GAS-LIQUID MASS TRANSFER: A COMPARISON OF DOWN- AND UP-PUMPING AXIAL FLOW IMPELLERS WITH RADIAL IMPELLERS

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ABSTRACT

The performance of a down- and up-pumping pitched blade turbine and A315 for gas-liquid dispersion and mass transfer was evaluated and then compared with that of Rushton and Scaba turbines in a small laboratory scale vessel. The results show that when the axial flow impellers are operated in the up-pumping mode, the overall performance is largely improved compared with the down-pumping configuration. Compared with the radial turbines, the up-pumping A315 has a high gas handling capacity, equivalent to the Scaba turbine and is economically much more efficient in terms of mass transfer than both turbines. On the other hand, the up-pumping pitched blade turbine is not as well adapted to such applications. Finally, the axial flow impellers in the down-pumping mode have the lowest performance of all the impellers studied, although the A315 is preferred of the pitched blade turbine.

Keywords: mixing; stirred tank; gas-liquid; mass transfer; gas holdup; up-pumping; axial flow impeller

INTRODUCTION

Fermentation, wastewater aeration, oxidation and hydrogenation represent only a small number of the complex mixing processes where gas dispersion is employed in mechanically agitated tanks. Traditionally, gas dispersion in agitated vessels is carried out using radial disc turbines, such as the Rushton turbine. However, over the last 10-15 years, an increased interest in axial flow impellers for gas-liquid applications has evolved in search of overcoming some of the weaknesses of disc turbines (eg. high power number, limited gas handling capacity, poor top-to-bottom mixing in multi-stage systems ... a complete description is given by McFarlane et al., 1995 and Nienow, 1996). Customarily, axial impellers are used in the down-pumping mode. In this operating mode, axial flow impellers have provided significant advantages over radial flow agitators, including a low unaerated power number and improved top-to-bottom mixing in multi-impeller vessels (McFarlane and Nienow, 1995; 1996). However, upon aeration, these impellers cause torque and flow instabilities (Nienow et al., 1983; Chapman et al., 1983; Bujalski et al., 1988; McFarlane and Nienow, 1996), which may lead to excessive vessel vibrations (Nienow, 1996). More recently, up-pumping axial flow impellers, which were first studied in the early 1980s (Kuboi and Nienow, 1982; Nienow et al., 1983), have been shown to provide further advantages over disc turbines and down-pumping axial impellers (Nienow, 1990a; 1990b; 1996), particularly for gas-liquid mixing. In single phase applications, the single up-pumping agitator generates two large circulation loops, which provide a higher circulation rate of liquid in the vessel and therefore leading to improved turnover and reduced dead zones

(Mishra *et al.*, 1998; Aubin *et al.*, 2001). Moreover, power numbers remain significantly lower than those of Rushton turbines (Nienow, 1996; Ozcan-Taskin *et al.*, 1996; Hari-Prajitno *et al.*, 1998) and mixing times are reduced significantly (Hari-Prajitno *et al.*, 1998; Aubin, 2001). Upon gassing, the problems of flow and torque fluctuations are totally eliminated and power loss is minimal ($P_g/P \sim 1$ for varying gas flow rates), which means that adequate gas dispersion is possible even at low impeller speeds (Nienow, 1996). In addition, a recent study has shown that the global gas holdup in a vessel stirred by an up-pumping pitched blade turbine is significantly greater than for the down-pumping configuration (Aubin *et al.*, 2004b). This suggests that by using up-pumping axial flow impellers, the mass transfer potential could be possibly improved due to a potential increase in the volumetric mass transfer coefficient.

Although several studies have dealt with up-pumping axial flow impellers and have highlighted the advantages of these agitators over others for both single- and multi-phase applications, these investigations have been focussed mainly on mixing time and power characteristics (McFarlane, 1991; Hari-Prajitno *et al.*, 1998; Kuzmanic and Ljubicic, 2001, or on the measurement (Mishra *et al.*, 1998; Aubin *et al.*, 2001; 2003a; Ozcan-Taskin and Wei, 2003) or prediction of turbulent flow fields (Jaworski *et al.*, 1998; 2001; Aubin *et al.*, 2003b; Ozcan-Taskin and Wei, 2003). To the authors' knowledge, it appears that there has been no attempt to characterise the performance of up-pumping axial flow impellers with respect to mass transfer potential, by measurement of the volumetric mass transfer coefficient.

In the present work, the performances of two axial flow impellers (pitched blade turbine and A315) in both the down- and up-pumping configuration are quantified in terms of the volumetric mass transfer coefficient, the overall transfer efficiency and the standard oxygen transfer efficiency, as well as the overall gas holdup and the power dissipation. These characteristics are then compared with those of two radial impellers, a Rushton turbine and a Scaba 6SRGT.

EXPERIMENTAL METHODS

Equipment & Experimental Conditions

The experiments were performed in a dished-bottom cylindrical vessel ($D_T = 0.19$ m) with an aspect ratio of 1, i.e. the liquid height (*H*) in the vessel was equal to the tank diameter (D_T). The tank was equipped with four baffles ($b = D_T/10$), which were placed 90° from one another, flush against the vessel wall. The impeller clearance was $C = D_T/3$, where *C* is defined as the distance from the vessel bottom to the lowest horizontal plane swept by the impeller. The performances of two axial impellers were studied: a 6-blade 45° pitched blade turbine

and a 4-bladed A315 hydrofoil (Lightnin) in both the down- and up-pumping modes, and then compared with a Rushton turbine and a Scaba 6SRGT. In all cases, the impeller diameter was equal to $D = D_T/2$ and the agitator shaft (s = 0.008 m) extended to the bottom of the vessel.

The experiments were carried out at room temperature and atmospheric pressure. Plain tap water was used as the working fluid (coalescent system) and air was fed into the tank via a ring sparger ($D_s = 1.05D$), which was placed below the impeller ($C_s = 0.75C$). For three different impeller rotational speeds (N = 300 rpm, 400 rpm and 500 rpm) corresponding to fully developed turbulent flow,, the gas flow rate varied between $0.3 - 3.5 \times 10^{-4}$ m³s⁻¹ (which is equivalent to 0.3 - 4.2 vvm).

In addition to the mass transfer experiments, which are detailed below, other basic parameters such as power consumption and global gas holdup were measured in order to characterise the agitation systems. The power consumption of the agitator, with and without gas, was determined by measuring the restraining torque of the motor and the global gas holdup was measured by comparing the liquid level in the tank with and without aeration. For these experiments, the rotational speed of the agitator was kept constant whilst the gas flow rate was varied. Measurements were taken for several impeller speeds.

Mass Transfer

The volumetric mass transfer coefficient, $k_L a$, was measured using a dynamic measurement method, assuming perfect mixing in the liquid phase and the first order no depletion model for the gas phase. The choice of a first order model for the gas phase is justified by the fact that such simplified models still preserve the relative order of merit of agitators, making them useful comparison purposes (Lopes de Figueiredo and Calderbank, 1979). Furthermore, for low $k_L a$ values (< 0.06 s⁻¹), like those obtained in this study, the difference between first and second order (eg. perfectly mixed or plug flow models) methods is negligible (Bakker, 1992).

Tap water was used as the operating fluid and was firstly deoxygenated by bubbling nitrogen through it until the dissolved oxygen concentration, C_i , was $\leq 1 \text{ mg.l}^{-1}$ using an impeller rotational speed of 5 s⁻¹. The increase in C_i was then measured over time as air re-oxygenated the tank water. The $k_L a$ was then calculated from:

$$C_t = C^* + \frac{\left(C^* - C_0\right)}{1 - k_L a \cdot \tau} \left(k_L a \cdot \tau e^{-t/\tau} - e^{-k_L a \cdot t}\right) \tag{1}$$

where C^* is the oxygen concentration at saturation, C_0 is the oxygen concentration at t = 0 s and τ is the response time of the oxygen probe. The response time of the oxygen probe can be evaluated using the first order relation given by equation (2) (Merchuk *et al.* (1990). In this work, the response time was measured to be 9.4 seconds.

$$\frac{dC_t^*}{dt} = \frac{1}{\tau} \left(C_t - C_t^* \right) \tag{2}$$

A temperature correction to the $k_L a$ was then applied using the relation (Bouaifi and Roustan, 1994):

$$(k_L a)_{20^{\circ}C} = 1.024^{(20-T)} (k_L a)_T$$
(3)

where T is the temperature of water during the experiment.

Measurements were made for various gas flow rates, ranging from 0.45×10^{-4} to 3.2×10^{-4} m³s⁻⁴ (0.5 – 4.0 vvm), whilst the impeller speed remained constant at 5 s⁻¹. In all cases a complete dispersion flow regime was obtained.

Using the $k_L a$ values, two performance criteria were deduced in order to compare the efficiencies of the different agitators. The first criterion is the Standard Oxygen Transfer Efficiency (*SOTE*), which is defined as the ratio of the mass of oxygen transferred to the liquid to the mass of oxygen supplied to the system per unit time (Gillot and Héduit, 2000):

$$SOTE = \frac{(k_L a)_{20^{\circ}C} C_{20^{\circ}C}^* V}{Q_{O_2}}$$
(4)

where V is the volume of liquid in the vessel and Q_{O_2} is the mass flow rate of oxygen into the system. The second criterion is the Overall Transfer Efficiency (*OTE*), which represents the mass of oxygen transferred to the liquid per kWh (Bouaifi and Roustan, 1994):

$$OTE = \frac{(3.6 \times 10^6) \times (k_L a)_{20^\circ C} \ C_{20^\circ C}^* \ V}{P_g}$$
(5)

where P_g is the gassed impeller power consumption.

RESULTS AND DISCUSSION

Power Consumption

The ungassed power numbers of the different agitators studied are shown in Table 1. The results are generally in very good agreement with those published in the literature. The power values measured for the PBTD and the A315D are however somewhat greater than the values reported by Bakker (1992). These discrepancies may be explained by the use of a different tank geometry, including a flat-bottomed vessel, a different impeller diameter ratio ($D=0.4D_T$) and the possible use of a different off-the-bottom clearance ratio ($C=0.4D_T$).

The gassed to ungassed power ratio, P_g/P , for each impeller configuration against the aeration number with varying gas flow rate and for three impeller speeds is plotted in Figures 1 (a)–(c). For the Rushton turbine and the PBTD at the three impeller speeds, a continual decrease in P_g/P from >0.9 to <0.6 is noticed as the gas

flow rate increases. However, the fall in P_g/P for the PBTD is generally about 15-20% greater than for the Rushton turbine for Na up to about 0.06. As the impeller speed increases, the P_g/P curves are shifted vertically towards lower values as expected (Warmoeskerken and Smith, 1982). For the A315U and the Scaba 6SRGT, on the other hand, the power fall upon gassing is negligible and $P_g/P \cong 1$ for all three impeller speeds. This means that even at high gas flow rates these impellers are still capable of dispersing gas and flooding is prevented. For the PBTU and the A315D, the vertical shift in the P_{o}/P curves is more significant than for the Rushton turbine and the PBTD. It is also noticed that at the lowest impeller speed and for Na greater than 0.04, P_{o}/P remains >0.8 for the PBTU, however a steep increase in P_{o}/P to 1 is observed for the A315D. At first sight, this increase in power demand appears somewhat surprising. Previous studies have shown that for a single down-pumping A315, P_{g}/P , falls a little at low gas flow rates and then significantly when the gas flow rate is more important (Bakker and Van den Akker, 1994; Hari-Prajitno et al., 1998; Aubin, 2001). It is interest to note, however, that the size of the sparger used in these previous works was at least 20% smaller than the impeller diameter ($D_{z}/D \leq$ 0.8), where as in the present study the sparger is slightly larger than the impeller ($D_s/D = 1.05$). A study carried out concerning the effect of sparger design on gas dispersion, for various disc turbines, has shown that the gassed power consumption using spargers with $D_s < D$ is remarkably different to that when $D_s > D$ (Birch and Ahmed, 1997). These authors reported that the large spargers $(D_s > D)$ lead to indirect loading of the impeller, which in turn, hindered the formation of the large gas cavities behind the impeller blades and thus significantly reduced the power loss with aeration. Bakker and Van den Akker (1994) also studied the effect of sparger size on the aerated power draw of an A315D, however, the sparger sizes tested were both smaller than the impeller. Their results suggest that P_g/P of the A315D is particularly sensitive to the sparger size. Thus, the unusual power increase observed for the A315D in this study could be explained by a modified loading regime due to the use of a larger sparger $(D_s > D)$. Due to the high solidity ratio of the impeller, gas may accumulate underneath the impeller and modify the forces acting on it, thus leading to improved pumping capacity and a higher power draw.

Gas Holdup

The experimental gas holdup for the different impellers has been determined only for the lowest impeller speed N = 300 rpm (Figure 2). Values at the rotational speeds have not been measured due to the difficulty in determining accurately the height of the free liquid surface when the impeller speed increases. At low aeration numbers (< 0.03), the gas holdup values are very similar for most of the impeller types. However, as the gas flow

rate increases one can distinguish different gas holdup capacities of the various agitators. The lowest gas holdup is observed for the Scaba 6SRGT, which entrains more than 20% less gas than the down-pumping PBT and A315. When these latter impellers are operated in the up-pumping mode, however, the gas holdup increases by 10-15% for the PBT and 20-25% for the A315. Thus, in the up-pumping mode, the gas holdup capacity of the A315U is indistinguishable from that of the Rushton turbine.

Mass Transfer

Firstly, in order to verify the accuracy of the technique adopted for the measurement of $k_L a$, the results for the Rushton turbine have been compared with the correlation reported by Van't Riet (1979):

$$k_L a = 0.026 \left(\frac{P_g}{V}\right)^{0.4} v_s^{0.5}$$
(6)

where P_g is the power consumption under aeration and v_s is the superficial gas velocity.

Figure 3 compares the experimental values for N = 300 rpm obtained in this work with the correlated values. The average difference between the experimental and correlated results is approximately 3%, with the maximal difference being no greater than 8%. Considering this comparison, the experimental method employed in this work is considered to be valid for the range of operating conditions used.

Volumetric Mass Transfer Coefficients

In order to assess the importance of surface aeration on the gas-liquid mass transfer, preliminary mass transfer measurements were made for the various impeller configurations without bubbling gas through the sparger. In general, the mass transfer coefficients due to surface aeration were not greater than 5 % of the mass transfer coefficients measured with sparger aeration, and were therefore judged as negligible.

The evolution of the volumetric mass transfer coefficients as a function of the aeration number with gas flow rate for all agitators at different rotational speeds is shown in Figures 4 (a)-(c). As expected, the mass transfer coefficients increase with increasing gas flow rate due to the increased gas holdup. Also, the k_La values for each impeller configuration increase with increasing *N*. This is because as the impeller speed increases, bubble breakup is enhanced, thus increasing the surface area needed for mass transfer. Clearly for all rotational speeds, the mass transfer coefficient for the Rushton turbine is much higher than those for the other impellers, being on average greater than 55% more. This can be explained by the fact that the experiments were carried out at constant rotational speeds and not at constant power dissipation. Since the k_La is a function of gassed power consumption, the Rushton turbine has a much larger mass transfer coefficient because it dissipates significantly more power than the other impellers studied at constant impeller speed. Physically, the energy dissipated by the Rushton turbine is used both for bubble break-up, thus increasing a, and for enhancing turbulence effects at the proximity of the gas-liquid interface, which increases k_L . Of course, there is a limit where an increase in P_g will no longer affect the $k_L a$.

For the other impellers studied, it can be seen that at N = 300 rpm, the $k_L a$ values are relatively similar. However, as N increases, the mass transfer coefficients of the up-pumping PBT and A315 increase at a higher rate than the down-pumping configurations and the Scaba. At 500 rpm, the $k_L a$ values for the up-pumping configurations are as much as 50% greater than those of the down-pumping impellers and the Scaba turbine. This could be explained by an increased gas holdup at higher rotational speeds: as the impeller speed increases, the lower circulation loop generated by the up-pumping impellers becomes stronger and entrains the gas bubbles in the lower half of the tank for a longer time, thus increasing the gas holdup and the capacity for mass transfer.

Standard Oxygen Transfer Efficiency (SOTE)

Figure 5 presents the oxygen transfer efficiency as a function of *Na* with varying gas flow rate for different impeller speeds. It can be seen that for all impellers the *SOTE* decreases with increasing gas flow and decreasing *N*. The former observation is contradictory to what one would expect, which would be constant oxygen transfer efficiency since the k_La increases with increasing oxygen flow. This counterintuitive result may be explained with respect to the gas flow regimes. At low gas flow rates, the gas is completely dispersed and the bubbles have a relatively long residence time, which promotes mass transfer and thus the *SOTE* values are relatively high. As the gas flow rate increases, however, the gas flow regime moves away from complete dispersion and towards flooding of the impeller. As a result, the bubbles are not as well dispersed and have a shorter residence time in the tank, which decreases the time for mass transfer and thus the quantity of oxygen transferred reduces even though the oxygen input increases. When N increases, however, the k_La increases, thus improving the *SOTE*.

Comparing the efficiencies of the different agitators, a similar trend to the evolution of the $k_L a$ is observed. The Rushton turbine produces significantly higher *SOTE* values than the other impellers. These values, however, decreases more quickly with increasing gas flow than the *SOTE* values of the other impellers, which follows the poor gas handling capacity of the Rushton turbine. For the other impellers, it can be seen that the uppumping configurations become clearly more efficient than the down-pumping impellers and the Scaba as the impeller speed increases. Generally, for the impellers studied, the oxygen transfer efficiencies are low, which is due to the fact that the experimental pilot used is small (approximately 5 L with a liquid height of only 0.19 m) and the residence time of the bubbles is therefore short, reducing the time for mass transfer to occur. Of course, in an industrial sized vessel where the bubble residence time would be much larger, higher *SOTE* values would be expected.

Overall Transfer Efficiency (OTE)

The evolution of the overall transfer efficiency of each impeller system as a function of the aeration number is shown in Figure 6 (a)-(c). For all impellers, the *OTE* increases with increasing gas flow. This is because as the gas flow rate increases, the k_La also increases but the gassed power consumption decreases or remains more or less constant. As the impeller speed increases, the *OTE* of course decreases due to the increased power consumption at higher rotational velocities.

In can be seen that when using this criterion, the Rushton and Scaba turbines generally have the lowest performance in terms of mass transfer efficiency, compared with the other impellers, and are as efficient as one another. Even though the Rushton turbine enables a large quantity of oxygen to be transferred to quickly to the liquid phase, it requires a large amount of power in order to carry out the process. On the other hand, the Scaba turbine requires less power than the Rushton turbine but transfers a smaller quantity of oxygen to the liquid phase. Looking at the effect of axial pumping direction on the *OTE* shows that there is little consequence on the values for the A315, however the up-pumping PBT is less efficient than the down-pumping mode except at the highest rotational speed. As the impeller speed increases however, the axial flow impellers appear to be equally efficient, and significantly more efficient than the radial turbines.

CONCLUSIONS

This paper deals with the performance of down- and up-pumping axial flow impellers (PBT and A315) for gasliquid mass transfer applications compared with the performance of the Rushton and the Scaba turbines. For each characteristic quantity, the performance of the impellers studied can be summarized in decreasing superiority as the following.

In terms of gas handling capacity (from gassed power consumption):

• A315U & Scaba 6SRGT > A315D & PBTU, Rushton turbine > PBTD

In terms of gas holdup capacity:

• A315U & Rushton turbine > PBTU > A315D & PBTD > Scaba 6SRGT In terms of *k*_L*a* and *SOTE*:

• Rushton turbine > A315U & PBTU > Scaba 6SRGT, A315D & PBTD In terms of *OTE*:

• A315U, A315D, PBTD & PBTU > Scaba 6SRGT & Rushton turbine

In general, it can be seen that when the axial flow impellers are operated in the up-pumping mode, they often give superior performance in gas-dispersion and gas-liquid mass transfer applications than the downpumping configuration (in the range of operating conditions studied). This is due to the extremely different flow patterns that are generated by up- and down-pumping impellers. In the up-pumping mode, axial flow impellers generate bulk flow patterns that are similar to those created by radial tubines: two primary circulation loops are formed, one in the lower half of the vessel and one in the upper half (Aubin et al., 2001; Jaworski et al., 1998, 2001). Of course the upper circulation loop enables increased recirculation of the gas phase, thus allowing more time for gas-liquid mass transfer to occur. Furthermore, the modification in the global flow pattern most likely alters the local pressure fields around the impeller blades, resulting in improved gas handling capacities. Regarding the radial impellers, it appears that although the Scaba 6SRGT has an extremely high gas handling capacity and relatively low power consumption, its performance for gas-liquid mass transfer is moderate due to its low gas holdup capacity and consequent low $k_L a$. The Rushton turbine, on the other hand, has a high gas holdup capacity and $k_L a$ but is energy demanding, making it economically inefficient. Overall, it appears that the A315U gives the highest performance for all impellers in terms of gas handling capacity and mass transfer efficiency. On the other hand, although the down pumping axial flow impellers have a better OTE than most others, they have additional disadvantages for gas-liquid dispersions, such as the generation of extremely high torque fluctuations, which can cause mechanical damage to the stirred tank system (Hari-Prajitno et al., 1998), making them perhaps the least well suited to gas-liquid mass transfer applications.

Finally, it should be pointed out that although the results presented here only concern the physico-chemical side of mass transfer, they will be particularly useful for the development of simple liquid flow models, such as the Compartment Model Approach (CMA) (Vrabel *et al.*, 2000), which can be easily used by process engineers for design purposes. In addition, since in industrial bioreactors and fermenters the physico-chemical gas-liquid mass transfer process is most often coupled with chemical reaction, it would be interesting to integrate into the model more detailed results concerning the transfer of oxygen to the liquid phase, which is then consumed by a

bioreaction. This would provide an advanced model for bioreactors, which could be used as a tool to obtain a more complex and complete picture of the relevant phenomena occurring in the reactor.

NOMENCLATURE

С	impeller clearance (m)
C_s	sparger clearance (m)
C_t	dissolved oxygen concentration at time t (kg.m ⁻³)
C_0	dissolved oxygen concentration at $t = 0$ (kg.m ⁻³)
C^{*}	oxygen concentration at saturation (kg.m ⁻³)
D	impeller diameter (m)
D_s	sparger diameter (m)
D_T	tank diameter (m)
Η	liquid height in tank (m)
<i>k_La</i>	volumetric mass transfer coefficient (s ⁻¹)
Ν	impeller rotational speed (s ⁻¹)
Р	ungassed impeller power consumption (W)
P_{g}	gassed impeller power consumption (W)
P_o	Dimensionless power number (-)
Q_G	gas flow rate (m ³ s ⁻¹ or vvm)
Q_{O_2}	mass flow rate of oxygen (kg. s^{-1})
S	shaft diameter (m)
t	time (s)
Т	temperature (°C)
τ	response time of the oxygen probe (s)
V_{S}	superficial gas velocity (ms ⁻¹)
V	volume of liquid in the tank (m ³)

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Table 1: Comparison of ungassed power numbers for the different impellers studied. ^aBertrand *et al.* (1980); ^bVrabel *et al.* (2000); ^cRanade and Joshi (1989); ^dMishra (1993); ^eJaworski *et al.* (1991); ^fBakker (1992); ^gAubin *et al.* (2001); ^hAubin (2001).

Impeller Type	P_o – This work (Error: ± 5%)	P_o – Literature
Rushton Turbine	4.60	4.9 ^a
Scaba 6SRGT	1.72	1.5 ^b
PBTD	1.96	$2.21^c - 2.1^d - 1.70^e - 1.55^f - 1.93{\pm}0.05^g$
PBTU	2.03	$2.58{\pm}0.04^{g}$
A315D	1.32	$0.76^{\rm f}-1.26{\pm}0.05^{\rm h}$
A315U	1.34	$1.17{\pm}0.07^{ m h}$

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Figure 1: Evolution of the gassed to ungassed power ratio with increasing gas flow rate: (a) N = 300 rpm; (b) N = 400 rpm; (c) N = 500 rpm.

Figure 2: Comparison of gas holdup at different gas flow rates for the various impellers studied (*N* = 300 rpm).

Figure 3: Comparison of experimental $k_L a$ values for the Rushton turbine with the correlation reported by Van't Riet (1979) (N = 300 rpm).

Figure 4: Volumetric mass transfer coefficients as a function of *Na* at constant impeller speed: (a) N = 300 rpm; (b) N = 400 rpm; (c) N = 500 rpm.

Figure 5: Comparison of the Standard Oxygen Transfer Efficiency (*SOTE*) as a function of *Na* at constant impeller speed: (a) N = 300 rpm; (b) N = 400 rpm; (c) N = 500 rpm.

Figure 6: Comparison of Overall Transfer Efficiency (*OTE*) as a function of *Na* at constant impeller speed: (a) N = 300 rpm; (b) N = 400 rpm; (c) N = 500 rpm.





















Figure 4













Figure 6