Rotating packed bed [8]	1080 m^{-1}	0.94	Absorption	Ethanol	0.42
Rotating spiral	983 m^{-1}	NA	Desorption	Acetone Ethanol	2.50 0.46
Rotating spiral [1]	5760 m^{-1}	NA	Distillation	C_5H_{10} and C_6H_{14}	1.10

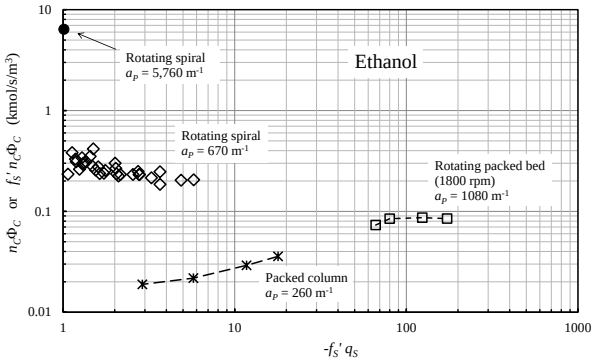

Figure 4. Comparison of Specific Throughput for Contacting with a Rotating Spiral and with a Packed Column. Absorption/Desorption of Ethanol For the Water-air System; Solid Symbols for Stripping Distillation of 2,2-Dimethylbutane and 2-Methyl-2-Butene [1].

Curiously, the packed column data in Figure. 4 does not extend into the optimum region of $-f'_S q_S$. The packed column data are shifted to higher $-f'_S q_S$ relative the acetone data by an amount suggesting q_S has not changed. And since the lower limit corresponds to lowest liquid flow rate (this is varied in the experiments) it may be that liquid distribution in the column becomes poor below this rate.

The rotating packed bed results are entirely in the wrong region of $-f'_S q_S$ and also show a specific throughput that is only about 30% of that achieved by the rotating spiral. The distillation result using smaller spiral channel size shows the expected increase in specific throughput. The small and large channel specific throughput values should be approximately in the ratio of inverse squared channel sizes, i.e. $(1.5/0.32)^2 = 22$, close to what is observed.

CONCLUSIONS

Transfer of acetone and ethanol in the water-air system has been used to compare mass transfer performance of rotating spiral channels to conventional packed column and rotating packed bed contactors. A common basis for comparison is developed and this is applied to data for both absorption and desorption over the range of contactor types. The rotating spiral is able to operate in

the appropriate range of relative phase flow rate ratio and gives the highest specific throughput for both solutes tested: a factor of 3 better than the rotating packed bed and a factor of 10 better than the packed column. Further increase in specific throughput is produced by decreasing channel size, as predicted.

ACKNOWLEDGEMENT

We thank the Libyan State for a Scholarship (NZ071-538-21368) supporting MKSZ and the Iraqi Government for an HCED Scholarship (D 11-3508) supporting AAA.

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BIOGRAPHIES

Dr. Jordan MacInnes a senior lecturer has joined the Chemical Engineering Department at the University of Sheffield, UK since 1993. His recent work has aimed to address fundamental technical issues within the emerging area of micro fluidics, including those of mixing and flow control. He received BS and MS degrees in Mechanical Engineering from the University of California Davis, and a PhD degree from University of Manchester Institute of Science and Technology.

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