Enhancing energy efficiency and mass transfer in solid-liquid systems using mechanical mixing and cavitation

A thesis submitted in fulfilment of the requirements for the degree of

Doctor of Philosophy

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Declaration

I certify that except where due acknowledgement has been made, the work is that of the author alone; the work has not been submitted previously, in whole or in part, to qualify for any other academic award; the content of the thesis is the result of work which has been carried out since the official commencement date of the approved research program; any editorial work, paid or unpaid, carried out by a third party is acknowledged; and, ethics procedures and guidelines have been followed.

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Peer reviewed conference papers


Conference presentations

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Nomenclature

A \quad \text{constant}
a \quad \text{constant}

\text{A310} \quad \text{Lightnin A310 hydrofoil turbine}

\alpha_p \quad \text{solid-liquid interfacial area per unit volume of solid, m}^{-1}

B \quad \text{baffle width, m}
b \quad \text{constant}

C \quad \text{impeller clearance from tank bottom, m}
c \quad \text{constant}

C_L \quad \text{bulk liquid concentration, mol/m}^3

(C_{Na}) \quad \text{NaOH concentration, mol/m}^3

(C_{Na})_0 \quad \text{initial NaOH concentration, mol/m}^3

C_S \quad \text{solute concentration at the solid surface, mol/m}^3

C_V \quad \text{volumetric solids concentration, (v/v)}

C_{V(max)} \quad \text{solids concentration with maximum k_{SL} value, (v/v)}

C_{V(op)} \quad \text{solids concentration with lowest } \varepsilon_{JS} \text{ value, (v/v)}

D \quad \text{impeller diameter, m}

d_{32} \quad \text{Sauter-mean diameter, m}

D_A \quad \text{molecular diffusivity, m}^2/s

d_p \quad \text{particle diameter, m}

E\% \quad \text{enhancement in mass ratio, } \%

g \quad \text{gravitational acceleration constant, m/s}^2
H  liquid height, m
HB  solids bed height, m
HS  solids cloud height, m
K  constant
k  constant
kD  constant
kSL  solid-liquid mass transfer coefficient, m/s
kSL,ap  solid-liquid volumetric mass transfer coefficient, s⁻¹
kSL(cal)  estimated solid-liquid mass transfer coefficient value, m/s
kSL(exp)  solid-liquid mass transfer coefficient value from experimental data, m/s
M  rate of diffusional mass transfer, mol/m³.s
ML  mass of liquid, kg
Ms  mass of solids, kg
MR(silent)  solids mass ratio in the absence of ultrasound
MR(US)  solids mass ratio in the presence of ultrasound
m  constant
N  number of data points
N  impeller speed, rps
NJCD  minimum impeller speed to attain ‘just complete dispersion’ condition, rps
NJJS  minimum impeller speed to achieve ‘just suspended’ condition, rps
NP  impeller power number
n  constant
P  impeller power consumption, W
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<td>PBT</td>
<td>6 bladed-45°pitch turbine</td>
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<tr>
<td>$P_{JS}$</td>
<td>impeller power consumption to achieve ‘just suspended’ conditions, W</td>
</tr>
<tr>
<td>Re</td>
<td>particle Reynolds number</td>
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<td>$Re_{Imp}$</td>
<td>impeller Reynolds number</td>
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<td>$Re_{P}$</td>
<td>modified particle Reynolds number</td>
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<td>RT</td>
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<td>Schmidt number</td>
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<td>Sh</td>
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<tr>
<td>t</td>
<td>time, s</td>
</tr>
<tr>
<td>$U_P$</td>
<td>Ultrasonic power, W</td>
</tr>
<tr>
<td>v</td>
<td>volume, m$^3$</td>
</tr>
<tr>
<td>X</td>
<td>ratio of mass solids to mass liquid, (kg/kg)</td>
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Greek symbols

$\varepsilon$ energy dissipation rate per unit mass liquid, W/kg

$\varepsilon_{\text{JS}}$ specific impeller power consumption at $N_{\text{JS}}$, W/kg

$\eta_r$ viscosity of the continuous phase, Pa.s

$\eta_{\text{Slurry}}$ apparent slurry viscosity, Pa.s

$\mu_L$ liquid phase viscosity, Pa.s

$\nu$ kinematic viscosity, m$^2$/s

$\rho_L$ liquid density, kg/m$^3$

$\rho_S$ solid density, kg/m$^3$

$\rho_{\text{slurry}}$ slurry density, kg/m$^3$

$\tau$ absolute torque, N.m

$\tau_m$ measured torque, N.m

$\tau_0$ residual torque, N.m

$\phi_m$ maximum volume fraction
Summary

Solid-liquid agitated vessels are widely used in the mineral process industry where there is a strong demand to intensify existing vessels to process more ore. It has been shown that process intensification can be achieved by increasing solids throughput or by optimising process equipment and conditions. This can be achieved by operating a taller/larger agitated vessel fitted with multiple impellers or by increasing the extraction yield via cavitation. These methods, however, require the addition of extra energy. Therefore, the main objective of this work is to determine if process intensification of solid-liquid agitated vessels is more efficiently achieved by increasing the vessel height and fitting a second impeller or by introducing cavitation into the slurry. This is achieved by determining the best impeller and baffle arrangement in a dual-impeller agitated vessel through a systematic experimental study, involving a range of solids concentration from 0.05 to 0.35 (v/v). The enhancement of solid-liquid mass transfer due to cavitation is also studied and compared to that in the dual-impeller system. This study also aims to study if the Zwietering correlation to predict the critical impeller speed ($N_{JS}$) in solid-liquid agitated vessels can be reliably applied to dual-impeller systems. It is also aimed to develop mathematical correlations using experimental data for estimating the specific impeller power consumption ($\varepsilon_{JS}$) and the solid-liquid mass transfer coefficient ($k_{SL}$) for dual-impeller systems. Another aim of this study is to determine the influence of particle at high concentrations on cavitation activity.

Impeller power consumption and mass transfer experiments in dual-impeller vessels were carried out in 0.2 m diameter (T) cylindrical tanks with liquid height (H) fixed at 1.5T to study solids suspension, dispersion, and mass transfer coefficient. Aqueous NaOH and cationic exchange resins were used as the liquid and solid phases, respectively. The impeller configurations used were either two 6-bladed Rushton turbines (RTRT) or an A310 hydrofoil and a 6 bladed-45°pitch turbine (A310PBT).

Experiments studying the influence of particles at high concentrations on cavitation activity were carried out in a 0.2 m agitated vessel irradiated with ultrasound and fitted with an A310 impeller. The effect of particle type and size were also determined using cation exchange resins, sand, and glass spherical beads of different sizes (206, 551 and 1290 µm) as the solid phases and aqueous potassium iodide (KI) as the liquid phase. Experiments investigating mass transfer enhancement with cavitation were carried out in a 0.2 m agitated vessel.
irradiated with ultrasound and fitted with a Rushton turbine. Aqueous NaOH and cation exchange resin were used in the ‘ion exchange’ system whereas polymeric resin saturated with phenol and water were used as the solid and liquid phases, respectively in the ‘desorption’ system.

Experimental results show that the applicability of the Zwietering correlation can be extended to vessels operating up to a solids concentration of 0.35 (v/v). This is found to be true for baffled and unbaffled vessels agitated with a dual-impeller configuration in addition to single-impeller agitated vessels. From the experimental data, new values of the Zwietering constant, S were determined for the geometrical setups used in this study. It was found that by operating the system at $C_V = 0.2$ (v/v), the impeller power consumption required for solids suspension per unit mass solids ($\varepsilon_{JS} = P_{JS}/M_S$) is minimised while achieving maximum $k_{SL}$ values at the same time. Particle dispersion was found to generally increase with increasing $C_V$ in all mixing systems used in this work. The results also showed that the removal of baffles leads to a reduction in power requirements for solids suspension as well as dispersion. But, the influence of baffles on $k_{SL}$ values is dependent on impeller type and vessel geometry. The results suggest that generally the radial pumping impellers are more energy efficient while achieving comparable $k_{SL}$ values. Overall, improved energy efficiency and increased solids throughput can be achieved using radial flow impellers under unbaffled conditions at relatively higher $C_V$.

Experimental data obtained in this work was used to derive mathematical correlations to estimate $\varepsilon_{JS}$, impeller power for solids dispersion, and $k_{SL}$ in solid-liquid agitated vessel fitted with single- or dual-impellers. The values estimated using these correlations were found to fit experimental data within a $\pm 15\%$ band for a $C_V$ range of 0.05 - 0.35 (v/v).

It was found experimentally that cavitation activity decreases with increasing solids concentration up to 0.1 (v/v) but increases thereafter up to 0.4 (v/v) followed by a further decrease due to the net effect of wave attenuation and increased number of nucleation sites available. Cavitation activity increases with increasing particle diameter due to a decrease in the liquid tensile strength when larger particles are present thereby decreasing the cavitation threshold. Increased surface roughness enhances the cavitation level. These results imply that particle size, and concentration, and surface roughness all play important roles in the formation and subsequent collapse of cavities thereby influencing sonochemical reaction yields.
Experimental results suggest that the presence of ultrasound has no influence on solid-liquid mass transfer in an ion exchange system for a $C_V$ range of 0.03-0.20 (v/v). However, the rate and amount of phenol desorbed from polymeric resin is higher under combined agitation and ultrasonic irradiation when compared to agitation alone. For the phenol desorption system, the influence of cavitation on mass transfer increases with time and is $C_V$ dependent. These results imply that ultrasonically enhanced solid-liquid mass transfer in agitated vessels is dependent on the physical and chemical properties of the solid and liquid phases as well as $C_v$. The ultrasonic power consumption and the impeller power consumption are compared and from the results, it is clear that mechanical agitation is more efficient at intensifying solid-liquid agitated vessels.
Chapter 1

Introduction
Introduction

Mechanically agitated vessels are widely utilised in many process industries to achieve the dispersion of solids in a liquid for mass transfer purposes (Wang et al., 2014). Mixing vessels are able to perform many operations such as dissolution, leaching, crystallization and precipitation, adsorption, desorption, ion exchange and solid-catalysed reactions. In the mineral industry, mixing is used to extract minerals from ore, with the extraction of copper from copper oxide bearing ore materials being one example (Davenport et al., 2002). Even though most copper ores contain copper-iron-sulphide for which the most viable processing method is pyrometallurgy, copper oxide and some copper sulphide ores can be processed via the hydrometallurgical method. Advantages of this method include lower energy cost, no SO$_2$ emission, and the ability to economically process low-grade ores (Davenport et al., 2002).

Uncertainty over metal prices and production costs has led to a strong demand in the mineral processing industry to intensify operations. Intensifying solid-liquid mass transfer processes to produce more material per unit power input without any major changes to existing infrastructure is, therefore, of great interest (Wang et al., 2014). Process intensification in solid-liquid agitated vessels can be achieved by recovering more processed materials. This can be achieved by increasing the solids throughput or by increasing the amount of solute extracted through efficient mixing or supplying additional energy.

Achieving higher solids throughput simply by increasing the solids concentration will result in more concentrated slurry which has an influence on the solid-liquid mass transfer coefficient and the impeller power required to achieve solids suspension. Another way of achieving increased solids throughput is to operate a larger or taller vessel. This will increase the vessel volume and, therefore, enable more material to be processed. However, taller vessels require multiple impellers to achieve better solids distribution and overall mixing with the consequence of increased total impeller power consumption (Paul et al., 2004). Having said this, process intensification of agitated vessels by means of operating at higher solids concentration or optimising process equipment is becoming an attractive option (Wu et al. 2010a).

There have been many studies in the literature on solid-liquid dispersion and mass transfer. While most of these studies have focused on systems with low solids concentrations, more recent investigations have suggested that operating solid-liquid agitated vessels at relatively
high solids concentrations led to improved energy efficiency as well as enhanced mass transfer (Wu et al., 2012; Bong et al., 2012). A few previous studies reported the influence of tank and impeller geometries on particle suspension and mass transfer (Tagawa et al., 2011; Ayranci and Kresta, 2011). While these studies are limited to agitated vessels fitted with a single impeller, numerous other studies involving multiple impeller systems focused on parameters associated with vessel geometry and impeller configuration such as liquid height to tank diameter ratio, number and type of impellers as well as their geometric position in the tank (Bujalski et al., 1999; Gogate et al., 2000; Angst and Kraume, 2006). The influence of solids concentration and vessel & tank geometry on solids suspension, solids dispersion, and mass transfer in dual-impeller operated solid-liquid agitated vessels has not been investigated in great detail. Therefore, the present work aims to investigate how these variables affect mass transfer and impeller power consumption in dual-impeller operated agitated vessels to determine its viability in achieving process intensification.

Process intensification can also be achieved by means of increasing the extraction yield per unit mass of solids. It has been shown that copper extraction rates can be improved with the use of ultrasound in leaching systems (Sarveswara Rao et al., 1997; Swamy and Narayana, 2001). It has been explained that cavitation is the mechanism by which enhanced extraction is achieved (Okur et al., 2002). Although ultrasound has been shown in the literature to enhance interphase mass transfer, the effect of the particulate matter, especially at high solids concentrations on cavitation events has been largely unexplored. Similarly, the influence of solids concentration and tank geometry on mass transfer and power usage in agitated sonoreactors has not been studied in detail. Previous studies have focused on the influence of particle type, size, and concentration on cavitation whilst operating at different ultrasonic frequencies (Keck et al., 2002; Lu et al., 2002; Shimizu et al., 2007). They have shown that there is a decrease in sonochemical yield with increasing particle concentration and size. Therefore, it is of great interest to understand how cavitation affects solid-liquid mass transfer whilst operating at high solid loadings as well as to determine whether the additional energy supplied to generate cavitation is efficiently used to achieve process intensification.

For diffusion-rate controlled processes such as solid-liquid mass transfer, suspending solids just off the tank bottom ensuring the availability of maximum surface area is sufficient. The minimum impeller speed to ‘just suspend’ all particles is denoted \( N_{JS} \) and is an important parameter for vessel design. Zwietering (1958) proposed a criteria and a mathematical correlation to estimate \( N_{JS} \). Though there have been more recent correlations (Baldi et al.,
the Zwietering correlation is still the most widely used. While recent studies have largely corroborated Zwietering’s findings (Nienow, 1968; Raghava Rao et al., 1988; Mak, 1992), others have identified systems where the correlation is not reliable. Choudhury (1997) pointed out one such system which operates with solid loadings greater than 0.15 (v/v). Other studies focused on systems with different vessel or impeller geometries (Mak, 1992; Ibrahim and Nienow, 1996; Armenante and Nagamine, 1998; Atiemo-Obeng et al., 2004; Ayranci and Kresta, 2011). However, these studies were confined to fully baffled vessels and therefore the reliability of the Zwietering correlation to estimate \( N_{JS} \) in unbaffled vessels fitted with single- or dual-impellers has not been tested yet. Another important design parameter is the solid-liquid mass transfer coefficient (\( k_{SL} \)). The \( k_{SL} \) is a function of many variables including diffusivity, viscosity, slurry density and tank configuration. The \( k_{SL} \) is a proportionality constant in an equation where the amount of mass transferred is related to the interfacial surface area and the concentration difference and is an indication of the system’s mass transfer efficiency (Cussler, 2009).

It is clear that knowledge on the effect of solids concentration in dual-impeller operated solid-liquid agitated vessels is not complete. This is especially the case for the solid-liquid mass transfer coefficient and impeller power consumption at high solids concentration. The reliability of the Zwietering correlation in estimating \( N_{JS} \) in such systems is also unknown. It is also clear that there is a lack of knowledge on how particles affect levels of cavitation in an agitated solid-liquid system operating at high solids concentration. Therefore, it would be beneficial to investigate the effects of particle concentration, size, and surface topography on cavitation levels, and its effect on solid-liquid mass transfer in agitated vessels. From this new knowledge, a more informed decision can be made in regards to the most efficient method of adding energy to achieve process intensification.

This study will investigate how particle concentration, size, and surface topography affect the cavity formation to determine its influence on sonochemical processes. This work will also determine the role of ultrasound on mass transfer as well as how solids concentration and tank geometry affect it. This study will provide a further understanding on the design of solid-liquid agitated vessels fitted with dual-impellers for handling high concentrations of solids. The results of this study will be beneficial for optimising impeller power consumption and achieve increased throughput efficiently. From the data obtained, design correlations will
be proposed so that $k_{SL}$ and energy dissipation rates can be using information such as vessel geometry and material properties.

**Main research objectives**

The main research objectives of this project are:

- To compare the energy efficiency of mechanical mixing and cavitation in achieving process intensification.
- To investigate the influence of the following variables on impeller power consumption and solid-liquid mass transfer coefficient
  - solids concentration
  - impeller type
  - baffles.
- To study the effect of employing a taller tank as a means of increasing throughput with the aim of process intensification.
- To determine whether the Zwietering correlation can be applied to solid-liquid mixing systems with
  - high solids concentrations
  - no baffles
  - dual-impellers.
- To study how particle concentration, size and surface topography affect sonochemical processes.
- To study the effect of cavitation on solid-liquid mass transfer in agitated systems with high solids concentration.
- To develop mathematical correlations using experimental data for estimating the solid-liquid mass transfer coefficient, and impeller power consumptions to ‘just’ suspend and completely disperse solids in liquid.
Organisation of thesis

The thesis is organised as follows:

Chapter 2: Literature Review

This chapter reviews the literature relating to solids suspension & distribution, and solid-liquid mass transfer in mechanically agitated vessels. A discussion on cavitation and the parameters that affect it are also presented. Also, the literature on impeller and ultrasonic power consumptions in several mixing systems with different tank and impeller geometries are discussed. Various approaches used in developing correlations to estimate specific power input and solid-liquid mass transfer coefficient in agitated vessels are also discussed.

Chapter 3: Experimental

This chapter describes the equipment, materials, and methodology used to measure $N_{JS}$, power consumption, the solid-liquid mass transfer coefficient and cavitation activity.

Chapter 4: Results and discussion – Predicting $N_{JS}$ using the Zwietering correlation for single- and dual-impeller systems

This chapter examines the reliable use of the Zwietering correlation in estimating $N_{JS}$ in solid-liquid mixing systems operating at high solids concentrations. The correlation will be tested in solid-liquid agitated vessels fitted with dual-impellers in the absence of baffles.

Chapter 5: Results and discussion – Impeller power consumption for single- and dual-impeller systems

This chapter analyses the experimental results on impeller power consumption required to 'just suspend' solids at concentrations up to 0.35 (v/v) in agitated vessels fitted with dual- and single-impellers. Total impeller power consumption is presented as a function of different tank and impeller geometries. This chapter also presents the experimental results on the impeller power consumption to distribute particles throughout the vessel. The chapter shows impeller power consumption to achieve varying degrees of solids dispersion in agitated vessels of different size and impeller configuration.
Chapter 6: Results and discussion – Solid-liquid mass transfer in agitated vessels with single- and dual-impellers

This chapter discusses the experimental results of solid-liquid mass transfer coefficient in an ion exchange system in dual- and single-impeller agitated vessels. The effect of solids concentration as well as different tank and impeller geometries on mass transfer coefficient are presented here. The optimum parameters for interphase mass transfer are determined on the basis of efficient specific impeller power consumption and high solid-liquid mass transfer coefficient values.

Chapter 7: Mathematical correlations to estimate the impeller power consumption for solids suspension & solids dispersion, and solid-liquid mass transfer coefficient

The experimental data obtained in this work are used to develop design correlations to estimate the solid-liquid mass transfer coefficient and impeller power consumption required to either ‘just’ suspend or completely disperse solids in liquid.

Chapter 8: Results and discussion – Particle influence on cavitation

This chapter discusses the results of cavitation activity in an agitated sonoreactor as a function of particle concentration, size, and type. The cavitational activity is quantified on the basis of tri-iodide production in the bulk solution.

Chapter 9: Solid-liquid mass transfer with ultrasound

This chapter discusses solid-liquid mass transfer in agitated vessels incorporating additional energy input in the form of cavitation. The results for two different solid-liquid systems (mechanical agitation and sonication) chosen in this work are discussed as a function of solids concentration. The results for total power consumption and mass transfer in ultrasonically enhanced solid-liquid systems are discussed and a comparison is made against mechanically agitated systems in regards to their efficiency of achieving process intensification.

Chapter 10: Conclusions and recommendations

This chapter presents the conclusions of this study and recommendations are made for future work.
Chapter 2

Literature Review
Introduction

This chapter is segregated into three sections with the first section critically reviewing the literature on solids suspension and solids dispersion as well as summarising various experimental measurement techniques used for determining critical impeller speed for off-bottom solids suspension, \( N_{JS} \). The review will also incorporate studies on the effect of solids concentration, impeller type, and baffling conditions on impeller power consumption. The second section of this chapter will present a general review of previous investigations on solid-liquid mass transfer as well as the effect of solids concentration, impeller type, and baffling conditions on the solid-liquid mass transfer coefficient, \( k_{SL} \). The approaches used in developing a correlation to estimate \( k_{SL} \) will also be discussed. Lastly, a review of the literature on cavitation and its effect on solid-liquid mass transfer will be presented.

2.1 Particle suspension in mechanically agitated vessels

The suspension of particles in an agitated vessel is an important unit operation in many process industries. As the particle is denser than the liquid, an additional force generated by the impeller is required to provide the energy to lift and suspend the particles. The energy input by the impeller creates a circulating turbulent flow, which counteracts the tendency of particles to settle due to gravity, and disperses them throughout the vessel. Particle suspension is a resultant of lift and drag forces of the moving fluid acting on the particles as well as bursts of turbulent eddies. It has been suggested that particles settled on the tank bottom tend to swirl around until they are suddenly lifted up by a turbulent burst (Atiemo-Obeng et al., 2004).

For diffusion-rate controlled processes, effective contact between suspended particles and the surrounding liquid is vital for mass transfer optimisation. The role of agitation is to ensure the entire solid surface area is available by allowing all particles to freely move without settling and providing bulk liquid turbulence to enhance mass and energy transfer (Nienow, 1968). The mean fluid velocity and its distribution, and the generation of anisotropic turbulent eddies are dependent on several factors including physical properties of the liquid and the geometric parameters of the impeller and vessel. Thus, different agitator designs achieve
different degrees of suspension at similar energy inputs. As in the case of particle suspension, mass transfer rates are also affected by impeller type as different impellers generate different bulk liquid flow patterns (Nienow and Miles, 1978; Upadhyay et al., 1994).

In agitated vessels, the degree of solids suspension is usually classified into three levels: on-bottom motion, complete off-bottom suspension, and uniform suspension (Paul et al., 2004). Many applications often require just enough agitation to just suspend the particles off the tank bottom so that maximum interfacial area is available for transfer processes. The minimum agitation speed to achieve this is designated as the critical impeller speed ($N_{JS}$), where the subscript ‘JS’ refers to ‘just suspended.’ Researchers have determined that operating the impeller below this critical impeller speed is not desirable for mass transfer processes (Harriott, 1962). Other researchers have also suggested that when operating the impeller above the critical speed, mass transfer rates increases only slightly and decreases eventually (Brucato et al., 2010). Therefore, for processes where diffusional mass transfer is the rate-limiting step, operating at critical impeller speed is suitable for mass transfer and impeller power consumption purposes.

### 2.1.1 Determination of the critical impeller speed ($N_{JS}$)

Much of the work in the literature pertaining to solids suspension is related to $N_{JS}$. Therefore, due to its importance in achieving effective mass transfer, it is important to understand how various parameters affect $N_{JS}$. Zwietering (1958) first introduced the visual method to determine $N_{JS}$. He defined $N_{JS}$ as the impeller speed at which no particle remains at the vessel bottom for more than 1 or 2 seconds and all particles are in motion. This criterion is frequently used by researchers as a measure of suspension level (Bujalski et al., 1999; Drewer et al., 2000). However, some researchers have questioned the accuracy of this method. Oldshue and Sharma (1992) reported that the method is limited to low solids concentrations (< 0.10 wt). While Kasat and Pandit (2005) pointed out that excessive energy is required to lift solids from relatively stagnant regions such as the periphery of the vessel bottom and near the baffles. They also mentioned that it is impractical to increase the impeller speed by up to 20 -50 % to suspend such small amounts of particles. Other researchers have also highlighted this issue in their investigations (Nienow at al., 1997; Wu et al., 2010a).
Another visual method was introduced by Einenkel and Mersmann (1977) based on observing the height of the interface between the solids rich slurry and the relatively clear liquid above. The distance from the vessel bottom to this interface is called the cloud height. They defined $N_{JS}$ as when the cloud height is 90% of the total liquid height. This method of determining $N_{JS}$ has been found to be 20 - 25% higher than that determined using Zwietering’s method (Kraume, 1992; Kasat and Pandit, 2005).

Wu et al. (2002) and Wu et al. (2012) proposed a different visualisation method to determine $N_{JS}$ based on the experimental technique proposed by Hicks et al. (1997). According to this method, the sedimentation bed height ($H_B$) is measured at various impeller speeds, and $N_{JS}$ is defined as the conditions at which the sedimentation bed height is zero ($H_B = 0$). Decreasing the impeller speed below $N_{JS}$ will result in a visible sedimentation bed height ($H_B > 0$). The repeatability of this method in measuring $N_{JS}$ has been found to give consistent results with an error of ±2%. In addition, this method has been demonstrated to be reliable when measuring $N_{JS}$ in suspensions with high solids concentrations (Wu et al., 2010b; Wang et al., 2012a). Due to the presence of stagnant regions and particles not being evenly distributed throughout the tank, $H_B$ was measured at midpoint between two consecutive baffles.

### 2.1.2 The Zwietering correlation to estimate the critical impeller speed ($N_{JS}$)

Zwietering (1958) conducted more than a thousand experiments investigating solids suspension in baffled vessels with diameters ranging from 0.15 to 0.60 m. He used a variety of impellers including 2-bladed paddles, 6-bladed Rushton turbine, vane disc, and marine propeller. The particles used were sodium chloride and sand with particle sizes ranging from 125 to 850 µm and at a solids concentration ranging from 0.05 to 0.2 (wt%). He also studied the effect of different fluid properties on solids suspension using liquid densities ranging from 790 to 1600 kg/m$^3$ and viscosity ranging from $0.31 \times 10^{-3}$ to $9.3 \times 10^{-3}$ Pa.s.

Zwietering proposed that the suspension of solid particles in stirred vessels were dependent on the following 10 factors:

- Impeller speed, $N$
- Vessel diameter, $T$
- Impeller clearance, $C$
- Impeller diameter, $D$
- Particle diameter, $d_p$
- Density of solids, $\rho_S$
- Density of liquid, $\rho_L$
- Kinematic viscosity, $\nu$
- Gravity constant, $g$
- Ratio of mass solids to mass liquid, $X$

From these factors, and following a dimensional analysis, seven dimensionless groups were obtained:

- Reynolds number, $\text{Re} = \frac{ND^2}{\nu}$
- Froude number, $\text{Fr} = \frac{N^2D}{g}$
- Density ratio, $\frac{\rho_S}{\rho_L}$
- Geometric ratios, $T/D$, $T/C$, $D/d_p$
- Ratio of mass solids to mass liquid, $X$

From the analysis of the experimental data, the following relationship was obtained by Zwietering (1958):

$$
\left(\frac{N D^2}{\nu}\right)^{0.1} \left(\frac{\rho_L D N^2}{g \Delta \rho}\right)^{0.45} \left(\frac{D}{d_p}\right)^{0.2} = K \left(\frac{T}{D}\right)^{\alpha} X^{0.13}
$$

(2.1)

The constant $K$ and the exponent $\alpha$ are dependent on the position and type of impeller. The above equation can be rewritten as:

$$
N_{JS} = S \nu^{0.1} \left[\frac{g(\rho_S-\rho_L)}{\rho_L}\right]^{0.45} d_p^{0.2} D^{-0.85} X^{0.13}
$$

(2.2)

where $S$ is a constant based on impeller type, impeller diameter, and impeller off-bottom clearance, $\nu$ is the kinematic viscosity of the slurry ($\text{m}^2/\text{s}$), $g$ is the gravitational acceleration constant ($\text{m/s}^2$), $\rho_S$ and $\rho_L$ are the densities of the solid and liquid phases, respectively ($\text{kg/m}^3$), $d_p$ is the particle diameter ($\text{m}$), $D$ is the impeller diameter ($\text{m}$) and $X$ is the ratio of mass solids to mass liquid (Zwietering, 1958).

Though many others have proposed correlations to determine $N_{JS}$, the Zwietering correlation is the most widely used and tested one and, therefore, is the only one discussed in this work.
The Zwietering correlation is purely empirical and while a few studies have largely corroborated Zwietering’s findings (Nienow, 1968; Raghava Rao et al., 1988; Mak, 1992), others have identified conditions at which the correlation is not reliable (Choudhury, 1997). Myers et al (2012) examined the effect of solid loadings on N_{JS} and found that the exponent on X increased with increasing solids concentration from 0.097 at 5 wt% to 0.34 at 40 wt%. Thus, it is clear that the validity of the Zwietering correlation is untested for high solids concentrations. As recent studies have shown that operating solid-liquid agitated systems at higher solids concentration (> 0.20 (v/v)) achieved greater energy efficiency (Wu et al., 2002; Wu et al., 2010b; Wang et al., 2012a), determining the reliability of the Zwietering correlation in such systems would be useful for process intensification purposes.

The effect of geometry is accounted for by the constant, S and the impeller diameter, D in the Zwietering correlation. Especially, S is a function of impeller type and diameter, and its off-bottom clearance. The possible combination of these parameters is infinite. In addition to Zwietering, other researchers have also reported S values for a wide range of impeller and tank geometries (Ibrahim and Nienow, 1996; Armenante and Nagamine, 1998; Atiemo-Obeng et al., 2004; Ayranci and Kresta, 2011). However, a majority of these studies were confined to baffled vessels. Therefore, the reliability of the Zwietering correlation in determining N_{JS} in unbaffled vessels is still unclear.

Moreover, information is scarce on the reliability of the Zwietering correlation for solid-liquid agitated systems with multiple impellers. Mak (1992) reported that N_{JS} for a solid-liquid agitated system fitted with a second impeller decreased or increased depending on impeller spacing due to the degree of interaction between liquid flows generated by the upper and lower impellers. Dutta and Pangarkar (1995) studied the effect of multiple impellers on N_{JS} in baffled vessels where the number of impellers was equal to the ratio of liquid height to vessel diameter. They concluded that the Zwietering correlation could be used to predict N_{JS} in multiple impeller systems though with some changes in the exponents. However, they did not report any S values for their system. There is a lack of information on the suitability of the Zwietering correlation to unbaffled vessels fitted with multiple impellers.

In summary, the Zwietering correlation is the most widely used correlation to estimate the critical impeller speed. Though the correlation is based on a very large number of experiments and is dimensionless, it should not be applied to solid-liquid systems outside its
tested range due to its empirical nature. So, it would be useful to determine the reliability of the correlation in solid-liquid systems other than those used to derive it.

2.1.3 Particle suspension in taller tanks involving multiple impellers

Industrial agitated vessels with a height to diameter ratio of > 1 are usually fitted with multiple impellers (Kadic and Heindel, 2010). The installation of additional impellers is to introduce an even distribution of shear and energy dissipation throughout the vessel. In a multiple-impeller arrangement, the bottom impeller is mainly responsible for particle suspension while the other impellers distribute solids throughout the tank resulting in enhanced overall mixing (Armenante and Li, 1993). They reported that impeller configurations that act to complement the flow produced by the bottom impeller results in a lower impeller speed for solids suspension, however, distortion in hydrodynamic flow patterns leads to chaotic mixing and higher energy requirements to achieve the same level of solids suspension. Other researchers have reported similar findings (Nocentini et al., 1988; Dutta and Pangarkar, 1995). Bakker et al. (1998) also showed that the difference in energy requirement for single- and dual-axial impeller systems was marginal. Using two A310 impellers, Wang et al. (2014) found that the presence of a second impeller led to higher energy consumption to attain N_{JS} conditions. They also found that the difference in energy consumption for single- and dual-A310 impeller systems decreased with increasing solids concentrations.

Numerous studies on solids suspension focused on parameters associated with vessel geometry and impeller configuration such as liquid height to diameter ratio, number and type of impellers as well as their geometric positions in the tank (Bujalski et al., 1999; Gogate et al., 2000; Angst and Kraume, 2006). Armenante et al. (1992) found that the influence of impeller diameter on N_{JS} did not significantly change with the number of impellers mounted on a shaft. However, they found that the clearance of the lowest impeller from the vessel bottom did have a significant role in determining N_{JS}. Impeller spacing and location were found to play important roles in determining rates of mass transfer and impeller power required for solids suspension (Saravanan et al., 1997). While it is generally accepted that additional impellers would result in increased power consumption, information on solids suspension in agitated vessels with multiple impellers operating at high solids loadings is
limited (Tatterson, 1994; Dutta and Pangarkar, 1995; Kasat and Pandit, 2005; Wang et al., 2014).

2.1.4 Cloud height and particle dispersion

Particle distribution in solid-liquid agitated vessels can be at varying degrees of homogeneity. When the solid phase is uniformly distributed throughout the stirred vessel, the degree of suspension is said to be homogenous. This level of suspension is usually more energy intensive than off-bottom suspension (degree of solids suspension at which just enough agitation is applied to just suspend all particles off the tank bottom) and not required in many industrial processes. However, in processes such as multiphase catalytic reactions and crystallization, off-bottom suspension is not sufficient and a greater degree of particle dispersion is required (Hicks et al., 1997; Bujalski et al., 1999). The extent of research in the area of solids distribution is limited compared to that in the area of solids suspension. This is mainly due to the difficulty in developing a reliable experimental technique and a measure to quantify particle distribution.

There are numerous methods of measuring local solids concentrations in slurry. The optical method has been widely used to characterise solids distribution in agitated vessels (Magelli et al., 1990; Magelli et al., 1991). This method is non-intrusive but is generally limited to solids concentrations of < 2% (v/v) due to scattering and blocking of light by particles between the source and receiver. Other methods include withdrawing samples at various locations in the vessel and conductivity measurement. These methods are intrusive but due to their simplicity, they were widely used (Barresi and Baldi, 1987; Considine and Considine, 1985; Spidlpa et al., 2005). The local solids concentration can be compared to average solids concentration to determine the distribution of solids concentration. Quantifying particle distribution is often quoted by measuring the relative standard derivation (RSD), which is the deviation of the local solids concentration from the average solids concentration.

Studying highly concentrated slurries at homogeneous conditions, Buurman et al. (1986) reported no difference in solids concentration at 3 sample points in the axial direction. Later, researchers reported similar findings where the axial solids concentration is roughly constant throughout the slurry suspension (Hicks et al., 1997; Wang et al., 2016). Studies on the radial
solids concentration gradient in agitated vessels have suggested that it is usually negligible (Yamazaki et al., 1986; Barresi and Baldi, 1987). However, subsequently a few studies have indicated the presence of a radial solids concentration gradient and its dependence on particle size, solids loading and impeller type (Micheletti et al., 2003; Spidla et al., 2005).

Modelling solids distribution in stirred vessels has been attempted by several authors using low concentration slurry. Several fluid dynamic models have been adopted in describing particle distribution, including one dimensional sedimentation dispersion model (Barresi and Baldi, 1987; Magelli et al., 1990; Shamlo and Koutsakos, 1989), multi-zone sedimentation dispersion model (Yamazaki et al., 1986), and two-or-three-dimensional network of zones model (Brucato et al., 1991). In addition to these models, more recent studies used computational fluid dynamics (CFD) simulation models (Khopkar et al., 2006; Guha et al., 2008).

Studies on impeller power consumption to achieve complete particle dispersion have been reported in the literature. Hicks et al. (1997) investigated the effects of different impeller and tank geometries and solids concentration on impeller power consumption. They showed that, while operating at $N_{JS}$, solids dispersion level decreased with increasing solids concentration up to a value of 0.40 wt% and increased thereafter. Recently, Wang et al. (2016) proposed a simple model using known values of volumetric solids concentrations ($C_V$) to estimate the impeller power consumption to achieve a homogenous suspension. Their work incorporated different impeller types and unbaffled vessels which showed that axial flow impellers were the most efficient at dispersing solids up to $C_V = 0.2$ (v/v). In another study, Wu et al. (2011) observed that a lower specific power input was required for particle dispersion with the removal of baffles when operating at very high solids concentration ($C_V = 0.4$ (v/v)).

2.2 Impeller power consumption

In solid-liquid mixing systems, impellers are used to supply energy to the system to achieve solids suspension or solids dispersion. Reporting impeller speed rather than the impeller power consumption for complete suspension of solids does not allow the comparison of energy efficiency reported in different studies (Wang et al., 2014). Therefore, it is highly desirable to compare solid-liquid mixing systems in terms of specific impeller power.
consumption at $N_{JS}$ to identify efficient processes. Impeller power consumption in solid-liquid mixing vessels is a fundamental measurement of the vessel performance and is a key design parameter. In solid-liquid agitated vessels, the impeller power consumption is dependent on a range of factors including size, speed, position and shape of the impeller, fluid and solid properties such as density and viscosity as well as vessel configuration. As the operating cost is directly proportional to the impeller power consumption, there is a strong demand from industry to intensify existing solid-liquid systems so that more solids can be processed with minimal energy input. Process intensification can be achieved by increasing the solids throughput or by optimising process equipment to enable more efficient mixing (Wu et al., 2010a). When operating at high solids concentration, increased impeller speeds and power consumptions are required to achieve off bottom solids suspension. Altering the impeller and vessel geometry such as impeller and baffle arrangement may aid in achieving process intensification. Recently, it has been reported that operating solid-liquid agitated vessels at higher solids concentrations leads to an increase in impeller energy efficiency as more solids are suspended per unit power input.

2.2.1 Effect of solids concentration on impeller power consumption

Solids concentration has a significant influence on the energy input in a solid-liquid agitated vessel. An increase in solids concentration leads to a decrease in the liquid circulation velocity and an increase in energy dissipated at the solid-liquid interface (Raghava Rao et al., 1988). To compensate for the energy loss due to solid-liquid friction, particle-particle friction and particle-equipment friction, additional energy is required (Bubbico et al., 1998). So, an increase in solids concentration leads to an increase in power consumption to attain $N_{JS}$ conditions. It has been reported that the impeller power consumption to achieve off bottom suspension ($P_{JS}$) increased exponentially at very high solids concentrations (>0.40 (v/v)) (Drewer et al., 2000; Wu et al., 2002; Wang et al., 2014). Raghava Rao et al. (1988) and Bubbico et al. (1998) presented similar findings but added that at very low solids concentrations (<0.04 (v/v)), the amount of energy loss is negligible.

Many investigators reported specific impeller power input on the basis of the vessel volume (Drewer et al., 1994; Wu et al., 2002). Drewer et al. (2000), on the other hand, proposed specific power input on the basis of mass of solids suspended stating that the rate of reaction
or mass transfer is independent of further increase in agitation or vessel volume once off-bottom solids suspension is achieved. They found that when the specific power consumption at $N_{JS}$ per unit mass of solids suspended ($P_{JS}/M_S$) is plotted against the solids concentration, the curve on the graph had a minimum value around a solids concentration of 0.30 (v/v). Similar findings were observed by Raghava Rao et al. (1988), who found that $P_{JS}/M_S$ decreased with increasing solids concentration. Subsequent studies by researchers using this concept found that, in $P_{JS}/M_S$ versus solids concentration plots, a minimum value for $P_{JS}/M_S$ occurs at around a solids concentration of 0.20 (v/v) and therefore a significant energy savings can be achieved by operating solid-liquid agitated systems at this solids concentration because more solids can be suspended per unit power input (Wu et al., 2010b; Wang et al., 2014; Bong et al., 2015).

2.2.2 Effect of impeller type on impeller power consumption

The impeller type has a crucial role in determining the power consumption required for solids suspension. This is because different impellers generate different flow patterns leading to different hydrodynamic conditions in the vessel thus affecting the impeller energy efficiency. Generally, impellers are classed as axial or radial flow impellers or a combination of both. The flow patterns of axial and radial flow impellers are completely different and therefore lead to different solid suspension mechanisms (Kresta and Wood, 1993; Bittorf and Kresta, 2000; Bittorf and Kresta, 2003).

Radial flow impellers, such as disc turbines or Rushton turbine (RT) generate liquid flow with radial direction movement and splits into two streams at the vessel wall. The streams generate circulation loops, one above and one below the impeller. The lower circulation loop reaches the tank bottom and flows towards the centre of the tank from where the solids are suspended. Axial flow impellers such as Lightnin A310 and mixed flow impellers such as a down pumping pitched blade turbine (PBT) generate liquid flows towards the tank bottom where they push the solids towards the periphery of the tank before suspending them.

Extensive studies have been conducted in determining a suitable impeller design to minimise energy consumption for solids suspension. It is well established that lower power number impellers such as A310 are more economical in baffled vessels in achieving $N_{JS}$ (Nienow,
However, a higher power number impeller such as the Rushton turbine was reported to be more energy efficient in unbaffled vessels (Wu et al., 2011; Wang et al., 2012b; Wang et al., 2014). Wu et al. (2010b) confirmed the previous findings reporting that axial flow impellers are more energy intensive than radial flow impellers under unbaffled conditions. They also demonstrated that the impeller power consumption to achieve off-bottom solids suspension decreased with the removal of baffles. Additionally, they found that a power saving of ≈70% could be achieved under unbaffled conditions even with relatively very high solids concentration (>0.40 (v/v)).

### 2.2.3 Effect of baffles on impeller power consumption

Baffles are installed in mechanically agitated vessels to prevent swirling and vortexing of liquid in the tank. It is generally agreed that the installation of baffles enhances axial and radial mixing as well as levels of turbulence (Nagata, 1975; Pangarkar et al., 2002). However, insufficient or excessive baffling may result in the reduction of mass flow and localisation of liquid flow in the vessel (Nishikawa et al., 1979). Also, excessive baffling would interrupt liquid mixing and lengthen mixing time, thus reducing the efficiency of the agitation system (Lu et al., 1997; Tagawa et al., 2011).

Unbaffled mixing vessels generally consume less power than baffled systems for suspending solids off the tank bottom (Markopoulos et al., 2005; Wu et al., 2010b; Tagawa et al., 2011). Liquid flows produced by the impeller in unbaffled agitated vessels move in a circular trajectory where the tangential velocities lead to lower impeller power draw than in baffled vessels (Assirelli et al., 2008). Vortexing also occurs in unbaffled mixing vessels. If it reaches the impeller, the impeller power draw decreases due to the impeller blade’s exposure to air (Scargiali et al., 2013). In addition to the significant reduction in the impeller power consumption required to suspend solids, the removal of baffles also lead to increased mixing time. However, Wu et al. (2010b) points out that the residence time in some mineral processing operations is much longer than the extended mixing times associated with baffle removal.
2.3 Solid-liquid mass transfer

Many industrial processes often involve interphase mass transfer at the solids surface while dispersed in a liquid phase. Mass transfer can be characterised from a microscopic or macroscopic perspective. The study of physical mass transfer is macroscopic and only this perspective of mass transfer will be discussed in this study.

Effective mass transfer depends on many parameters including external parameters such as vessel and impeller geometry as well as impeller rotation speed. Parameters associated with the solid and liquid phases such as density, viscosity, particle size and shape all have an impact on mass transfer. In addition, hydrodynamics of flow and degree of solids suspension, which in turn, depend on levels of turbulence, also influence mass transfer. As turbulence intensity varies from point to point in the vessel, it is difficult to establish a uniform hydrodynamic environment or to conduct a mathematical analysis (Bong et al., 2015).

The complex nature of the flow patterns generated and the influence of several variables permit the measurement of only an average mass transfer coefficient for a given solid-liquid mass transfer system. Extensive studies have been conducted investigating solid-liquid mass transfer using a variety of geometries and operating conditions. In this section, a brief review of some of the parameters that affect mass transfer will be presented.

2.3.1 Solid-liquid mass transfer coefficient

Solid-liquid mass transfer is interphase migration of molecules due to a chemical potential. Effective mass transfer depends on physical processes such as mixing and factors affecting the interaction between the solid and liquid phases. The hydrodynamic environment around a particle controls the diffusivity of a solute either to or from the solid surface. Diffusion mass transfer is considered to occur via two mechanisms:

1) Molecular diffusion by microscopic movement of individual molecules; and
2) Eddy or turbulent diffusion by macroscopic fluid motion.

These mechanisms are the means by which mass transfer between the solid and liquid phases occur. The most common example of solid-liquid mass transfer is the dissolution of solids
where mass is transported from the solid surface into the adjacent liquid layer. The solid particle decreases in size as it is incorporated as solute in the solvent. On the other hand, in adsorption and desorption operations, mass transfer occurs between the solid and liquid phases due to a concentration difference. In ion-exchange systems mass transfer occur via an exchange of ions between the two phases with ion affinity playing an important role. Leaching processes involve the dissolution of the soluble component of the solid, leaving behind a solid of different size, structure or porosity.

In solid-liquid mixing operations, the rate of diffusion in solid-liquid mass transfer is highly dependent on liquid turbulence. As the liquid becomes more turbulent, the diffusional boundary layer decreases leading to an increased rate of mass transfer (Sterbacek and Tausk, 1965).

As solid-liquid mass transfer is film diffusion controlled, the rate of mass transfer is dependent on the refresh rate as well as the size of the boundary layer around the particle. The diffusional mass transfer, M is defined by the following equation:

\[ M = k_{SL}a_p(C_s - C_L) \]  \hspace{1cm} (2.3)

where \( k_{SL} \) is the solid-liquid mass transfer coefficient, \( a_p \) is the interfacial surface area per unit volume and \( (C_s - C_L) \) is the concentration driving force. \( C_s \) is the solute concentration at the solid surface and \( C_L \) is the solute concentration in the solvent. The interfacial surface area per unit volume can be defined by:

\[ a_p = 6 \frac{C_v}{d_{32}} \]  \hspace{1cm} (2.4)

where \( C_v \) is the volume fraction of solids in the slurry and \( d_{32} \) is the Sauter-mean particle diameter. From equation (2.3), it is clear that the mass transfer rate can be increased by increasing \( k_{SL} \), the driving force \( (C_s - C_L) \) or \( a_p \). The solid-liquid mass transfer coefficient, \( k_{SL} \) is affected by many variables including physical properties, geometrical parameters, and solid-liquid interactions.
2.3.2 Effect of solids concentration on the solid-liquid mass transfer coefficient

While there are numerous studies on solid-liquid mass transfer, a majority of them involve dissolution processes containing highly inert particle concentrations or systems restricted to solids concentration of less than 0.01 (v/v) (Kato et al., 1998; Kasat and Pandit, 2005; Tagawa et al., 2011). Harriot (1962) showed that the mass transfer coefficient was not affected by solids concentration up to 0.053 (v/v). Similar observations were reported by Lal et al. (1988) up to a solids concentration of 0.1 (v/v).

A correlation relating solids concentration with the solid-liquid mass transfer coefficient was developed by Cline (1978) who suggested that the mass transfer coefficient would decrease with increasing solids concentration. Cline (1978) used ion exchange resins suspended at constant impeller speed and varied the solids concentration from 0.05 to 0.40 (v/v). He did not observe any significant effect of solids concentration on mass transfer coefficient at constant impeller power input. Harriot (1962) also developed a correlation to predict the solid-liquid mass transfer coefficient up to 0.3 (v/v). He predicted that the mass transfer coefficient would increase with increasing solids concentration but could not validate his predictions with experimental data. The experimental conditions used in obtaining solid-liquid mass transfer coefficient values by these researchers are shown in Table 2.1.

More recent investigations have suggested that operating at relatively high solids concentrations led to improved energy efficiencies as well as enhanced mass transfer (Wu et al., 2010a; Wang et al., 2012a). Bong et al. (2015) reported that optimum mass transfer and impeller power consumption occurred at a solids concentration of 0.2 (v/v) whilst operating the stirrer at $N_{JS}$. These studies utilised vessels agitated with a single impeller, therefore, the effect solids concentration and tank geometry on mass transfer in dual-impeller systems is lacking.

2.3.3 Effect of baffles on the solid-liquid mass transfer coefficient

The hydrodynamic flow produced by an impeller in a stirred vessel contributes to convective mass transfer. Therefore, baffles are widely used in industrial agitated vessels to achieve better mixing due to higher levels of turbulence generated under baffled condition (Nagata,
Baffles promote liquid circulation by increasing the axial liquid velocity and reducing the swirl velocity. Higher relative liquid velocities lead to better solids distribution and thus promote effective contact between the solid surface and the surrounding liquid thereby enhancing mass transfer.

The effect of baffles on mass transfer coefficient at high impeller speeds have been investigated by several researchers (Hixson and Wilkens, 1933; Barker and Treybal, 1960; Lal et al., 1988). They reported that under baffled conditions, solids were fully suspended due to greater levels of liquid velocity fluctuations. However, baffles lead to the formation of stagnant regions behind baffles. Particles tend to accumulate in these regions thereby leading to ineffective solid-liquid mass transfer between the particles involved and the surrounding liquid.

Levins and Glastonbury (1972) reported that the effect of impeller speed on the mass transfer coefficient was less pronounced under unbaffled conditions than under baffled conditions. Nienow et al. (1997) reported that mass transfer coefficient was lower in unbaffled vessels due to vortex formation occurring resulting in surface aeration. It was also reported that when operating at partially suspended conditions, unbaffled condition leads to higher mass transfer coefficient values than those obtained under baffled condition. This was ascribed to the fraction of particles suspended at low speeds being greater under unbaffled condition than that under baffled condition. However, in the case of fully suspended condition, baffled vessels were reported to lead to higher or similar mass transfer coefficient values (Harriott, 1962; Barker and Treybal, 1960). Other studies have shown that whilst operating at high solids concentrations, unbaffled tanks were more energy efficient in suspending particles while also obtaining similar or slightly lower mass transfer coefficient values than those obtained under baffled conditions (Wu et al., 2010a; Tagawa et al., 2011; Bong et al., 2012; Wang et al., 2012a).

### 2.3.4 Effect of impeller type on the solid-liquid mass transfer coefficient

As mention in Section 2.2.2, impellers are normally classified as radial, mixed or axial flow impellers (Sterbacek and Tausk, 1965; Uhl and Grey, 1966). The selection of impellers is largely dependent on the application or process objective as different impellers produce
different liquid flow patterns and hydrodynamic environments. There have been numerous studies into the effect of impeller type on the solid-liquid mass transfer coefficient. It has been reported that different impellers produce different levels of solids suspension and turbulence at the same location in the vessel, and therefore influence diffusional mass transfer differently (Pangarkar et al., 2002).

Studies have shown that radial flow impellers generate higher levels of turbulence than mixed or axial flow impellers and in turn achieve higher solid-liquid mass transfer coefficient values (Nienow and Miles, 1978; Chapman et al., 1983; Jadhav and Pangarkar, 1991; Zhou and Kresta, 1996). Nienow and Miles (1978) reported that at constant impeller speed, radial flow impellers achieved higher solid-liquid mass transfer coefficient values than axial flow impellers. However, at complete suspension conditions, the solid-liquid mass transfer coefficient values obtained using either radial or axial flow impellers were similar. Based on impeller power consumption, Doraiswamy and Sharma (1984) suggested the use of a 45° pitched blade turbine or a propeller to achieve a desired mass transfer coefficient.

2.3.5 Effect of multiple impellers on solid-liquid mass transfer coefficient

Multiple impeller agitated vessels are used when shear sensitive and high viscosity materials need to be handled. Operations requiring a high surface-to-volume ratio also utilises vessels agitated with multiple impellers. As Paul et al. (2004) stated, the fluid velocity decreases with increasing distance from the impeller region and therefore multiple impellers are needed to induce greater levels of turbulence in the upper regions of the vessel and increase overall mass transfer. Also, the flows generated from each impeller may interfere with each other leading to increased levels of chaotic mixing and in turn increased levels of mass transfer (Dutta and Pangarkar, 1995). El Shazly (2016) measured mass transfer (corrosion) rates from the vessel wall of a dual-impeller agitated vessel and reported that mass transfer rate was dependent on vessel/impeller geometry and that it increased with increasing $C_v$. Although widely used in industry, research into solid-liquid mass transfer in agitated vessels with multiple impellers is scarce. Much of the work reported in the literature focused on solid-liquid mass transfer in three-phase single-impeller systems (Dutta and Pangarkar, 1996; Pangarkar et al., 2002; Winterbottom et al., 2003).
Dutta and Pangarkar (1994) studied solid-liquid mass transfer in solid-liquid vessels agitated with multiple impellers. They derived a correlation to predict the solid-liquid mass transfer coefficient and found that it applies equally well in single-impeller systems at $N_{JS}$. The solids concentration used in their study was not more than 0.5 wt%. So, the effect of high solids concentration on solid-liquid mass transfer coefficient in multiple impeller systems is still lacking.

### 2.4 Correlation for solid-liquid mass transfer coefficient

There are many correlations reported in the literature for estimating the solid-liquid mass transfer coefficient in agitated vessels (Harriott, 1962; Levins and Glastonbury, 1972; Kato et al., 1998). Due to many variables that affect the solid-liquid mass transfer coefficient and the complex nature of liquid flow patterns, most of the work on correlations is only applicable to systems similar to the one used to develop the correlation. For this reason, the dimensional analysis has become the method of choice in developing a correlation to estimate the solid-liquid mass transfer coefficient, $k_{SL}$. The most general form of the correlation to estimate $k_{SL}$ is

$$Sh = 2 + ARe^m Sc^n$$  \hspace{1cm} (2.5)

where $Sh$ is the Sherwood number, $Re$ is the Reynolds number, $Sc$ is the Schmidt number, $A$ is a constant, and $m$ and $n$ are exponents.

Sherwood number ($Sh$) is the ratio of convective mass transfer to diffusive mass transfer and is defined as:

$$Sh = \frac{k_{SL}d_p}{D_A}$$  \hspace{1cm} (2.6)

where $k_{SL}$ is the solid-liquid mass transfer coefficient, $D_A$ is the molecular diffusivity and $d_p$ is particle diameter.

Schmidt number ($Sc$) is the ratio of kinematic viscosity to molecular diffusivity:

$$Sc = \frac{\mu_L}{\rho_L D_A}$$  \hspace{1cm} (2.7)
where $\mu_L$ and $\rho_L$ is the liquid viscosity and liquid density, respectively. $Sc$ also represents the ratio of the thicknesses of hydrodynamic layer to boundary layer.

Impeller Reynolds number ($Re$) is the ratio of inertial to viscous forces and is given as:

$$Re = \frac{ND^2\rho_L}{\mu_L} \quad (2.8)$$

where $N$ is the impeller speed, $D$ is the impeller diameter and $\mu_L$ and $\rho_L$ are the liquid viscosity and liquid density, respectively.

Mass transfer coefficient correlations depend on several factors as mentioned previously, which has led to differences in experimental and predicted mass transfer coefficient values (Kuboi et al., 1974; Lal et al., 1988; Grisafi et al., 1998). In addition, three approaches have been used to determine Reynolds number used in the correlation to estimate the solid-liquid mass transfer coefficient. The first approach is the use of impeller tip velocity, the second uses slip velocity while the third utilises Kolmogoroff theory of isotropic turbulence. Only the Kolmogoroff theory of isotropic turbulence approach in determining Reynolds number will be used in this work.

### 2.4.1 Reynolds Number calculation using Kolmogoroff theory of isotropic turbulence

Kolmogoroff theory (1941) is widely accepted due to its simplicity and its ability to relate turbulence, power input, and solids concentration to the rate of mass transfer. The theory proposes that large eddies are formed due to the rotating impeller, generating turbulence. These eddies eventually disintegrate into smaller eddies as it moves away from the impeller until all the energy is dissipated as viscous flow. As these eddies break down further into ‘terminal eddies’, the directional element of the flow is lost and the turbulence is said to be isotropic. Hinze (1975) discusses the theory further in detail.

Reynolds number using this approach is calculated using:

$$Re_K = \frac{d_p^{4/3} \varepsilon^{1/3}}{\nu} \quad (2.9)$$

where $\varepsilon$ is the rate of energy dissipation per mass of liquid (W/kg) and $\nu$ is kinematic viscosity of the liquid (m$^2$/s).
Using Eq. (2.9) to calculate Reynolds number and incorporating it into the functional relationship between Sherwood number, Reynolds number and Schmidt number (equation 2.5), the following equation is obtained:

\[ Sh = 2 + ARe^\frac{m}{n}Sc^n \quad (2.10) \]

This correlation suggests that Sherwood number and in turn the solid-liquid mass transfer coefficient \((k_{SL})\) is a function of power input per mass of liquid \((\varepsilon)\), which is incorporated in \(Re\). Although the local energy dissipation rate varies throughout the vessel, which means the mass transfer coefficient will also vary throughout the vessel, determining the average power input per mass of liquid will help in predicting the average mass transfer coefficient (Pangarkar et al., 2002). Also, it has been reported that a constant mass transfer coefficient value was obtained for a wide range of specific power input values at \(N_{JS}\) (Nienow and Miles, 1978).

### 2.4.2 Limitations to mass transfer coefficient correlations

Due to complex flow patterns in a solid-liquid agitated system, the solid-liquid mass transfer coefficient varies with tank and impeller geometry and other parameters associated with the liquid and solid phases used in the mixing system. This has led to researchers determining the mass transfer coefficient based on a dimensional approach including impeller geometry, impeller speed and type, diffusivity, viscosity, density difference and particle diameter. Hence, differences in experimental and estimated mass transfer coefficient values have been reported. It means that the correlations developed from experimental results are usually only applicable to solid-liquid systems for which they have been developed. A summary of operating parameters and correlations developed by different researchers are listed in Table 2.1.
<table>
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<th>Approach to calculate Re</th>
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<td>Harriott (1962)</td>
<td>Ion exchange resin</td>
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<td>T = 0.07 – 0.53m;</td>
<td>6 bladed disc</td>
<td>0.001 – 0.053 (v/v)</td>
<td>Slip velocity</td>
<td>$Sh = 2 + 0.6Re^{0.5}Sc^{0.33}$</td>
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<td></td>
<td></td>
<td>glycerol;</td>
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<td>Levins and Glastonbury (1972)</td>
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<td>$Sh = 2 + 0.5Re^{0.203}Sc^{0.33}$</td>
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<td>baffled and unbaffled</td>
<td>bladed turbine; marine propeller</td>
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<tr>
<td>Cline (1978)</td>
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<td>NaOH</td>
<td>T = 0.16m;</td>
<td>4 bladed pitch</td>
<td>0.05 – 0.40 (v/v)</td>
<td>Slip velocity</td>
<td>$Sh = 2 + 0.95Re^{0.5}Sc^{0.33}$</td>
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<td>Lal et al. (1988)</td>
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<td>baffled and unbaffled</td>
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<td></td>
<td>$Sh = 2 + 7.53Re^{0.25}Sc^{0.33}$ for Re&gt;800</td>
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<tr>
<td>Study</td>
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<tr>
<td>Jadhav and Pangarkar (1991)</td>
<td>Benzoic acid</td>
<td>Water + CMC</td>
<td>0.15 – 0.30 m</td>
<td>Disc turbine; 4 &amp; 6 bladed pitch turbine (upward)</td>
<td>0.05 (v/v)</td>
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<tr>
<td>Dutta and Pangarkar (1994)</td>
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<td>Water + CMC</td>
<td>0.15 – 0.30 m</td>
<td>Disc turbine; 6 bladed pitch turbine; multiple impeller setup</td>
<td>0.5 (wt/wt)</td>
<td>$k_{SL} = 1.34 \times 10^{-3}$ (N/N_{JS})^{1.07}S_{c}^{-0.52}</td>
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<tr>
<td>Kato et al. (1998)</td>
<td>Ion exchange resin</td>
<td>NaOH; KOH; Ca(OH)$_2$</td>
<td>0.12 – 0.17 m</td>
<td>Shaking vessel with current pole</td>
<td>0.01 (v/v)</td>
<td>$Sh = 0.112Re^{0.29}S_{c}^{0.33}$</td>
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<tr>
<td>Bong et al. (2015)</td>
<td>Ion exchange resin</td>
<td>NaOH</td>
<td>0.2 – 0.3 m</td>
<td>6 bladed disc turbine</td>
<td>0.05 – 0.40 (v/v)</td>
<td>$Sh = 2+(-aRe^{0.3}+b-cRe^{-0.5})Re^{0.5}S_{c}^{0.33}$ where a, b and c is 3.7, 56.7, 211.3 for T = 0.2m and 2.3, 32.4, 109.5 for T = 0.3m, respectively</td>
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</table>
2.5 Ultrasound and Cavitation

Ultrasound are sound waves that occur at frequencies higher than the audible frequency of the average human ear, and is typically in the range of 20 kHz to 500 MHz. As the ultrasonic frequency is inversely proportional to power output, low frequency ultrasound is used in sonochemical applications (Thompson and Doraiswamy, 1999). These sound waves are mechanical oscillations in time and space that cause molecules in a medium to vibrate about their mean position (Kuttruff, 1991). As the wave moves through the liquid medium, it compresses and stretches the molecular spacing between the molecules. The negative pressure created during the rarefaction stage, if large enough, causes the liquid to break apart creating voids in the liquid medium. These voids generally occur in ‘weak spots’ or in ‘nucleation sites’ and are called cavitation bubbles (Mason, 1992).

Cavitation can be described as one of two types, stable cavitation and transient cavitation (Atchley and Crum, 1988). Stable cavitation involves bubbles that are formed at low ultrasonic intensities and oscillate about some equilibrium size over many acoustic cycles. There is little bubble growth and these bubbles can lead to transient cavities or dissolve back into the solvent. Transient cavitation on the other hand, are formed at high ultrasound intensities and expand and compress during only a few acoustic cycles until the size of the bubble reaches a maximum before violently collapsing (Mason, 1990).

The chemical effects of ultrasound referred to as sonochemistry are attributed to the collapse of both stable and transient cavitation. Two competing theories exist to explain cavitation’s effect on chemical processes:

1) The hot spot theory; and
2) The electrical theory

The hot spot theory postulates that when the cavity implodes, localised hot spots as well as intense pressures are formed in the order of 5000K and 200 atm, respectively (Suslick, 1990; Mason, 1999). These extreme localised conditions instigate the formation of free radicals that act as catalyst to initiate chemical reactions (Mason, 1999). The electrical theory postulates that an electrical charge is created on the cavity surface, forming an electrical field gradient across the cavity, which is then able to initiate chemical reactions (Margulis, 1984).
However, Lepoint-Mullie et al. (1996) discounted the electrical theory as a valid mechanism for sonochemistry and therefore the hot spot theory is more widely accepted.

2.5.1 Cavitation and solid-liquid mass transfer

Solid-liquid mass transfer enhancement has been attributed to the physical effects of ultrasound, which are acoustic streaming and acoustic cavitation. Acoustic streaming is the non-cavitating, sonically induced movement of liquid due to the conversion of sound energy into kinetic energy (Hamdaoui and Naffrechoux, 2007). Acoustic cavitation, on the other hand, is the violent collapse of a transient bubble, which produces several phenomena that enhances solid-liquid mass transfer. Cavitation is a physical phenomenon that is affected by the frequency and intensity of the ultrasonic wave, the type and temperature of the solvent, and the presence of dissolved gasses. These parameters have an effect on cavitation as they all affect the cohesive properties of the liquid molecules and/or the vapour pressure of the solvent.

One of the phenomena that enhance solid-liquid mass transfer can be attributed to the production of microjets. Microjets occur if the bubble collapse is close enough to the absorbent surface where the microjet impacts the particle at a velocity of several hundred metres per second (Mason, 1991). This microjet of liquid can travel into a porous particle or produce newly exposed surfaces for interphase mass transfer. High-pressure shock waves can also be produced from cavitating bubbles situated away from any solid-liquid boundary layer. These shockwaves cause increased mass transfer by decreasing the diffusion layer thickness at the solid surface (Leong et al., 2011). Another cavitation event that enhances mass transfer at the solid-liquid interface is microstreaming or the formation of micro eddies (Hamdaoui and Naffrechoux, 2007). Microstreaming enhances mass transfer by decreasing the thickness of the boundary layer around the particle. The performance of these cavitation events produced by acoustic cavitation is dependent on a range of ultrasonic and solid-liquid parameters.

There have been many studies in the literature to investigate the use of ultrasound as a means to improve interphase mass transfer in solid-liquid systems (Schueller and Yang, 2001; Breitbach et al., 2003; Hamdaoui et al., 2003). Ultrasound has been shown to increase the
dissolution rates of minerals from ores such as galena, crocoite and arsenolite by 20, 30 and 50 times, respectively, when compared to traditional leaching methods (Swamy and Narayana, 2001). The leaching of copper and other metals from their associated ores has been found to be enhanced with ultrasound (Sarveswara Rao et al., 1997; Grennan et al., 2007; Razavizadeh and Afshar, 2008). The adsorption rate of various compounds including organics onto activated carbon in the presence of ultrasound has also been investigated and found to be enhanced when compared to systems without ultrasound (Hamdaoui et al., 2003; Juang et al., 2006; Milenkovic et al., 2013). Other studies have shown that the elution of organic compounds from absorbents can also be accelerated by ultrasound (Hamdaoui and Naffrechoux, 2007). Schueller and Yang (2001) who studied the adsorption and desorption of phenol from activated carbon and polymeric resin, found that cavitation promoted desorption by decreasing the activation energy for surface diffusion as well as generating localised heating.

Although the physical effects associated with cavitation is proposed as the cause for increased mass transfer rates, the mechanism by which it is achieved varies. Bassler et al. (1997) has suggested that enhanced desorption from polymeric resin resulted from acoustic cavitation. Other studies have suggested that increased desorption rates of phenol from activated carbon and polymeric resin was due to cavitation and acoustic vortex microstreaming (Rege et al., 1998; Schueller and Yang, 2001). Qin et al. (2001) postulated that desorption of organic adsorbates from various resins was accelerated due to thermal effects. On the other hand, Hamdaoui et al. (2005) suggested that accelerated desorption rate was due to a combination of thermal and non-thermal effects. While these studies identified an increase in mass transfer when operating at very low solids concentrations (<0.04 (v/v)), the influence of ultrasound on solid-liquid mass transfer in slurries with higher solids concentration, which is more relevant for industrial operations, is still unclear.

Moreover, information is scarce on the effect of ultrasound on mass transfer in an ion exchange system. While applying an ultrasonic field to a CaCl₂ solution containing 13X zeolite with exchangeable Na⁺ cations, Kiezel et al. (1977) found that the ion exchange rate was greater than that found with mechanical mixing alone. They found that initial exchange rates were rapid and similar in both ultrasonic and silent conditions before witnessing an increased exchange rate in the ultrasonic system. They attributed this rate increase to the greater periodic vibrational potential of the sodium ion which increases its diffusion rate through zeolite. Also, when using 13X zeolite in LiCl (aq), CaCl₂ (aq) and CeCl₃ (aq)
solutions, Erten-Kaya and Cakicioglu-Ozkan (2012) found that the initial rate of exchange was very fast followed by a slow rate under both ultrasonic and silent conditions. They also found that the amount exchanged at equilibrium increased in the ultrasonic system, and they were able to fit the experimental data to kinetic models. Entezari and Tahmasbi (2009) also observed that, when ultrasound was applied, the ion exchange rate increased compared to agitation alone. When using very low solid loadings (0.0016 and 0.004 (v/v)), they found that the amount exchanged was dependent on the sorbate/sorbent ratio. In contrast, Cheng and Wang (1982) found that mechanical mixing was more effective than ultrasound in accelerating ion exchange equilibria. They suggested that the shockwaves produced during ultrasonic irradiation interrupted the film diffusion process and therefore lowered the ion exchange rate.

2.5.2 Effect of solids concentration on cavitation

Addition of particles into the liquid phase has been shown to enhance the yield of a sonochemical reaction (Sekiguchi and Saita, 2001; Keck et al., 2002). Particles provide additional nucleation sites for cavity formation due to the roughness of the particle surface thereby leading to a decrease in the cavitation threshold (Marschall et al., 2003; Borkent et al., 2007). Suspended particles also cause asymmetric collapse of bubbles present near the solid surface leading to the generation of high-speed microjets of liquid that causes dramatic physical effects on the solid surface. These microjets are responsible for pitting, erosion, and corrosion of the solid surface leading to the production of increased nucleation sites (Suslick and Price, 1999). On the other hand, the presence of particles also attenuates ultrasonic wave energy thus lowering the ultrasonic intensity propagating through the liquid. The net effect of these opposing influences will determine whether the presence of particles will promote or suppress cavitation formation.

Gogate et al. (2004) found that increasing solids concentration up to 500 ppm resulted in a decrease in phenol degradation via cavitation and suggested this was due to the dominant effect of wave scattering and attenuation. The addition of particles at concentrations less than 0.1 (v/v) was found to have a detrimental effect on cavitation due to the substantial prevention of sound propagation (Lu and Weavers, 2002; Lu et al., 2002).
Tuziuti et al. (2005) studied hydroxyl radical formation in the presence of alumina particles up to a solids concentration of 0.025 (v/v) and found cavitation increased initially up to a solids concentration of 0.005 (v/v) and then decreased thereafter. They proposed that the increase in cavitation with an increase in solids concentration was due to an increase in nucleation sites and the effect of soundwave attenuation at higher solids concentrations led to a net negative effect thereafter. Keck et al. (2002) also found similar results when using water irradiated with ultrasound at 206 kHz in the presence of quartz particles. They measured hydrogen peroxide formation rates, which increased initially then decreased with increasing solids concentration and particle size. The rate of hydrogen peroxide formation in the absence of quartz particles was found to be lower than those for all solids concentration studied. They explained their findings by suggesting that the asymmetric shaped bubbles formed due to the presence of particles had larger surface areas than spherical bubbles. The larger surface area enabled more free radicals to escape into the bulk solution forming more $\text{H}_2\text{O}_2$. Lu et al. (2002), however, suggested that radical production decreases when bubbles collapse asymmetrically.

**2.5.3 Effect of particle size on cavitation**

It has also been shown that the presence of suspended particles decreases the tensile strength of a liquid and that the decrease is greater with increasing particle size (Marschall et al., 2003). The decrease in liquid tensile strength results in more cavitation events due to lowering of the cavitation threshold. It has been stated that particles greater than 150 µm can act as a wall, which causes asymmetric bubble collapse resulting in microjet formation (Doktycz and Suslick, 1990). The asymmetric collapse leads to the generation of a large number of tiny bubbles thus resulting in enhanced sonochemical yields. Several researchers reported similar results and have shown that an increase in particle size leads to a decrease in ultrasonic wave attenuation thereby allowing more cavitation to occur (Zanwar and Pangarkar, 1988; Romdhane et al., 1997). However, other researchers suggest that increasing particle size has a detrimental effect on cavitation formation.

In studying nano/microparticles, it was found that cavitation preferentially occur on the surface of smaller particles due to a lower nucleation energy barrier (Zhang et al., 2014). They also confirmed that particle breakage was more apparent in larger particles, not only
due to interparticle collisions but also due to cavitation-induced microjets and shock waves. Tuziuti et al. (2005) found that particle size \(d_p = 1-80\ \mu m\) had no effect on cavity formation rates. They were not sure of the reason but proposed that small particles did not play a role in cavitation as they were in motion together with the liquid phase and did not act as a solid boundary to induce cavitation.

### 2.5.4 Particle surface influence on cavitation

It is largely accepted that surface roughness can have a dramatic effect on cavitation inception (Holl, 1960; Arndt and Ippen, 1968). This is due to the higher degree of lowering of the liquid pressure by rough particles compared to smooth particles (Holl, 1970). The importance of surface roughness in facilitating the entrapment and stabilization of gas pockets on the particle surface has been previously outlined (Borkent et al., 2007). Borkent et al. (2007) showed that the influence of particle type and surface roughness was more important in determining cavitation yield than surface area.

Various investigators have studied how particle parameters such as hydrophobicity, size, and concentration affect cavity formation. Greater hydrophobicity was found to enhance cavitation as bubbles formed on hydrophobic surfaces have higher contact angles between the solid and liquid phases. A higher contact angle causes a lowering of the energy barrier required for cavitation formation (Belova et al., 2011).

### 2.6 Ultrasonic power consumption

In addition to the impeller power consumption, power is also added into the system via ultrasound. A key issue for any process including sonochemical processes is the ability to measure energy input and operational efficiency. The universal procedure for acoustic power measurements is the calorimetric method (Kimura et al., 1996; Ji et al., 2006; Juang et al., 2006). Despite some drawbacks such as convective cooling, heating of the transducer and the presence of an intrusive sensor, it is a fairly precise method of quantifying cavitation effects (Martin and Law, 1980). The calorimetric method is based on the assumption that the
ultrasonic waves generated (mechanical energy) are degenerated into heat (Toma et al., 2011). From this assumption, the ultrasonic power \( U_p \) can be calculated from the rate of temperature increase by the following equation (Mason, 1992):

\[
U_p = C_p M_L \frac{dT}{dt}
\]  

(2.11)

where \( C_p \) is the heat capacity of the liquid (J kg\(^{-1}\) K\(^{-1}\)), \( M_L \) is the mass of the liquid (kg) and \( \frac{dT}{dt} \) is the temperature change per second (K t\(^{-1}\)).

It has been shown that the temperature rise of the system is independent of the initial bulk liquid temperature (below 40°C), the liquid height and the probe height (Kimura et al., 1996; Ratoarionoro et al., 1995). Hagensen and Doraiswamy (1998), however, found that the power estimated using Eq. (2.11) was inadequate. They suggested that Eq. (2.11) needed to be modified to incorporate the heat absorbed by the reaction vessel in addition to the heat absorbed by the solvent. In addition, experimental equipment such as baffles, impellers, cooling coils as well as particles will cause losses in energy conversion.

Other methods of estimating the power dissipated into the system are by using chemical dosimeters such as the Weissler reaction and measuring the generation of HNO\(_3\) from NO\(_3\) in water (Koda et al., 1996). Kimura et al. (1996) compared ultrasonic power values estimated using the calorimetric and Weissler reaction methods, and found both methods predicted similar values. Therefore, due to its simplicity and being one of the most common approaches used to determine the ultrasonic power, the calorimetric method is adopted in this study. In addition, the total power delivered into the system is the summation of power consumed by the impeller and the acoustic power.
2.7 Summary

From the above literature review, it can be concluded that information on how particle concentration, size, and surface topography affect cavity formation and its influence on sonochemical processes is limited. The role of ultrasound on solid-liquid mass transfer, particularly at high solids concentration is still not established. There are many studies in the literature that focuses on solids suspension and solid-liquid mass transfer in agitated vessels. However, information on dual-impeller vessels is limited, even with its extensive use in industry. Therefore, in this study, the investigation focuses on improving impeller power consumption and achieving increased throughput to evaluate whether process intensification is achieved more efficiently through mechanical agitation or cavitation.
Chapter 3

Experimental
Introduction

This chapter describes the equipment, materials and methodology used in this study. The specifications of the mixing vessels, impeller type, and ultrasonic device used in the study are described here. The techniques used to determine the critical impeller speed and cloud height, and the experimental procedure used to measure impeller power consumption, mass transfer rate and mass transfer coefficient are described. The experimental techniques used to determine particle size and particle influence on cavitation activity are also explained.

3.1 Materials

Specifications of the solid and liquid phases used in this work are shown in Tables 3.1 and 3.2, respectively.

3.1.1 Solid phase

Table 3.1 Solid phase specifications

<table>
<thead>
<tr>
<th></th>
<th>Dowex marathon C (H) ion exchange resin</th>
<th>Sand</th>
<th>Glass beads</th>
<th>Polymeric resin</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Shape</strong></td>
<td>Spherical</td>
<td>Spherical</td>
<td>Spherical</td>
<td>Spherical</td>
</tr>
<tr>
<td><strong>Matrix</strong></td>
<td>Polystyrene</td>
<td>SiO$_2$</td>
<td>Na$_2$SiO$_3$ / Na$_2$O / CaO</td>
<td>Polystyrene Divinylbenzene (macroporous)</td>
</tr>
<tr>
<td>Sauter-mean</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>diameter (d$_{32}$) (mm)</td>
<td>0.624</td>
<td>0.303</td>
<td>0.206</td>
<td>0.625</td>
</tr>
<tr>
<td><strong>Density (kg/m$^3$)</strong></td>
<td>1220</td>
<td>2500</td>
<td>1518</td>
<td>1160</td>
</tr>
</tbody>
</table>
3.1.2 Liquid phase

Table 3.2 Liquid phase specifications

<table>
<thead>
<tr>
<th></th>
<th>NaOH</th>
<th>KI</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Concentration (M)</td>
<td>0.025</td>
<td>0.1</td>
<td>-</td>
</tr>
<tr>
<td>Supplier</td>
<td>Chem-supply, Australia</td>
<td>Chem-supply, Australia</td>
<td>-</td>
</tr>
<tr>
<td>Physical state purchased</td>
<td>Dry solid pellets</td>
<td>Dry solid pellets</td>
<td>-</td>
</tr>
</tbody>
</table>

3.2 Measurement techniques

This section describes the measurement techniques used to determine the critical impeller speed for just off-bottom solids suspension ($N_{JS}$), cloud height, the minimum impeller speed to just disperse solids throughout the liquid ($N_{JCD}$), solid-liquid mass transfer rate, solid-liquid mass transfer coefficient ($k_{SL}$), impeller power consumption, ultrasonic energy dissipated, and the concentration of $I_3^-$ to quantify cavitation activity. The experimental technique to acquire micrographs using scanning electron microscope (SEM) is also discussed here. Experiments were carried out in triplicate and results were found to vary within ± 5% of the average value.
3.2.1 Determination of critical impeller speed for just off-bottom suspension $N_{JS}$

The critical impeller speed ($N_{JS}$) was determined in this study using a technique involving the measurement of the settled solid-bed height. This method was originally proposed by Hicks et al. (1997). This method is similar to Zwietering’s method except the bottom is illuminated and the tank bottom is observed from the side. This method has been demonstrated to be quite reliable for suspensions of solids at higher concentrations (Wu et al., 2010b; Wang et al., 2012a). The $N_{JS}$ in this work is defined at the point when the settled-solids bed disappears as the impeller speed is slowly increased from speeds lower than $N_{JS}$. This method of determining $N_{JS}$ was done by initially operating the impeller at high speeds to ensure all particles are suspended and no particle is stationary on the tank bottom. The impeller speed was then decreased gradually until a thin layer of particles appears on the tank bottom and then increased until the thin solid-bed disappears. The impeller speed at which the thin solid-bed disappears was designated as $N_{JS}$. Due to the types of flow produced by various impellers, particles did not evenly distribute throughout the tank bottom. Therefore, the bed height was measured at a point midway between two consecutive baffles. A light source was also used to illuminate the tank bottom to aid in the visualization of solids suspension, especially at higher solid loadings.

3.2.2 Determination of solids cloud height

The cloud height, $H_{S}$ was determined visually at $N_{JS}$ as well as at other impeller speeds. It was found that the slurry was not necessarily homogenous when the cloud height reached the liquid surface ($H_{S} = H$). The minimum impeller speed required to ‘just completely disperse’ the solids or $H_{S} = H$ was denoted as $N_{ICD}$. 
3.2.3 Measurement of impeller power consumption

The impeller power consumption was determined by measuring the torque experienced by the impeller. The torque was measured using a torque transducer (Burster 8645-500) assembly which is shown in Fig. 3.2. The motor and torque assembly was mounted on a metal frame to ensure the transducer is not affected by vibrations during agitation and make sure the transducer is correctly aligned. A pillow bearing and two flex couplers were used to connect the transducer to the shaft on both sides. This was done to prevent any extra bending experienced by the transducer during the experiment. The torque transducer was connected to a sensor interface (LCV-USB), which amplifies the measurements and converts analog signals into digital signals, which are then processed into torque readings using the Lorenz Messtechnik GmbH software on a PC.

The absolute torque $\tau$ experienced by the impeller shaft was determined using the following equation:

$$\tau = \tau_m - \tau_0$$

(3.1)

where $\tau_m$ is the measured torque during experimentation and $\tau_0$ is the torque measured at 0 rpm.
The impeller power consumption was determined using the following equation:

\[ P = 2\pi N \tau \]  

(3.2)

where \( P \) is the impeller power draw (W) and \( N \) is the impeller rotational speed in revolutions per second (rps).

The specific power consumption based on mass of suspended solids at ‘just suspended’ conditions (\( \epsilon_{JS} \)) was calculated using Eq. (3.3):

\[ \epsilon_{JS} = \frac{P_{JS}}{M_S} \]  

(3.3)

where \( \epsilon_{JS} \) (W/kg) denotes the specific impeller power consumption, \( P_{JS} \) (W) is the impeller power draw and \( M_S \) (kg) is the total solids suspended in the vessel.

Figure 3.2 Schematic diagram of equipment setup: \( H = T \), \( C = T/4 \), \( D = T/3 \), \( B = T/12 \)
3.2.4 Determination of ultrasonic power consumption

The ultrasonic power delivered into the system was measured using the calorimetric method (Eq. 3.4). This was achieved by adding 1 Litre of deionised water into a tank insulated with aluminium foil and measuring the temperature rise as a function of time under sonication. Taking into account the heat capacity and mass of water, the power dissipated into the system was then calculated. The problem of convective cooling, when the ultrasound processor is switched off, was mitigated by measuring and adding the cooling rate to the heating rate. The ultrasonic power \( (U_p) \) was calculated from the rate of temperature rise using the following equation:

\[
U_p = C_p M_L \frac{dT}{dt}
\]  

(3.4)

where \( C_p \) is the heat capacity of the liquid (J kg\(^{-1}\) K\(^{-1}\)), \( M_L \) is the mass of the liquid (kg) and \( \frac{dT}{dt} \) is the temperature rise per second (K t\(^{-1}\)).

3.2.5 Determination of particle stability

It has been shown in a previous study that particles exposed to ultrasound in a liquid medium can be eroded or broken up (Whillock and Harvey, 1997). For this reason, a particle size analysis was performed before and after irradiating the slurry. The particle size distribution and the average particle size were determined using particle size analyser (Malvern Mastersizer 3000). Solids samples were removed before sonication and after 10 minutes of combined agitation and sonication. Ultrasonic irradiation with power intensity of 131 W was applied to the slurry during the test. Sauter-mean particle diameter \( (d_{32}) \) values before and after ultrasonic irradiation are shown in Table 3.3 for the four types of particles used in this study.
Table 3.3 Impact of ultrasonic irradiation on particles of different sizes

<table>
<thead>
<tr>
<th>Irradiation time (min)</th>
<th>0</th>
<th>10</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>Spherical glass beads, $d_P$ (µm)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>206</td>
<td>204</td>
</tr>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>551</td>
<td>552</td>
</tr>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>1270</td>
<td>1360</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cation exchange resin, $d_P$ (µm)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>625</td>
<td>624</td>
</tr>
<tr>
<td>$C_V$ 0.05 (v/v)</td>
<td>624</td>
<td>624</td>
</tr>
<tr>
<td>$C_V$ 0.30 (v/v)</td>
<td>624</td>
<td>623</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sand, $d_P$ (µm)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>303</td>
<td>302</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Polymeric resin, $d_P$ (µm)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_V$ 0.01 (v/v)</td>
<td>625</td>
<td>625</td>
</tr>
</tbody>
</table>

3.2.6 Determination of $[I_3^-]$ using UV-Vis Spectrometry

Ultrasonic irradiation of an aqueous solution results in the formation of OH radicals due to the thermolysis of the water molecule according to reaction (3.1). Water in an aqueous solution can also decompose to form $H_2$ and O according to reaction (3.2). The high localised temperatures associated with cavitation can also cause reactions (3.3) and (3.4) to occur readily. To a large extent, the newly formed hydroxyl radicals ($\cdot$OH) react with each other according to reaction (3.5) forming hydrogen peroxide (Iida et al., 2005). The hydrogen peroxide formed in ultrasonically irradiated aqueous KI solution reacts with free $\Gamma$ anions to form iodine ($I_2$), which then reacts with excess $\Gamma$ to form $I_3^-$ according to reactions (3.6) and (3.7), respectively (Hart and Henglein, 1985). The concentration of $I_3^-$ formed is an indication...
of cavitation activity and was measured using a UV-Vis spectrometer (Thermo Scientific Helios Zeta) at 355 nm.

\[ H_2O \rightarrow H \cdot + \cdot OH \]  \hspace{1cm} (3.1)

\[ H_2O \rightarrow H_2 + O \]  \hspace{1cm} (3.2)

\[ H \cdot + H_2O \rightarrow H_2 + \cdot OH \]  \hspace{1cm} (3.3)

\[ O + H_2O \rightarrow \cdot OH + OH \]  \hspace{1cm} (3.4)

\[ \cdot OH + \cdot OH \rightarrow H_2O_2 \]  \hspace{1cm} (3.5)

\[ H_2O_2 + 2I^- + 2H^+ \rightarrow I_2 + 2H_2O \]  \hspace{1cm} (3.6)

\[ I_2 + I^- \rightarrow I_3^- \]  \hspace{1cm} (3.7)

The experimental procedure used to determine the sonochemical oxidation of KI in the presence of particles is as follows: A known amount of 0.1M KI solution was added to the baffled tank and the impeller was operated at the pre-determined critical speed (\(N_{JS}\)) required to ‘just suspend’ all particles off the tank bottom (Zwietering, 1958). A chosen amount of particles was then added to the tank and the slurry was sonicated (131 W) for 10 minutes. Ten mL of aliquots was removed every 2 minutes for the absorbance measurements. Silent experiments were also conducted under agitation-only conditions by switching off the ultrasonic generator but with the probes still submerged in the tank.

3.2.7 Obtaining micrographs using a Scanning Electron Microscope (SEM)

A scanning electron microscope (SEM) analysis was performed to study the morphology of the particles using a Phillips XL30 SEM and a FEI Quanta 200 SEM, fitted with a tungsten filament. The working distance was set at 10 mm. Samples were gold sputter coated with a coating thickness of \(~25\) nm to alleviate sample charging. A low electron beam accelerating voltage of either 5 kV or 30 kV was used to ensure the surface features were imaged accurately.
The mass transfer coefficient was determined by measuring the changes in NaOH concentration due to the interphase exchange of cations. The change in sodium concentration was determined by measuring the solution conductivity using an electrical conductivity meter (HACH, sension™ HQ40d). The conductivity probe was placed adjacent the shaft at mid-liquid height position. A calibration curve, which is a plot of solution specific conductance versus solute concentrations, was prepared using standard NaOH solutions. This plot was used to determine the concentration of NaOH from its specific conductance.

It is assumed that the rate-limiting step in interphase mass transfer is diffusion across the liquid film surrounding the solid particle. Also, since the conductivity probe was calibrated to measure Na\(^{+}\) ion concentration, a differential mass balance could be written to express the rate of Na\(^{+}\) disappearance from the bulk liquid to the solid phase as follows:

\[
\frac{\partial [C_{Na}]}{\partial t} = k_{SL}a_p([C_{Na}] - [C_{Na}]_0)
\]  
(3.5)

The volumetric solid-liquid mass transfer coefficient, \(k_{SL}a_p\) (s\(^{-1}\)) is the product of the solid-liquid mass transfer coefficient, \(k_{SL}\) (m/s) and the solid-liquid interfacial area per unit volume of solid, \(a_p\) (m\(^{-1}\)). On integration and applying limits, equation (3.5) can be written in the form of equation (3.6), which can be used to determine \(k_{SL}a_p\) by measuring the changes in solute concentration in the solvent (Levins and Glastonbury, 1972; Tezura et al., 2008):

\[
\ln\left(\frac{[C_{Na}]}{[C_{Na}]_0}\right) = -k_{SL}a_p t
\]  
(3.6)

where \([C_{Na}]\) and \([C_{Na}]_0\) (mol/L) are the sodium molar concentrations at time \(t\) and \(t = 0\), respectively. The solid-liquid interfacial area, \(a_p\) was calculated using the following equation:

\[
a_p = \frac{6C_v}{d_{32}}
\]  
(3.7)

where \(C_v\) (v/v) is the volumetric solids concentration and \(d_{32}\) (m) is the Sauter-mean particle diameter. Using Eq. (3.6), \(k_{SL}a_p\) was obtained as the slope from the linear plot of ln([C\(_{Na}\)]/[C\(_{Na}\)\(_0\)]) versus \(t\). The solid-liquid mass transfer coefficient \(k_{SL}\) was then determined by dividing \(k_{SL}a_p\) by the interfacial surface area \(a_p\).
3.2.9 Measurement of solid-liquid mass transfer rate in phenol-polymeric resin system (desorption system)

Polymeric resin saturated with phenol and deionised water were used as the solid and liquid phases, respectively in all desorption experiments. The required amount of polymeric resin saturated with phenol was added to water while operating the stirrer at $N_S$ under ultrasound irradiated condition. Ten mL aliquots were removed at time intervals of 0.5, 1, 2, 3, 4, 5, 6, 8 and 10 minutes for the purpose of absorbance measurements. The amount of phenol desorbed from the polymeric resin into water was determined by measuring the concentration of phenol in water using a UV-Vis spectrometer (Thermo Scientific Helios Zeta) at 270 nm. In this study, the volumetric solids concentration ($C_V$) was varied from 0.03 to 0.20 (v/v) to study its effect on solid-liquid mass transfer under sonication.
3.3 Study 1: Impeller power consumption and solid-liquid mass transfer in an agitated vessel fitted with dual-impellers

3.3.1 Experimental set up

The impellers were mounted on a centrally driven shaft attached to a torque transducer (Burster 8645) which had a measurement range of 0 ± 5 Nm. The impeller shaft was driven by an electric motor (Heidolph RZR 2102 control) and the impeller speed was varied by adjusting the rotational speed on the motor control panel. The torque experienced by the impeller shaft was recorded as a function of time on a personal computer.

A schematic diagram of the experimental set-up is shown in Fig. (3.3). Experiments were carried out in a cylindrical, flat-bottomed Perspex tank with a diameter (T) of 0.2 m inside an outer square Perspex tank. The slurry height H was set to 1.5 T and H=T for vessels agitated with dual- and single-impellers, respectively. The baffled tank was fitted with four equally spaced baffles with width (B) equal to T/12. The outer square tank was filled with water to minimise optical distortion during flow visualisation.

For the dual-impeller experiments, two impeller combinations namely, two 6-bladed Rushton turbines (RTRT) and an A310 hydrofoil and a 6-45°pitch bladed turbine (A310PBT), were used. The single-impeller experiments were carried out with Rushton turbine (RT) and A310 hydrofoil impeller. The impeller diameter (D) was equal to T/3 except for the hydrofoil turbine which was 0.064 m in diameter. The off-bottom clearance (C) was set equal to T/4 for all experiments. The impeller spacing (S) in the dual-impeller experiments was equal to the impeller diameter. The A310 was set as the bottom impeller for A310PBT configuration. The impellers used in this work are shown in Fig. (3.4).
3.3.2 Materials used

Ion exchange resin and analytical grade NaOH solution were used as the solid and liquid phases, respectively in this study. The volumetric solids concentration ($C_V$) in this work was varied from 0.05 to 0.35 (v/v) while keeping the slurry volume constant. Experiments were carried using liquid (NaOH) concentration of 0.025 M. This aqueous solution concentration was chosen in all experiments because the decrease in NaOH concentration did not occur rapidly thereby enabling sufficient number of data points to be collected for both higher and lower $C_V$. 
3.4 Study 2: Impact of particle size, roughness and concentration on sonochemical reactions in potassium iodide solution

3.4.1 Experimental set up

Experiments were conducted in a 0.2 m diameter (T) cylindrical, flat-bottomed perspex tank fitted with four equally spaced baffles with width (B) equal to T/12 and placed inside a square outer perspex tank. The space between the inner and outer tanks was filled with running water to maintain the temperature of tank liquid at 23 ± 2°C. The slurry height (H) in all experiments was set equal to T. A centrally driven shaft mounted with A310 hydrofoil turbine of diameter (D) = 0.0635 m was used as the agitator. The off-bottom impeller clearance was set equal to H/4. Ultrasound was applied using a dual ultrasonic probe (20 kHz Sonics and Materials VCX 1500). The probe tips were 0.019 m in diameter, 0.114 m apart and were submerged 0.1 m from the liquid surface adjacent to the impeller shaft.

3.4.2 Materials used

Aqueous potassium iodide (KI) solution was used as the liquid phase and cationic exchange resin, sand, and glass particles were used as the solid phases. The volumetric solids concentration (C_V) in this work was varied from 0.05 to 0.35 (v/v) while keeping the slurry volume constant. Experiments were carried out using KI concentration of 0.1 M.
3.5 Study 3: Solid-liquid mass transfer with ultrasound in cation-NaOH solution system and polymeric resin-phenol-water system

3.5.1 Experimental set up

All experiments were conducted in a 0.2 m diameter (T) cylindrical, flat-bottomed perspex tank placed inside a square outer tank. The space between the inner and outer tank was filled with running water to maintain the tank liquid temperature at 23 ± 2°C. Four equally spaced baffles were installed in the inner tank with width (B) equal to T/12. The slurry height (H) was set to equal T. A six bladed Rushton turbine with impeller diameter of D = T/3 mounted on a centrally driven shaft was used as the agitator. The off bottom impeller clearance was set to equal T/4. Ultrasound experiments were conducted using a dual ultrasonic probe (20 kHz Sonics and Materials VCX 1500). The ultrasonic probe tips (0.019 m in diameter) were submerged 0.1 m adjacent the impeller shaft and they were 0.114 m apart. Silent experiments were conducted with the probes still submerged in the vessel with the ultrasound generator switched off.

3.5.2 Materials used

Ion exchange resin and analytical grade NaOH solution were used as the solid and liquid phases, respectively in the ion exchange system. The initial liquid concentration was 0.025 M and the resin had an average diameter (d_P) of 0.6 - 0.7 mm and a density (ρ_S) of 1220 kg/m³. The resin was washed with deionised water and dried using suction filtration to remove any contaminants prior to use.

Polymeric resin used in the phenol desorption experiment was Dowex Optipore L-493 resin, which is a methylene bridged copolymer of styrene and divinylbenzene. The particle diameter was between 0.6 - 0.7 mm and the particle density was 1160 kg/m³. The polymeric resin was pretreated with a deionised water wash before being saturated with phenol. This was done by adding 6 kg of resin to 40L of 0.05 M phenol and agitating the tank for 48 hours. The resin was then dried with suction filtration and used in the desorption experiments. The volumetric solids concentration (C_V) in this work was varied from 0.05 to 0.35 (v/v) while keeping the slurry volume constant.
Chapter 4

Results and Discussion

Predicting $N_{JS}$ using the Zwietering correlation for single- and dual-impeller systems
Introduction

The chapter presents $N_{JS}$ values obtained in solid-liquid agitated vessels with single- and dual-impellers under baffled and unbaffled conditions. $N_{JS}$ was determined using a technique involving the measurement of the settled solid-bed height as shown in Chapter 3, for a solids concentration up to 0.35 (v/v). The applicability of the Zwietering correlation in solid-liquid systems operating at high solids concentration in agitated vessels with single- and dual-impellers under baffled and unbaffled conditions is also discussed. In addition, this chapter discusses whether the Zwietering correlation can reliably predict $N_{JS}$ in solid-liquid systems with different impeller and vessel geometries than those used in the original work by Zwietering.

The results presented in Chapter 4 have been submitted as part of a manuscript for peer review, and has been accepted for publication.


4.1 Results and Discussion

4.1.1 Effect of solids concentration on $N_{JS}$

The critical impeller speed ($N_{JS}$) values obtained in this work for RT in the single-impeller system are shown in Fig. 4.1, whereas those of RTRT in the dual-impeller system are shown in Fig. 4.2 as a function of $C_V$ under baffled conditions. As expected, the $N_{JS}$ value increases with an increase in $C_V$ up to 0.35 (v/v). This trend indicates that increased impeller speeds are required to just suspend solids off the tank bottom when $C_V$ increases, which is consistent with the findings of many researchers including that of Ayranci et al. (2013). From Figs. 4.1 and 4.2, it can be seen that $N_{JS}$ values for RT under baffled conditions are greater than those for RTRT at a given $C_V$ value. In the case of dual impellers, the bottom impeller is largely responsible for solids suspension and the upper impeller is largely associated with particle distribution (Hicks et al., 1997). The addition of a second impeller enhances the overall mixing in the vessel by distributing solids throughout the tank thereby decreasing the amount
of solids that drop out of suspension. In addition, dual-impeller systems with impeller configurations that generate liquid flow that complements that of the bottom impeller results in lower $N_{JS}$ values (Mak, 1992).

Figure 4.1 $N_{JS}$ values for RT under baffled conditions as a function of $C_V$

Figure 4.2 $N_{JS}$ values for RTRT under baffled conditions as a function of $C_V$
4.1.2 Effect of impeller type on $N_{JS}$

To investigate the effect of impeller type, $N_{JS}$ values for RT and A310 in the single-impeller system are shown in Figs. 4.3a and 4.3b, whereas those of RTRT and A310PBT in the dual-impeller system are shown in Figs. 4.3c and 4.3d as a function of $C_V$ under both baffled and unbaffled conditions, respectively. The flow generated by RT and A310 impellers represents radial and axial flows, respectively. From Figs. 4.3a to 4.3d, it can be seen that $N_{JS}$ values for RT and RTRT (radial pumping impellers) are lower than those for axial and axial/mixed pumping impellers for single- and dual-impeller configurations. High shear impellers such as RT generate radial liquid flow with higher levels of turbulence compared to the turbulence generated by low shear impellers such as A310 at the same impeller speed (Zhou and Kresta, 1996). Consequently, A310 impeller requires higher speeds to generate the turbulence required for solids suspension at the tank bottom. Similar to the results mentioned above, Wang et al. (2012b) reported that A310 has higher $N_{JS}$ values than RT under both baffled and unbaffled conditions for a given $C_V$. Ayranci and Kresta (2014) also reported that $N_{JS}$ values for A310 were greater than those for PBT under baffled conditions.

Figure 4.3 Effect of impeller type on $N_{JS}$ as a function of $C_V$: (a) baffled vessel, single-impeller, (b) unbaffled vessel, single-impeller, (c) baffled vessel, dual-impeller, (d) unbaffled vessel, dual-impeller
4.1.3 Effect of baffles on $N_{JS}$

Baffles are used in agitated vessels to enhance mixing, eliminate vortexing, and improve mass transfer. The effect of baffles on $N_{JS}$ can be seen from Figs. 4.4a and 4.4b for RT and A310 in single-impeller systems and from Figs. 4.4c and 4.4d for RTRT and A310PBT in dual-impeller systems. It can be seen that $N_{JS}$ values for RT under baffled conditions for all $C_V$ are greater than those under unbaffled conditions. For the RT impeller, a 30-35% decrease in $N_{JS}$ is observed due to the removal of baffles for a $C_V$ up to 0.35 (v/v) (Fig. 4.4a). Davoody et al. (2016) also reported recently that $N_{JS}$ values for RT under baffled conditions are greater than those under unbaffled conditions up to a $C_V$ of 0.4 (v/v). On the other hand, no significant difference in $N_{JS}$ values between those for baffled and unbaffled tanks was found for systems using A310, RTRT and A310PBT impellers (Figs. 4.4b to 4.4d). Wang et al. (2012b) reported similar results for RT. They also reported that there is no significant difference in $N_{JS}$ for PBT and A310 impellers under baffled and unbaffled conditions.

Figure 4.4 Effect of baffle removal on $N_{JS}$ as a function of $C_V$: (a) RT, (b) A310, (c) RTRT, (d) A310PBT
4.2 Estimation of $N_{JS}$ using the Zwietering correlation

The $N_{JS}$ values predicted using the Zwietering correlation (Eq. 2.2, shown in Figs. 4.5a to 4.5d as continuous lines) are compared to experimental results in Figs. 4.5a to 4.5d. The $S$ values used in the estimations were obtained using back-solving method and are shown in Table 4.1. The $S$ value is a function of impeller type, impeller diameter, and off-bottom clearance, and it is sensitive to impeller geometry. Zwietering reported $S$ values for several combinations of impeller diameter and type, and some off-bottom clearances. Many other studies reported $S$ values for a wide range of geometries (Ibrahim and Nienow, 1996; Atiemo-Obeng et al., 2004; Ayranci and Kresta, 2011). The possible geometric combinations are infinite and the $S$ values reported in this study are additions to the literature. The $N_{JS}$ values predicted agree well with the experimental values for single-impeller systems under both baffled and unbaffled conditions. However, for dual-impeller systems, the $N_{JS}$ values predicted are slightly higher than experimental values at low $C_V$ ($< 0.2 \ (v/v)$) and lower at higher $C_V$ (Figs. 4.5c and 4.5d). This is especially the case for A310PBT under unbaffled condition (Fig. 4.5d). Ayranci and Kresta (2014) suggested that the exponent for $X$ in the Zwietering correlation is greater for higher solids concentration and is dependent on particle type. However, results from this work suggest that the value of 0.13 for the exponent $X$ accurately represents the concentration dependence in the correlation proposed by Zwietering. Zwietering (1958) used solids concentration only up to $C_V \approx 0.12 \ (v/v)$ but the results of this study suggest that the Zwietering correlation is applicable up to 0.35 $(v/v)$ under both baffled and unbaffled conditions for both single- and dual-impeller systems.

The $N_{JS}$ values determined using the Zwietering correlation and $S$ values shown in Table 4.1 are compared to experimental $N_{JS}$ values in a parity plot shown in Fig. 4.6. It can be seen that all data points are situated within a ±15% band indicating that the Zwietering correlation can be reliably extended to solid-liquid systems agitated with dual-impellers under both baffled and unbaffled conditions.
Figure 4.5 Experimental and predicted $N_{JS}$ values using the Zwietering correlation: (a) baffled vessel, single-impeller system, (b) unbaffled vessel, single-impeller system, (c) baffled vessel, dual-impeller system, (d) unbaffled vessel, dual-impeller system

Figure 4.6 Comparison of experimental data in solid-liquid systems with values predicted using $S$ values found in Table 4.1
Table 4.1 Values of experimental Zwietering constants, S

<table>
<thead>
<tr>
<th>Zwietering constant, S</th>
<th>RT</th>
<th>A310</th>
<th>RTRT</th>
<th>A310PBT</th>
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<tbody>
<tr>
<td></td>
<td>Baffled</td>
<td>Unbaffled</td>
<td>Baffled</td>
<td>Unbaffled</td>
</tr>
<tr>
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<td>5.5</td>
<td>10.5</td>
<td>10</td>
<td>5.8</td>
</tr>
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</table>
Summary

The critical impeller speed to ‘just suspend’ particles off the tank bottom (N_{JS}) were determined experimentally using a system involving cation exchange resin and water for a range of solids concentrations (0.05-0.35 (v/v)). The impellers used in this study for a single-impeller system were the radial flow Rushton turbine (RT) and the axial flow hydrofoil turbine (A310). The impeller combinations used in the dual-impeller systems were the dual radial flow RTRT and the axial/mixed flow A310PBT. These impellers were operated under both baffled and unbaffled conditions to determine the impeller type effect on N_{JS}. Results show that, with an increase in C_v, N_{JS} increases regardless of baffle arrangement or impeller type. Among the impeller configurations used, RT and RTRT achieved ‘just suspended’ conditions at lower impeller speeds than A310 and A310PBT in both single- and dual-impeller systems, respectively. This was the case for both baffled and unbaffled conditions. The removal of baffles had led to a decrease of 30-35% in N_{JS} for RT over a C_v range of 0.05-0.35 (v/v). On the other hand, N_{JS} values remained more or less the same for A310, RTRT and A310PBT regardless of baffle arrangement.

The reliability of the Zwietering correlation in predicting N_{JS} in solid-liquid systems under conditions that were different from those used in the original work was investigated. The results show that the correlation can be extended up to a solids concentration of 0.35 (v/v) for baffled and unbaffled vessels agitated with single- and dual-impeller configurations. Zwietering constant, S was determined from the experimental data for the tank and impeller geometries used in this study. N_{JS} values estimated using the Zwietering correlation and S values determined in this study were compared to those found experimentally in a parity plot, with all data points situated within ± 15% band. It is suggested that Zwietering correlation can be reliably extended to estimate N_{JS} in solid-liquid systems similar to those used in this work.
Chapter 5

Results and Discussion

Impeller power consumption for single- and dual-impeller systems
Introduction

In Chapter 4, the critical impeller speed to ‘just suspend’ particles off the tank bottom (N\textsubscript{JS}) was shown to increase with increasing C\textsubscript{V} and was dependent on impeller type and baffling arrangement. Following on from Chapter 4, this chapter discusses the effect of solids concentration C\textsubscript{V}, impeller type, and baffles on specific impeller power input per solids mass \(\varepsilon\textsubscript{JS}\) in both single- and dual-impeller systems. The results presented in this chapter will help evaluating the optimum solids concentration and impeller configuration to attain ‘just suspended’ or ‘just complete dispersion’ conditions.

The results presented in Chapter 5 have been submitted as part of a manuscript for peer review, and has been accepted for publication.


5.1 Results and Discussion

5.1.1 Effect of solids concentration on specific impeller power consumption

Impeller power consumption at N\textsubscript{JS} values based on slurry volume (P\textsubscript{JS}/V) for RT and A310 are shown in Figs. 5.1a and 5.1b as a function of C\textsubscript{V} under baffled and unbaffled conditions, respectively. For both baffled and unbaffled vessels, (P\textsubscript{JS}/V) values increase with increasing C\textsubscript{V}. While the (P\textsubscript{JS}/V) values for RT are higher than those for A310 under baffled conditions at all C\textsubscript{V}, they are nearly equal to those of A310 under unbaffled conditions. The increase in (P\textsubscript{JS}/V) with increasing C\textsubscript{V} can be ascribed to increased slurry viscosity. The apparent viscosity \(\eta\textsubscript{slurry}\) of the solid-liquid mixture was calculated at all solids concentration used in this work using equation (5.1) assuming uniform particle concentration (Fedors, 1974).

\[
\eta\textsubscript{slurry} = \eta_r \left(1 + \frac{1.25C_V}{\varphi_m - C_V} \right)^2
\]

(5.1)

where \(\eta_r\) is the viscosity of the carrier fluid (Pa.s), \(\varphi_m\) is the maximum volume fraction to which the particle can pack. Fedors (1974) used \(\varphi_m = 0.63\) for a system containing permanent
aggregates in a Newtonian liquid. However, in later studies, $\phi_m$ was reported to be equal to 0.68 which gave good agreement between experimental data and model predictions (Honek et al., 2005). Therefore, $\phi_m = 0.68$ was used in this work.

Fig. 5.2 shows that effective slurry viscosity values predicted using Fedors’ (1974) correlation, as reported by Honek et al. (2005), for $C_V$ up to 0.35 (v/v). Fedors’ (1974) correlation was modified from Eilers’ (1941) model, which was suggested for systems consisting of bitumen and soap solution. Fedors proposed his correlation for a solid-liquid system consisting of permanent aggregates in Newtonian liquids. The increase in $(P_{JS}/V)$ with increasing effective slurry viscosity is consistent with the results presented by other authors (Ibrahim and Nienow, 1999; Wang et al., 2010). The increase in $(P_{JS}/V)$ with increase in $C_V$ can also be attributed to increased particle-particle, particle-liquid, and particle-equipment collisions as was done by Bubbico et al. (1998).

The impeller power input values at $N_{JS}$ based on slurry volume $(P_{JS}/V)$ in a dual-impeller system are shown in Figs. 5.3a and 5.3b as a function of $C_V$ under baffled and unbaffled conditions, respectively. The impeller configurations used were RTRT and A310PBT. It can be seen that $(P_{JS}/V)$ increases with increasing $C_V$ in both baffled and unbaffled vessels similar to that observed in the single-impeller system shown in Figs. 5.1a and 5.1b. It is also clear that the removal of baffles leads to significant decrease in specific power input in dual-impeller systems. However, in contrast to the results of single-impeller systems, the $(P_{JS}/V)$ values for RTRT (radial pumping impellers) are lower than those for A310PBT (axial/mixed pumping impellers) under both baffled and unbaffled conditions.
Figure 5.1a Impeller power input at $N_{JS}$ per unit slurry volume ($P_{JS}/V$) as a function of $C_V$: baffled vessel

Figure 5.1b Impeller power input at $N_{JS}$ per unit slurry volume ($P_{JS}/V$) as a function of $C_V$: unbaffled vessel
Figure 5.2 Effect of $C_V$ on apparent slurry viscosity (correlation proposed by Fedors (1974))

Figure 5.3a Impeller power input at $N_{JS}$ per unit slurry volume ($P_{JS}/V$) as a function of $C_V$: baffled vessel
Another approach by which the specific impeller power consumption at $N_{JS}$ can be expressed on the basis of total slurry mass ($P_{JS}/(M_S+M_L)$), where $M_S$ and $M_L$ are total mass solids and liquid, respectively. The experimental data for ($P_{JS}/(M_S+M_L)$) at $N_{JS}$ for RT and A310 are shown in Figs. 5.4a and 5.4b as a function of $C_V$ under baffled and unbaffled conditions, respectively in single-impeller systems. The trends in ($P_{JS}/(M_S+M_L)$) curves are similar to those for ($P_{JS}/V$) curves as shown in Figs. 5.1a and 5.1b. The main reason for this is the nearly constant $(M_S+M_L)$ values (5.26 to 5.58 kg) for a $C_V$ range of 0.05 – 0.35 (v/v) due to nearly similar densities of the cation particles and liquid phases ($\rho_S = 1220 \text{ kg/m}^3$ and $\rho_L = 1000 \text{ kg/m}^3$) used in our experiments. Values of impeller power input at $N_{JS}$ on the basis of total mass ($P_{JS}/(M_S+M_L)$) in dual-impeller systems are shown in Figs. 5.5a and 5.5b for baffled and unbaffled conditions, respectively. When compared to results shown in Figs. 5.3a and 5.3b, the trends of power input curves are the same.
Figure 5.4a Impeller power input per unit total slurry mass \( \frac{P_{JS}}{M_S+M_L} \) as a function of \( CV \): baffled vessel

Figure 5.4b Impeller power input per unit total slurry mass \( \frac{P_{JS}}{M_S+M_L} \) as a function of \( CV \): unbaffled vessel
Figure 5.5a Impeller power input per unit total slurry mass ($P_{JS}/(M_S+M_L)$) as a function of $C_V$: baffled vessel

Figure 5.5b Impeller power input per unit total slurry mass ($P_{JS}/(M_S+M_L)$) as a function of $C_V$: unbaffled vessel

Figs. 5.6a and 5.6b show experimental $\varepsilon_{JS} = (P_{JS}/M_S)$ values as a function of $C_V$ for RT and A310 under baffled and unbaffled conditions, respectively in single-impeller systems. For both impellers, the $\varepsilon_{JS}$ value decreases with increasing $C_V$, reaches a minimum, and thereafter either increases slightly (A310 under baffled conditions) or remains constant. This trend is consistent with the results reported by Wang et al. (2012a), who studied the effect of high
solids concentration on $\varepsilon_{JS}$ using glass particles. Similar results were also reported by Bong et al. (2015) who found that the minimum $\varepsilon_{JS}$ value is around 0.20-0.25 (v/v) for ion exchange particles. They defined the $C_V$ at which the $\varepsilon_{JS}$ value was minimum as the “optimum solids concentration ($C_{V(\text{op})}$”). The $C_{V(\text{op})}$ values obtained for single-impeller systems under both baffled and unbaffled conditions are at higher $C_V$ ranges due to $\varepsilon_{JS}$ values remaining constant with increasing $C_V$. The exception to this is A310 under baffled conditions where $C_{V(\text{op})}$ is around 0.2-0.3 (v/v) (Fig. 5.6a).

The $\varepsilon_{JS}$ values for RTRT and A310PBT impeller systems are shown in Figs. 5.7a and 5.7b for baffled and unbaffled conditions, respectively in dual-impeller systems. Similar to that in single-impeller systems, $\varepsilon_{JS}$ values decreases with increasing $C_V$, reaches a minimum at a $C_V$ value of 0.2 (v/v), and increases thereafter. The $C_{V(\text{op})}$ value for dual-impeller systems is at $C_V = 0.2$ (v/v). At $C_{V(\text{op})}$, more solids are suspended per unit impeller power (kg/W) than at any other solids concentration. At lower $C_V$, the energy supplied by the impeller is not fully utilised for suspension because a majority of it is used unnecessarily to move the liquid around. These findings indicate that operating the solid-liquid mixing vessel with dual impellers at a $C_V$ of 0.20 (v/v) would result in more efficient specific impeller power usage. Similar findings were observed by Wang et al. (2014) for a dual-A310 configuration under baffled conditions.

**Figure 5.6a $\varepsilon_{JS}$ as a function of solids concentration: baffled vessel**
Figure 5.6b $\varepsilon_{JS}$ as a function of solids concentration: unbaffled vessel

Figure 5.7a $\varepsilon_{JS}$ as a function of solids concentration: baffled vessel
Effect of impeller type on specific impeller power consumption

The key to selecting the most suitable type of impeller for a solid-liquid mixing system is to determine which impeller set-up draws the least impeller power (Kasat and Pandit, 2005). Of the two impellers used in single-impeller systems, RT has the higher $\varepsilon_{JS}$ values at all $C_V$ than A310 under baffled conditions (Fig. 5.6a). The higher power draw required by RT could be explained by the radial liquid flow produced by the impeller. The liquid flow generated by RT flows in a radial direction towards the vessel wall before it is separated into two circulating loops, one above and one below the impeller. As a result, only a fraction of energy supplied by the impeller is used for solids suspension (Raghava Rao et al., 1988; Ayranci and Kresta, 2011). The purely axial flow A310 impeller leads to lower $\varepsilon_{JS}$ values than RT for a given $C_V$ under baffled conditions. Similar results were reported by Wu et al. (2011). The $\varepsilon_{JS}$ results for unbaffled conditions in single-impeller systems exhibit a trend opposite to those for baffled conditions (Fig. 5.6b). For all $C_V$, $\varepsilon_{JS}$ values for RT are lower than those of A310, indicating that RT, which has a higher impeller power number, is more efficient than A310, which is the lower power number impeller. Similar results were reported by Wang et al. (2012a) for both baffled and unbaffled conditions for spherical glass particles.
It can also be seen from Figs. 5.7a and 5.7b that RTRT impellers consume less power at $N_{JS}$ compared to A310PBT impellers under both baffled and unbaffled conditions, respectively. The difference in $\varepsilon_{JS}$ values between these two impeller combinations is around 40-50% and 60-70% for baffled and unbaffled vessels, respectively. Similar findings were made by Wu et al. (2011) who reported that $\varepsilon_{JS}$ values for low power number dual-axial pumping impellers are higher than those for high power number dual-axial pumping impellers at a solids concentration of 0.4 (v/v) under both baffled and unbaffled conditions. The above results can be attributed to the homogenous distribution of particles observed in our experiments for the RTRT impellers. In general, the bottom impeller in a dual-impeller system is largely responsible for solids suspension at the tank bottom while the top impeller is responsible for particle distribution in the upper volume of the vessel. In the case of RTRT impellers, the top RT impeller ensured even dispersion of particles thereby complimenting the flow generated by the bottom RT impeller. The combined flow fields generated by upper and lower RT impellers led to fewer solids dropping out of suspension thereby leading to a lower $N_{JS}$ value and consequently lower impeller power consumption (Paul et al., 2004).

5.1.3 Effect of baffles on specific impeller power consumption

It has been reported that the removal of baffles leads to a significant reduction in impeller power consumption for particle suspension even at high solids concentration (Markopoulos et al., 2005; Wu et al., 2010b). Figs. 5.6a and 5.6b show that $\varepsilon_{JS}$ values for both RT and A310 are lower under unbaffled conditions compared to those under baffled conditions. The reduction in $\varepsilon_{JS}$ under unbaffled conditions is about 80% for RT and 60% for A310. The flow phenomena that could be attributed to the reduction is an inward-spiralling liquid flow that is dominant under unbaffled conditions which moves the particles away from the tank walls towards the centre and suspending them below the impeller. Assirelli et al. (2008) also suggested that the unchaotic liquid swirling motion formed in unbaffled tanks leads to a reduction in the impeller power consumption compared to baffled tanks. Therefore, operating the vessel without baffles could enhance the overall impeller energy efficiency regardless of impeller type.

The impeller power consumed in vessels with multiple impellers largely depends on the impeller type and geometry, number of impellers, and impeller spacing. Figs. 5.7a and 5.7b
show that unbaffled tanks require significantly less power for solids suspension for both RTRT and A310PBT impeller configurations. The reduction in $\varepsilon_{JS}$ due to the removal of baffles is in the order of 70-75% for RTRT and 50-60% for A310PBT impellers. Similar reductions in $\varepsilon_{JS}$ due to the removal of baffles were reported by Wu et al. (2011) for dual-radial and dual-axial pumping impellers at $C_v = 0.4$ (v/v). They noted that the reduction in $\varepsilon_{JS}$ for dual-radial pumping impellers is greater than that for dual-axial pumping impellers.

5.2 Particle dispersion at $N_{JS}$

At $N_{JS}$, nearly all particles are suspended off the tank bottom and a major fraction of them are lifted to higher levels in the liquid. The maximum height to which the particles are lifted is called the ‘cloud height’. Experimental cloud height values in single-impeller systems normalised against liquid height ($H_S/H$) at $N_{JS}$ for RT and A310 are shown in Figs. 5.8a and 5.8b as a function of $C_v$ under baffled and unbaffled conditions, respectively. Under baffled conditions, the cloud height is equal to liquid level for RT ($H_S = H$) for all $C_v$ (Fig. 5.8a). For A310, under baffled conditions, the cloud height varies from 0.8H at $C_v = 0.05$ to nearly H at $C_v = 0.35$ (Fig. 5.8a). For both RT and A310 impellers, under unbaffled conditions, the cloud height is lower than the liquid level at $C_v = 0.05$ and it increases with increasing $C_v$ and becomes equal to the liquid height at $C_v = 0.25$ (Fig. 5.8b). These results indicate that RT achieves higher cloud height than A310 at $N_{JS}$ regardless of the baffle arrangement. Similar results were reported by Wang et al. (2012b) using the same impellers for smaller particles ($d_p = 0.09$ mm) at $C_v = 0.4$ (v/v). Wang et al. (2014) found that the cloud height achieved at $N_{JS}$ varied from the highest to lowest for RT, A310 and PBT. As the power number $N_P$ of RT>PBT>A310, it is clear that particle dispersion is influenced by the liquid flows produced by the impeller, and not due to the changes in impeller power number. These results indicate that the cloud height is the result of interaction between the flow patterns generated by the impeller, which depends on the baffle arrangement, and the particle settling velocity, which has a distribution depending on the particle size distribution.

Fig. 5.9 compares the impeller speed required to ‘just-suspend’ the particles off the tank bottom ($N_{JS}$) and that required to ‘just completely disperse’ solids ($N_{JCD}$) for single-impeller systems. It is interesting to note that $N_{JCD}$ values for RT under baffled conditions are lower than $N_{JS}$ for all $C_v$ used in this work indicating that the impeller speed to ‘just completely
disperse’ solids may not always be greater than \( N_{JS} \) as commonly thought (Fig 5.9a). For A310, under baffled conditions, \( N_{JCD} \) values are higher than \( N_{JS} \) values for all \( C_V \) (Fig. 5.9c). However, under unbaffled conditions, \( N_{JCD} \) values for A310 increases with increasing \( C_V \), reaches a maximum at \( C_V = 0.2 \) (v/v) before decreasing thereafter. Consequently, \( N_{JCD} \) values at \( C_V > 0.25 \) (v/v) are lower than \( N_{JS} \) values (Fig. 5.9d). Hicks et al. (1997) found that \( N_{JCD} \) values were higher than \( N_{JS} \) values at \( C_V = 0.09 \) (v/v) for axial flow impellers like four-bladed PBT and Chemineer high-efficiency impeller which are different from those used in this study. Using data from Hicks et al. (1997) and Bujalski et al. (1999), Bittorf and Kresta (2003) suggested that \( N_{JCD} > N_{JS} \) using a model to predict cloud height that only applies to purely axial flow impellers.

Cloud height values at \( N_{JS} \) normalised with liquid height (\( H_S/H \)) are shown as a function of \( C_V \) for dual-impeller systems in Figs. 5.10a and 5.10b for baffled and unbaffled vessels, respectively. The liquid level was set equal to 1.5\( T \) in all dual-impeller experiments. Similar to single-impeller systems, the cloud height increases with increasing \( C_V \) until \( H_S \) is equal to \( H \). The cloud height becomes equal to the liquid level at \( C_V = 0.15 \) (v/v) for the baffled tank and at \( C_V = 0.1 \) (v/v) for the unbaffled tank. This result is further substantiated in Fig. 5.11 which shows \( N_{JCD} \) values are either equal or slightly greater than \( N_{JS} \) values at lower \( C_V \) indicating that the cloud height would be lower than or equal to \( H \) at \( N_{JS} \). At higher \( C_V \), \( N_{JS} \) values are greater than \( N_{JCD} \) values indicating that complete particle dispersion is attained under these conditions at \( N_{JS} \).

Figs. 5.10a and 5.10b show that the cloud height values for RTRT impellers at \( N_{JS} \) in the unbaffled vessel are essentially the same as those observed in the baffled vessel. But, the cloud height values for A310PBT impellers at \( N_{JS} \) in the baffled vessel are lower than those in the unbaffled vessel at low \( C_V \) (<0.2 (v/v)). At \( C_V >0.2 \) (v/v), the cloud height for A310PBT impellers becomes equal to the liquid height in both baffled and unbaffled vessels. These results also show that the effect of impeller type on the cloud height in dual-impeller systems is marginal. At low \( C_V \), RTRT impellers at \( N_{JS} \) under baffled conditions outperformed A310PBT slightly in achieving ‘just complete dispersion’ condition while the effect is opposite under unbaffled conditions. At higher \( C_V \), there is no difference in cloud height between the two impeller types.
Figure 5.8a Normalised cloud height ($H_s/H$) while operating at $N_{JS}$ as a function of $C_V$ for single-impeller system: baffled vessel

Figure 5.8b Normalised cloud height ($H_s/H$) while operating at $N_{JS}$ as a function of $C_V$ for single-impeller system: unbaffled vessel
Figure 5.9 $N_{JS}$ and $N_{JCD}$ values as a function of $C_V$: (a) baffled vessel, RT impeller, (b) unbaffled vessel, RT impeller, (c) baffled vessel, A310, (d) unbaffled vessel, A310

Figure 5.10a Normalised cloud height ($H_s/H$) while operating at $N_{JS}$ as a function of $C_V$ for dual-impeller system: baffled vessel
Figure 5.10b Normalised cloud height ($H_s/H$) while operating at $N_{JS}$ as a function of $C_V$ for dual-impeller systems: unbaffled vessel.

Figure 5.11 $N_{JS}$ and $N_{JCD}$ values as a function of $C_V$ for dual-impeller systems: (a) baffled vessel, RTRT impeller, (b) unbaffled vessel, RTRT impeller, (c) baffled vessel, A310PBT, (d) unbaffled vessel, A310PBT.
5.3 Impeller power consumption to attain ‘just completely dispersed’ conditions

The impeller power consumption to achieve ‘just complete dispersion’ (P_{JCD}) is shown in Figs. 5.12a and 5.12b as a function of C_V under baffled and un baffled conditions, respectively for single-impeller systems. In both baffled and un baffled vessels, P_{JCD} increases with increasing C_V up to 0.2 (v/v) before decreasing slightly there after. The only exception is for A310 under baffled conditions where its P_{JCD} value keeps increasing with increasing C_V. Wang et al. (2016) observed a similar trend of increasing P_{JCD} with increasing C_V up to a value of 0.2 (v/v) under both baffled and un baffled conditions for several impellers including A310. It was observed in our experiments that N_{JCD} also increases with increasing C_V, reaches a maximum at C_V = 0.2 (v/v) before decreasing slightly thereafter for RT under both baffle arrangements and A310 under un baffled conditions (Figs. 5.9a, 5.9b and 5.9d). The only exception is that for A310 under baffled conditions where N_{JCD} increases with increasing C_V continuously (Fig. 5.9c). As the impeller power draw is a function of impeller speed, the value of N_{JCD} for a given C_V will therefore affect the P_{JCD} value. It can be seen from Figs. 5.12a and 5.12b that P_{JCD} values for RT are lower than those for A310 at any given C_V in both baffled and un baffled vessels. While it is well established that it is more energy efficient to employ axial flow impellers for achieving off-bottom solids suspension, our results indicate that RT (radial flow impeller) is more energy efficient in achieving ‘just complete dispersion’ conditions.

The impeller power consumption to achieve ‘just complete dispersion’, (P_{JCD}), in dual-impeller systems are shown in Figs. 5.13a and 5.13b for baffled and un baffled vessels, respectively. The P_{JCD} value increases with increasing C_V for both RTRT and A310PBT impeller configurations under baffled conditions (Fig. 5.13a). However, under un baffled conditions, P_{JCD} increases slightly with increasing C_V, reaches a maximum at C_V = 0.2 (v/v) and decreases slightly thereafter (Fig. 5.13b). These results can be explained using N_{JCD} results shown in Fig. 5.11. As P_{JCD} is directly proportional to N_{JCD}, an increase in N_{JCD} would lead to an increase in P_{JCD} and vice versa.

A comparison of Figs. 5.13a with 5.13b shows that there is a substantial decrease in P_{JCD} in dual-impeller systems with the removal of baffles at any given C_V for both impeller configurations used. However, the extent of the energy savings is the highest at higher C_V.
(C_V >0.2 (v/v)). These results are substantiated by other studies in the literature which showed that unbaffled tanks lead to lower specific impeller power consumption for solids suspension (Wang et al., 2012b; Davoody et al., 2016; Wang et al., 2016). Figs. 5.13a and 5.13b also show that RTRT has lower P_JCD values compared to A310PBT under both baffled and unbaffled conditions. The decrease in P_JCD for RTRT compared to A310PBT is significant (50 - 58%) under baffled conditions while it is about 40% under unbaffled conditions. The results suggest that dual-radial flow impellers are more efficient at achieving ‘just complete dispersion’ conditions than axial/mixed flow impellers in dual-impeller systems.

![Figure 5.12a P_JCD values as a function of C_V for single-impeller system: baffled vessel](image-url)
Figure 5.12b $P_{JCD}$ values as a function of $C_v$ for single-impeller system: unbaffled vessel

Figure 5.13a $P_{JCD}$ values as a function of $C_v$ for dual-impeller systems: baffled vessel
Figure 5.13b $P_{JCD}$ values as a function of $C_V$ for dual-impeller systems: unbaffled vessel

The impeller power consumption to attain varying levels of cloud heights was also measured to investigate its impact on cloud height at different solids dispersion levels and the results are shown in Figs. 5.14a and 14b for single- and dual-impeller tanks, respectively. For all experiments, $C_V$ was maintained at 0.2 (v/v) and power measurements were obtained at $H_S = 0.5H, 0.7H, 0.9H, \text{ and } H,$ and also $N > N_{JCD}$. Cloud height values shown in Fig. 5.14 indicate that radial impellers require lower power to attain at any level of cloud height than axial impellers. It can also be seen that unbaffled systems require lower power to achieve a certain level of cloud height. The cloud height is more sensitive to changes in power input for radial impellers under unbaffled conditions. This is shown as a rapid increase in $H_S/H$ when impeller power input is increased. Similar findings were reported by Wang et al. (2016) but for baffled tanks. They found that the decrease in cloud height with decrease in the impeller speed from 500 to 300 rpm is significant ($10 – 20\%$) for baffled tanks compared to unbaffled tanks.
Figure 5.14a Dispersion of solid particles at constant particle loading ($C_V = 0.2 \ (v/v)$): single-impeller system

Figure 5.14b Dispersion of solid particles at constant particle loading ($C_V = 0.2 \ (v/v)$): dual-impeller system
5.4 Optimum solids concentration, $C_{V\text{(op)}}$

The specific impeller power consumption results for solids suspension for a range of solids concentrations under different impeller and vessel configurations were discussed above. It was shown in both dual- and single-impeller systems that $\varepsilon_{JS}$ can be minimised by operating the solid-liquid mixing system at a higher solids concentration called $C_{V\text{(op)}}$ which is higher than $C_V$ values hitherto used. It has been suggested by Wang et al. (2014) that the value of $C_{V\text{(op)}}$ is dependent on the slurry flow pattern within the tank. Changes in the flow pattern can be estimated from the impeller Reynolds number, which can be expressed as

$$Re_{imp} = \frac{\rho_{\text{slurry}} N_{JS} D^2}{\eta_{\text{slurry}}}$$  \hspace{1cm} (5.1)

where $\rho_{\text{slurry}}$ is the slurry density (kg/m$^3$), $N_{JS}$ is the minimum impeller speed to achieve ‘just suspended’ condition (1/s), $D$ is the impeller diameter (m) and $\eta_{\text{slurry}}$ is the slurry viscosity (Pa.s) which can be estimated using the correlation proposed by Fedors (1974). The impact of solids concentration on $Re$ for single- and dual-impeller configurations in baffled and unbaffled vessels is shown in Fig. 5.15. It can be seen that $Re$ decreases with increasing $C_V$ regardless of baffling condition or impeller configuration. Fig. 5.15 also shows that $Re$ remains above 10000 at $C_V = 0.35$ (v/v) for all impeller types except for unbaffled vessels with RT or RTRT. These results indicate that up to a $C_V$ value of 0.35 (v/v), the flow regime in the tank is turbulent and solids suspension can still be achieved without excessive power draw at such high solids concentrations. By operating at higher $C_V$, the mixing vessel is able to achieve a higher throughput or the vessel size required to achieve the same throughput is smaller, which will lead to an increase in process efficiency or decrease in capital costs.

Wang et al. (2012a) examined various ways of optimising solid-liquid systems by considering impeller and tank geometry. They showed that the removal of baffles led to significant increases in energy efficiency. Similar observations are found in this study for both single- and dual-impeller systems. Though the removal of baffles has been shown to increase mixing time, this is not an issue in mineral processing operations because the time scales for reactions or mass transfer are still an order of magnitude greater than mixing time (Wu et al., 2010b). In addition to the removal of baffles, further energy savings can be achieved by optimising impeller design such as using radial flow impellers as discussed earlier. Table 5.1 summarises the $C_{V\text{(op)}}$ values and corresponding $\varepsilon_{JS}$. 

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Figure 5.15 Effects of $C_V$ on impeller Reynolds number ($Re_{imp}$) operating at $N_{JS}$ using viscosity ($\eta_{slurry}$) estimated using the correlation proposed by Fedors (1974)

Table 5.1 $C_{V(op)}$ values with corresponding $\varepsilon_{JS}$ values

<table>
<thead>
<tr>
<th>Impeller</th>
<th>Liquid height</th>
<th>Baffle arrangement</th>
<th>$C_V$ with lowest $\varepsilon_{JS}$ ($C_{V(op)}$) (v/v)</th>
<th>Corresponding $\varepsilon_{JS}$ (W/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RTRT</td>
<td>$H = 1.5 , D$</td>
<td>Baffled</td>
<td>0.15-0.20</td>
<td>2.31-2.33</td>
</tr>
<tr>
<td>RTRT</td>
<td>$H = 1.5 , D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>0.63</td>
</tr>
<tr>
<td>A310PBT</td>
<td>$H = 1.5 , D$</td>
<td>Baffled</td>
<td>0.15-0.25</td>
<td>4.33-4.42</td>
</tr>
<tr>
<td>A310PBT</td>
<td>$H = 1.5 , D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>1.85</td>
</tr>
<tr>
<td>RT</td>
<td>$H = D$</td>
<td>Baffled</td>
<td>0.15-0.35</td>
<td>4.84-5.20</td>
</tr>
<tr>
<td>RT</td>
<td>$H = D$</td>
<td>Unbaffled</td>
<td>0.25-0.35</td>
<td>0.83-0.86</td>
</tr>
<tr>
<td>A310</td>
<td>$H = D$</td>
<td>Baffled</td>
<td>0.20-0.30</td>
<td>2.43-2.60</td>
</tr>
<tr>
<td>A310</td>
<td>$H = D$</td>
<td>Unbaffled</td>
<td>0.20-0.35</td>
<td>0.92-0.98</td>
</tr>
</tbody>
</table>
**Summary**

The impeller power consumption for solids suspension in a mechanically agitated vessel on the basis of slurry volume, total slurry mass, and mass of solids has been investigated for a solids concentration range of 0.05 - 0.35 (v/v) at ‘just suspended’ conditions. Baffled and unbaflled vessels were used with a single impeller and dual-impellers. The impellers used in the study were RT and A310 for the single-impeller system, while RTRT and A310PBT impeller configurations were used in the dual-impeller system. Results show that impeller power input expressed on the basis of slurry volume or total slurry mass increases with increasing solids concentration for a C_V range of 0.05 – 0.35 (v/v) under both baffled and unbaflled conditions for all impeller types studied. Specific impeller power consumption, $\varepsilon_{JS}$ (expressed as power per unit mass of solids) values for single-impeller systems decrease with increasing C_V, reaches a low critical value and remains constant or increases slightly thereafter depending on impeller type and baffle condition. For dual-impeller systems, $\varepsilon_{JS}$ decreases with increasing C_V, reaches a minimum at C_V = 0.2 (v/v) and increases thereafter. This indicates that lower impeller power per unit mass of solids is required to achieve ‘just suspended’ conditions at higher solids concentrations than hitherto used.

It was also found that removing baffles led to lower $\varepsilon_{JS}$ values for the impeller configurations used in this work. Vessels fitted with radial pumping impellers were found to require less energy to satisfy N_JS conditions compared to axial pumping impellers, except in the baffled single-impeller system where A310 outperformed RT.

It was found in both single- and dual-impeller systems that cloud height (H_S) at N_JS generally increases with increasing C_V until H_S reaches the liquid surface. In the single-impeller system, cloud height values at N_JS are higher in baffled vessels especially at lower C_V ranges. Also, RT achieves higher cloud heights than A310 in single-impeller systems at any given C_V. However, in dual-impeller systems, cloud height is independent of baffling and impeller type. P_{JC} is found to increase with increasing C_V under baffled conditions while it remains roughly constant under unbaflled condition. This result is observed in both single- and dual-impeller systems under all configurations used this this work.

In order to improve energy efficiency for solids suspension or solids dispersion in an agitated vessel, three main approaches are recommended. The first approach is to operate the solid-liquid agitated vessel at higher solids concentrations (C_V > 0.2 (v/v)). The second approach is
to remove baffles and use a radial pumping impeller such as RT. The third approach is to operate a taller vessel fitted with dual-radial pumping impellers such as RTRT. It is suggested that operating a taller solid-liquid agitated vessel fitted with dual-radial pumping impellers in the absence of baffles at higher solids concentrations will improve impeller energy efficiency. This leads to an investigation on the solid-liquid mass transfer coefficient, whose results are presented in Chapter 6.
Chapter 6

Results and Discussion

Solid-liquid mass transfer in agitated vessels with single- and dual-impellers
Introduction

The work presented in Chapter 5 focused on impeller power consumption for solids suspension and solids dispersion and evaluated the performance of a mixing vessel on the basis of solids concentration, impeller type and configuration, and baffle arrangement. It was found that operating a taller vessel fitted with dual radial pumping impellers (RTRT) under un baffled conditions leads to significant energy savings when operated at higher solids concentrations than those that have been used in industry hitherto. To investigate whether further advantages could be obtained by using a dual-impeller system, solid-liquid mass transfer studies were carried out using the same solid-liquid mixing systems as those in Chapter 5.

In this chapter, the changes in $k_{SL}$ values due to the change in solids concentration were analysed for single- and dual-impeller systems. The results presented in this chapter will be useful in determining the best configuration among the baffle and impeller arrangements used in this study for achieving maximum possible solid-liquid mass transfer. A case study is also presented to highlight the benefits of adopting some of the recommendations made in this chapter.

The results presented in Chapter 6 have been submitted as part of a manuscript for peer review, and has been accepted for publication.


6.1 Results and Discussion

6.1.1 Effect of solids concentration on solid-liquid mass transfer coefficient

The solid-liquid mass transfer coefficient ($k_{SL}$) is a primary design parameter to evaluate the performance of a solid-liquid agitated vessel. Experiments were carried out in this study by varying the solids concentration to examine its effects on $k_{SL}$ for both single- and dual-impeller systems. The impellers used in this study were RT and A310 for single-impeller
systems and RTRT and A310PBT for dual-impeller systems. The $k_{SL}$ values obtained in baffled and unbaffled tanks are shown in Figs. 6.1a and 6.1b, respectively for RT and A310 in the single-impeller system. It can be seen that $k_{SL}$ increases with increasing $C_V$, reaches a maximum and decreases slightly thereafter. The $C_V$ with the highest $k_{SL}$ value is defined as $C_V(\text{max})$ and it occurs at a $C_V$ value of 0.2 (v/v) for both RT and A310. Similar findings were reported by Bong et al. (2015) who showed that the $k_{SL}$ value in a single-impeller system operating at $N_{JS}$ increased with increasing $C_V$ up to a maximum at $C_V = 0.20$ (v/v) and decreased thereafter. They attributed the increase in $k_{SL}$ with increasing $C_V$ to increasing $N_{JS}$ values and associated increased turbulence intensity levels. Turbulence intensity in the continuous phase influences advective as well as diffusional mass transfer at the solid-liquid interface. Therefore, an increase in turbulence intensity around the particle would lead to an increase in the mass transfer rate and thus the mass transfer coefficient. The decrease in $k_{SL}$ for $C_V > 0.20$ (v/v) was attributed by Bong et al. (2015) to turbulence dampening due to increasing effective viscosity of slurry ($\eta_{\text{slurry}}$) and its effect on diffusional mass transfer. In addition, particle-particle interactions become more frequent with increasing $\eta_{\text{slurry}}$, which decreases the solid-liquid interfacial area thereby decreasing the mass transfer rate (Conway et al., 2002; Tagawa et al., 2011).

The $k_{SL}$ values obtained using RTRT and A310PBT in baffled and unbaffled vessels are shown in Figs. 6.2a and 6.2b for a range of $C_V$ for dual-impeller system. It can be seen that, similar to the single-impeller systems, $k_{SL}$ at $N_{JS}$ increases with increasing $C_V$, reaches a maximum at $C_V = 0.2$ (v/v) and decreases thereafter. The $C_V(\text{max})$ values at which $k_{SL}$ is maximum in dual-impeller systems ($C_V(\text{max}) = 0.2$ (v/v)) are the same as those in a single-impeller system. Based on these results, it is concluded that operating the system at solids concentration of greater than or equal to 0.2 (v/v) will be beneficial in terms of solid-liquid mass transfer as well as impeller power consumption for both single- and dual-impeller systems.

6.1.2 Effect of impeller type on solid-liquid mass transfer coefficient

Different combination of impellers and vessel geometry may have a significant effect on mass transfer as different impellers at the same speed can generate different types of liquid flow and levels of turbulence. One of the important parameters that control the value of $k_{SL}$
values is the impeller type (Nienow and Miles, 1978). It can be seen from Figs. 6.1 and 6.2 that, under baffled conditions, RT leads to higher $k_{SL}$ values than those obtained with A310 at any given $C_V$ (Fig. 6.1a). This can be attributed to higher levels of turbulence generated by the high-shear RT compared to low-shear A310 at respective $N_{JS}$ values (Zhou and Kresta, 1996). Kumaresan and Joshi (2006) also suggested that using higher power number impellers such as RT leads to higher mass transfer coefficient values due to higher levels of turbulence that can be generated by it. Interestingly, under unbaflled condition, $k_{SL}$ values for RT and A310 are nearly equal for all $C_V$ (Fig. 6.1b).

It can also be seen from Fig. 6.2a that, under baffled condition, the $k_{SL}$ values obtained for both RTRT and A310PBT are nearly the same at any given $C_V$. The results also show that under unbaflled condition, the $k_{SL}$ values for A310PBT are higher than those for RTRT for a $C_V$ range of 0.05 - 0.35 (v/v) (Fig. 6.2b). As mentioned earlier, it was observed during experiments that particle distribution was more homogenous in the case of RTRT suggesting that the liquid flow produced by each RT impeller complimented each other and aided in particle suspension and dispersion. On the other hand, the flow generated by A310PBT under unbaflled conditions may have interfered with each other leading to an increase in levels of chaotic mixing and therefore increased mass transfer (Dutta and Pangarkar, 1995).

6.1.3 Effect of baffles on solid-liquid mass transfer coefficient

The effect of baffles on $k_{SL}$ can be seen by comparing Figs. 6.1a and 6.1b for the single-impeller system, and Figs. 6.2a and 6.2b for the dual-impeller system. The $k_{SL}$ values for RT under unbaflled conditions are lower than those under baffled conditions for all $C_V$. However, the difference in $k_{SL}$ values for A310 under unbaflled and baffled conditions is negligible for all $C_V$. These results are consistent with those reported by Bong et al. (2012) who reported that $k_{SL}$ values for A310 at $N_{JS}$ were similar under both baffled and unbaflled conditions for a $C_V$ range of 0.08 – 0.3 (v/v). Harriot (1962) also found that $k_{SL}$ values in both baffled and unbaflled tanks were similar at low solids concentration. However, Nienow et al. (1997) reported that surface aeration occurred at high impeller speeds due to vortex formation in unbaflled tanks resulting in lower mass transfer coefficient values compared to those in baffled tanks. The higher $k_{SL}$ values at a given $C_V$ for RT in baffled tanks can be attributed to higher fluctuating liquid velocities generated at higher $N_{JS}$ required for RT
(Levins and Glastonbury, 1972). Pangarkar et al (2002) also suggested that different impellers produce different levels of turbulence and therefore have different levels of influence on solid-liquid mass transfer.

It is also clear from Figs. 6.2a and 6.2b, at any given \(C_V\), baffles have minimal effect on \(k_{SL}\) especially for RTRT. There is a slight increase in \(k_{SL}\) with the removal of baffles for A310PBT. These results are in contrast to \(k_{SL}\) results obtained in single-impeller systems, where the removal of baffles led to a reduction in \(k_{SL}\) for RT and a small difference for A310. Several researchers investigated the effect of baffles on the mass transfer coefficient at high impeller speeds (Hixson and Wilkens, 1933; Barker and Treybal, 1960; Lal et al., 1988). They reported that solids were fully suspended due to greater liquid velocity fluctuations in the presence of baffles. As the impeller speeds to achieve \(N_{JS}\) in the dual-impeller systems as well as the A310 in the single-impeller system were roughly the same, the \(k_{SL}\) values obtained in these systems were also roughly the same. Harriot (1962) and Bong et al., (2012) also reported similar \(k_{SL}\) values in baffled and unbaffled vessels but at low solids concentration. As the liquid flow patterns and turbulence intensity produced by dual-impellers are different from those of single-impeller systems, the influence of baffles on the hydrodynamics and consequently on \(k_{SL}\) would also be different.

![Graph](image)

**Figure 6.1a Solid-liquid mass transfer coefficient as a function of \(C_V\) for single-impeller system: baffled vessel**
Figure 6.1b Solid-liquid mass transfer coefficient as a function of $C_V$ for single-impeller system: unbaflled vessel

Figure 6.2a Solid-liquid mass transfer coefficient as a function of $C_V$ for dual-impeller system: baffled vessel
6.2 Solids concentration with the highest $k_{SL}$ value, $C_{V(max)}$

The effect of solids concentration on $k_{SL}$ at $N_{JS}$ for single- and dual-impeller systems under different baffle and impeller arrangements has been discussed so far. The results presented above suggest that operating a solid-liquid system at higher $C_V$ helps not only in achieving significant energy savings as highlighted in Chapter 5 but it also leads to increased levels of mass transfer. As noted in Section 6.1.1, with an increase in solids concentration, $k_{SL}$ increases up to a maximum value before decreasing thereafter regardless of impeller type or baffle arrangement. The value of $C_{V(max)}$ is the same for all mixing systems studied in this work and is shown along with its corresponding $k_{SL}$ value in Table 6.1. The effects of impeller design and baffling on $k_{SL}$ in single-impeller systems are different from those in dual-impeller systems as shown in Sections 6.1.2 and 6.1.3. On comparing the results between single- and dual-impeller systems at $C_{V(max)}$, it is clear that the highest $k_{SL}$ value was obtained for RT under baffled conditions in the single-impeller system, whereas in the dual-impeller system, A310PBT under unbaffled conditions has the highest $k_{SL}$ value.
The $C_{V(\text{max})}$ for achieving the highest $k_{\text{SL}}$ value occurred at $C_{V} = 0.2$ (v/v), regardless of vessel or impeller geometry. These $C_{V(\text{max})}$ results are similar to the $C_{V(\text{op})}$ values obtained based on $\varepsilon_{\text{JS}}$ in Chapter 5. This shows that the $C_{V}$ value for the highest $k_{\text{SL}}$ coincide with the $C_{V}$ value that has the lowest $\varepsilon_{\text{JS}}$ value. These findings suggest that operating a solid-liquid agitated vessel at $C_{V} = 0.2$ (v/v) is optimum from both mass transfer and impeller power consumption criteria. Also, as shown in Chapter 5, the mixing system with the lowest $\varepsilon_{\text{JS}}$ value at $C_{V(\text{op})}$ is the RTRT system under unbaffled conditions (Table 5.1). Therefore, as the $k_{\text{SL}}$ values among the mixing systems studied are very similar, it is concluded that RTRT in the unbaffled system is the best arrangement among the systems studied in this work for achieving the lowest specific impeller power input and highest possible $k_{\text{SL}}$.

Table 6.1 $C_{V(\text{max})}$ values with corresponding $k_{\text{SL}}$ values

<table>
<thead>
<tr>
<th>Impeller</th>
<th>Liquid height</th>
<th>Baffle arrangement</th>
<th>$C_{V}$ with highest $k_{\text{SL}}$ ($C_{V(\text{max})}$) (v/v)</th>
<th>Corresponding $k_{\text{SL}}$ (mm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RTRT</td>
<td>$H = 1.5\ D$</td>
<td>Baffled</td>
<td>0.20</td>
<td>0.11</td>
</tr>
<tr>
<td>RTRT</td>
<td>$H = 1.5\ D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>0.11</td>
</tr>
<tr>
<td>A310PBT</td>
<td>$H = 1.5\ D$</td>
<td>Baffled</td>
<td>0.20</td>
<td>0.12</td>
</tr>
<tr>
<td>A310PBT</td>
<td>$H = 1.5\ D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>0.13</td>
</tr>
<tr>
<td>RT</td>
<td>$H = D$</td>
<td>Baffled</td>
<td>0.20</td>
<td>0.13</td>
</tr>
<tr>
<td>RT</td>
<td>$H = D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>0.11</td>
</tr>
<tr>
<td>A310</td>
<td>$H = D$</td>
<td>Baffled</td>
<td>0.20</td>
<td>0.10</td>
</tr>
<tr>
<td>A310</td>
<td>$H = D$</td>
<td>Unbaffled</td>
<td>0.20</td>
<td>0.10</td>
</tr>
</tbody>
</table>
6.3 Improved mass transfer rate and energy efficiency: a case study

To illustrate the benefits of adopting some of the recommendations made in earlier sections of this chapter, a case study is presented here for the parameters shown in Table 6.2. A hypothetical benchmark design, which is considered as the ‘standard’ design for solid-liquid systems (Wang et al., 2014), is used as the existing design in this case study. Wang et al. (2014) suggested that solid-liquid mixing tanks in the minerals industry are fully baffled and operate at relatively low solids concentration. Based on this, 0.1 (v/v) is chosen as the operating $C_V$ and single A310 is chosen as the impeller for the standard design.

The new designs for both single- and dual-impeller systems are also shown in Table 6.2 to highlight the difference between the two. Both mixing vessels include radial pumping impellers under unbaffled conditions. The vessels are operated at a solids concentration $C_V = 0.2$ (v/v), which is significantly higher than that in the standard design. The impeller speed to ‘just suspend’ and ‘just completely disperse’ solids for the new designs are found to be lower than those in the standard design resulting in significant savings in power usage. It should also be noted that the degree of solids dispersion (cloud height) and $k_{SL}$ increase in the new designs. For both single- and dual-impeller systems, the cloud height and $k_{SL}$ values are very similar. However, $\varepsilon_{JS}$ and $P_{JCD}$ values are different from each other for the two new designs. Though the power consumption to achieve ‘just complete dispersion’ conditions ($P_{JCD}$) in the dual-impeller system is greater than that in single-impeller system, the lower value of $\varepsilon_{JS}$ implies better energy utilisation for solids suspension in the dual-impeller system.
Table 6.2 Vessel/Impeller dimensions and design parameters for an existing (standard) and new solid-liquid mixing vessel designs. Values of design parameters were determined in this study

<table>
<thead>
<tr>
<th>Specifications</th>
<th>Standard design</th>
<th>New design (single impeller)</th>
<th>New design (dual impellers)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tank diameter (T)</td>
<td>0.2 m</td>
<td>0.2 m</td>
<td>0.2 m</td>
</tr>
<tr>
<td>Liquid height (H)</td>
<td>H = T</td>
<td>H = T</td>
<td>H = 1.5T</td>
</tr>
<tr>
<td>Baffles</td>
<td>4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Impeller</td>
<td>A310</td>
<td>RT</td>
<td>RTRT</td>
</tr>
<tr>
<td>Impeller diameter (D)</td>
<td>T/3.125</td>
<td>T/3</td>
<td>T/3</td>
</tr>
<tr>
<td>Solids concentration (C_v)</td>
<td>0.1 (v/v)</td>
<td>0.2 (v/v)</td>
<td>0.2 (v/v)</td>
</tr>
<tr>
<td>Agitator speed for just off-bottom solids suspension (N_{JS})</td>
<td>730 rpm</td>
<td>400 rpm</td>
<td>430 rpm</td>
</tr>
<tr>
<td>Agitator speed for complete dispersion of solids (N_{JCD})</td>
<td>760 rpm</td>
<td>400 rpm</td>
<td>390 rpm</td>
</tr>
<tr>
<td>Cloud height (H_S/H) at N_{JS}</td>
<td>0.84</td>
<td>0.97</td>
<td>1.0</td>
</tr>
<tr>
<td>Specific impeller power consumption at N_{JS} (\varepsilon_{JS})</td>
<td>3.94 W/kg</td>
<td>0.88 W/kg</td>
<td>0.63 W/kg</td>
</tr>
<tr>
<td>Power consumption at N_{JCD} (P_{JCD})</td>
<td>2.98 W</td>
<td>1.16 W</td>
<td>1.44 W</td>
</tr>
<tr>
<td>Solid-liquid mass transfer coefficient (k_{SL})</td>
<td>0.097 mm/s</td>
<td>0.11 mm/s</td>
<td>0.11 mm/s</td>
</tr>
</tbody>
</table>
Summary

The solid-liquid mass transfer coefficient ($k_{SL}$) values in single- and dual-impeller systems were determined experimentally using a NaOH - cation exchange resin system at $N_{JS}$ for a range of $C_V$ (0.05 – 0.35 (v/v)) under baffled and unbaffled conditions. The impellers used in this study were RT, A310, RTRT and A310PBT. The results showed that with increasing $C_V$, $k_{SL}$ increases, reaches a maximum at $C_V = 0.2$ (v/v) and decreases again for both single- and dual-impeller systems. Experiments showed that higher values of $k_{SL}$ can be obtained at solids concentrations higher than hitherto used in industry. The removal of baffles leads to reduction in $k_{SL}$ for RT whereas $k_{SL}$ increases when A310PBT is employed. Baffling has no effect on $k_{SL}$ when other impeller configurations are used. Higher $k_{SL}$ values are observed for RT compared to A310 in single-impeller systems. Between the two impeller configurations used in the dual-impeller system, A310PBT leads to higher $k_{SL}$ values.

When $\varepsilon_{JS}$ values are considered along with $k_{SL}$ values, the unbaffled vessel fitted with RTRT is shown to be the most economical configuration because it leads to the lowest $\varepsilon_{JS}$ values with comparable $k_{SL}$ values. A case study is presented highlighting the benefits of adopting some of the recommendations made in this chapter. Two new designs involving a single-impeller system and a dual-impeller system are compared to a standard mixing system commonly used in industry. The new design involving the dual-impeller system outperforms the existing design in terms of energy consumption as well as mass transfer.
Chapter 7

Mathematical correlations to estimate the impeller power consumption for solids suspension & solids dispersion, and solid-liquid mass transfer coefficient
Introduction

In Chapter 4, it was shown that the Zwietering correlation can be extended up to a solids concentration of 0.35 (v/v) for baffled and unbaffled vessels agitated with single- and dual-impeller configurations. Chapters 5 and 6 discussed the effect of solids concentration, impeller type and baffle arrangement on impeller energy consumption and solid-liquid mass transfer, respectively. As estimation of the impeller power consumption and the solid-liquid mass transfer coefficient are essential for the reliable design of solid-liquid agitated vessels, Chapter 7 presents mathematical correlations for the estimation of these important design parameters. Due to the complexity of the flow patterns within the slurry system and the large number of variables that may affect it, especially in agitated vessels fitted with multiple impellers, accurate estimation of $k_{SL}$ is difficult. Although there are a large number of correlations available in the literature to predict $k_{SL}$, a majority of them are applicable to systems similar to the ones used in deriving the correlation. An aim of this study is to add to the body of knowledge by deriving design correlations for estimating $k_{SL}$ and impeller power consumption in single- or dual-impeller agitated vessels operating at high solids concentration under baffled and unbaffled conditions. Experimental values of $k_{SL}$ obtained from mass transfer experiments carried out in this study are used in developing the design correlations.

As some solid-liquid mixing operations in industry require solids to be homogenously suspended throughout the agitated vessel, a simple correlation to estimate the power required to ‘just completely disperse’ solids is proposed. The design correlation is to be added to the body of knowledge for estimating the impeller power consumption in single- or dual-impeller agitated vessels operating at high solids concentration under baffled and unbaffled conditions. The mathematical correlations presented in Chapter 7 have been submitted as part of a manuscript for peer review, and has been accepted for publication.

7.1 Correlation for estimating the specific impeller power consumption, $\varepsilon_{JS}$

Recently, Bong et al. (2015) has shown that the impeller power consumption at $N_{JS}$ can be determined using the following equation:

$$P = N_P \rho_{slurry} N_{JS}^3 D^5$$  \hspace{1cm} (7.1)

where $N_P$ is the impeller power number (which can be found in Table 7.1 for the impellers used in this work), $\rho_{slurry}$ is the density of the slurry (kg/m$^3$) which is calculated using $\rho_{slurry} = \rho_s C_v + \rho_L (1 - C_v)$ and $D$ is the impeller diameter (m).

To account for the influence of particles on impeller hydrodynamics and therefore impeller power consumption, Bong et al. (2015) incorporated the parameter $k$ into Eq. (7.1) and when combined with Eq. (3.3), obtained the following equation:

$$\varepsilon_{JS} = \frac{P}{M_S} = \frac{k N_P \rho_{slurry} N_{JS}^3 D^5}{M_S}$$  \hspace{1cm} (7.2)

The parameter $k$ is a function of vessel and impeller geometries and is shown in Table 7.1 for the mixing systems studied in this work. These values were obtained using experimental data for the impeller power and speed and performing a back-solving analysis.

The critical impeller speed $N_{JS}$ can be estimated using the Zwietering correlation (Eq. 2.2). The $X$ term in Eq. (2.2) can be expressed as a function of $C_v$ by the equation $X = \left(\frac{\rho_s}{\rho_L}\right)^{100 C_v} (1-C_v)$ (Wang et al., 2014). Though the exponent for $X$ varies, it is equal to 0.13 according to Zwietering (1958). The same value of 0.13 is assumed applicable for solids concentration from 0.05 to 0.35 (v/v) and is used in this work. Even though Eq. (2.2) was derived using solid-liquid mixing vessels fitted with a single impeller, Dutta and Pangarkar (1995) and this work (Chapter 4) showed that the empirical correlation for $N_{JS}$ for single-impeller systems can also be applied to agitated reactors fitted with multiple impellers.

By substituting Eq. (2.2) into Eq. (7.2), the following equation for the specific impeller power consumption is obtained:

$$\varepsilon_{JS} = k \left(\frac{N_P \rho_{slurry} D^5}{M_S}\right) \left(\frac{S_U^{0.1} d_P^{0.2}}{\rho_L^{0.45}} \frac{g(\rho_s - \rho_L)}{D^{0.85}} \left(\frac{\rho_s}{\rho_L}\right)^{100 C_v} \left(1-C_v\right)^{0.13}\right)^3$$  \hspace{1cm} (7.3)
where $\varepsilon_{JS}$ is the specific impeller power consumption (W/kg), $k$ is a constant, $N_P$ is the impeller power number, $\rho_{\text{Slurry}}$ is the slurry density (kg/m$^3$), $D$ is the impeller diameter (m), $M_S$ is the mass solids (kg), $S$ is the Zwietering constant, $\nu$ is the kinematic viscosity of the slurry (m$^2$/s), $d_p$ is the particle diameter (m), $g$ is the gravitational acceleration constant (m/s$^2$), $\rho_S$ and $\rho_L$ are the densities of the solid and liquid phases, respectively (kg/m$^3$) and $C_V$ is the volumetric solids concentration (v/v).

Figs. 7.1a and 7.1b compared experimental data obtained in this study with values obtained using Eq. (7.3) for single- and dual-impeller systems, respectively. It can be seen that the values estimated by Eq. (7.3) are in good agreement with experimental data for both single- and dual-impeller systems with the vast majority of data points situated within ± 15% band.

**Figure 7.1a** Comparison of experimental data in solid-liquid systems with values estimated using Eq. (7.3): single-impeller system
7.2 Correlation to estimate the solid-liquid mass transfer coefficient, $k_{SL}$

There are many correlations reported in the literature for estimating the solid-liquid mass transfer coefficient in agitated vessels over a wide range of Reynolds numbers. Most of the mathematical correlations are in the form of the following equation (Levins and Glastonbury, 1972; Kato et al., 1998; Bong et al., 2015):

$$Sh = 2 + ARe^{0.5}Sc^{0.33}$$  \hspace{1cm} (7.4)

where $Sh$ is the Sherwood number, $Re$ is the Reynolds number, $Sc$ is the Schmidt number and $A$ is a constant. $A$ is determined using a regression analysis of the experimental data.

Sherwood number ($Sh$) is a dimensionless number which is the ratio of the convective mass transfer to diffusive mass transfer. It can be defined as:

$$Sh = \frac{k_{SL}d_p}{D_A}$$  \hspace{1cm} (7.5)

The contribution of asymptotic molecular diffusion from a fixed sphere based on film theory is taken into account in Eq. (7.4) by the use of the constant 2 (Armenante and Kirwan, 1989).
This value is widely used in mathematical correlations for mass transfer in agitated slurries involving spherical particles.

Schmidt number (Sc) is the ratio of the kinematic viscosity to the molecular diffusivity:

\[ Sc = \frac{\mu_L}{\rho_L D_A} \]  

(7.6)

where \(d_p\) is the particle diameter (m), \(D_A\) is molecular diffusivity \((m^2/s)\), \(\mu_L\) is the liquid viscosity \((Pa.s)\) and \(\rho_L\) is the liquid density \((kg/m^3)\).

Based on boundary layer theory, the exponent of the Schmidt number is chosen as 0.33, though it varies from 0 - 0.5 depending on hydrodynamic conditions (Harriot, 1962; Levins and Glastonbury, 1972). Many investigators using a wide range of solids concentrations have agreed that the exponent for the Reynolds number should be equal to 0.5 for solid-liquid mixing systems and is therefore chosen in this work (Harriot, 1962; Levins and Glastonbury, 1972; Cline, 1978).

The particle Reynolds number used in Eq. (7.4) is based on Kolmogoroff’s theory of isotropic turbulence and is defined by Eq. (7.7) (Hinze, 1975):

\[ Re = \left(\frac{\varepsilon d_p^4}{\nu^3}\right)^{1/3} \]  

(7.7)

where \(\varepsilon\) = energy dissipation rate per unit mass liquid \((W/kg)\), \(d_p\) is the particle diameter (m) and \(\nu\) is the kinematic viscosity of the liquid \((m^2/s)\).

The energy dissipation rate per unit mass liquid, \(\varepsilon\), is defined as follows:

\[ \varepsilon = \frac{P}{M_L} \]  

(7.8)

where \(P\) is the impeller power consumption \((W)\) and \(M_L\) is the total mass liquid in the tank \((kg)\).

By substituting Eq. (2.2), Eq. (7.1), Eq. (7.8) and incorporating \(k\) into Eq. (7.7), the following equation for the modified particle Reynolds number \((Re_p)\) is obtained as a function of \(C_V\):

\[ Re_p = \left(\frac{k(\frac{Np}{M_L})^{5/4} \left(\frac{\rho}{\rho_L}\right)^{0.15} \left(\frac{\rho}{\rho_L}(1-C_V)^{0.13}\right)}{d_p^{0.45} \left(\frac{\rho}{\rho_L}\right)^{0.85}} \right)^{1/3} \]  

(7.9)
Eq. (7.9) can be used to determine Reₚ in the absence of experimental impeller power consumption data as it’s a function of solids volume fraction Cᵥ.

The mass transfer correlation (Eq. 7.4) can now be rewritten as:

\[
Sh = 2 + ARe_p^{0.5}Sc^{0.33} \tag{7.10}
\]

The value of A is obtained by rearranging Eq. (7.10) and is as follows:

\[
\frac{Sh - 2}{Sc^{0.33}} = ARe_p^{0.5} \tag{7.11}
\]

By plotting \(\frac{Sh - 2}{Sc^{0.33}}\) versus \(Re_p^{0.5}\) in an x-y graph, the value of A can be determined as shown in Fig. 7.2a and 7.2b for single- and dual-impeller systems, respectively. It can be seen that the plots for all the mixing systems studied are not linear. \(\frac{Sh - 2}{Sc^{0.33}}\) increases with increasing \(Re_p^{0.5}\), reaches a maximum, and decreases thereafter. This trend is similar to the experimental results for \(k_{SL}\) with increasing Cᵥ. This is expected as Sh is a function of \(k_{SL}\) (Eq. 7.5) and \(Re_p^{0.5}\) accounts for changes in Cᵥ (Eq. 7.9).

![Figure 7.2a](image-url)

**Figure 7.2a (Sh-2)/Sc^{0.33} versus Re_p^{0.5} for Cᵥ = 0.05 - 0.35 (v/v), continuous lines represent equation:**

\[
\frac{Sh - 2}{Sc^{0.33}} = -a(Re_p^{0.5})^2 + b(Re_p^{0.5}) - c: \text{single-impeller system}
\]

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A mathematical equation that represents the parabolic trend of the data points was determined by carrying out a non-linear regression analysis. The parabolic nature of the data point trend indicated that it could be simulated mathematically using a quadratic equation:

\[
\frac{Sh-2}{Sc^{0.33}} = -a\left(Re_p^{0.5}\right)^2 + b\left(Re_p^{0.5}\right) - c \tag{7.12}
\]

From Eq. (7.11) and Eq. (7.12), we get

\[
A = (-a\left(Re_p^{0.5}\right) + b - c\left(Re_p^{0.5}\right)) \tag{7.13}
\]

The constants a, b and c can be determined by performing a regression analysis. The values of a, b and c are shown in Table 7.1 for the mixing systems studied in this work. To determine the difference between calculated and experimental values, the mean absolute % error is commonly used and it can be calculated using the following equation (Mayer and Butler, 1993):

\[
\text{mean absolute % error} = \frac{1}{N} \sum \left| \frac{k_{SL(cal)} - k_{SL(exp)}}{k_{SL(exp)}} \right| \times 100\% \tag{7.14}
\]

where N is the number of data points, \(k_{SL(cal)}\) is the solid-liquid mass transfer coefficient value estimated using Eq. (7.15) and \(k_{SL(exp)}\) is the experimental solid-liquid mass transfer coefficient value.
coefficient value. The mean absolute % error calculated for each set of experiments is shown in table 7.1.

Table 7.1 Parameters in Eq. (2.2), (7.2), (7.9), (7.14) and (7.15)

<table>
<thead>
<tr>
<th></th>
<th>RTRT</th>
<th>A310PBT</th>
<th>RT</th>
<th>A310</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Baffled</td>
<td>Unbaffled</td>
<td>Baffled</td>
<td>Unbaffled</td>
</tr>
<tr>
<td>S</td>
<td>5.8</td>
<td>5.5</td>
<td>10.5</td>
<td>10.5</td>
</tr>
<tr>
<td>k</td>
<td>1.1</td>
<td>1.1</td>
<td>1.1</td>
<td>1.1</td>
</tr>
<tr>
<td>N_p</td>
<td>6.5</td>
<td>2.3</td>
<td>2.3</td>
<td>1</td>
</tr>
<tr>
<td>a</td>
<td>0.81</td>
<td>2.55</td>
<td>1.46</td>
<td>1.85</td>
</tr>
<tr>
<td>b</td>
<td>7.0</td>
<td>17.6</td>
<td>14.1</td>
<td>14.9</td>
</tr>
<tr>
<td>c</td>
<td>9.38</td>
<td>24.7</td>
<td>27.6</td>
<td>23.0</td>
</tr>
<tr>
<td>Average error (%)</td>
<td>2.1</td>
<td>5.1</td>
<td>5.9</td>
<td>3.3</td>
</tr>
</tbody>
</table>

By substituting Eq. (7.13) into Eq. (7.10), the following equation is obtained:

\[ Sh = 2 + (-a(Re_p^{0.5}) + b - c(Re_p^{-0.5})Re_p^{0.5}Sc^{0.33}) \]  \hspace{1cm} (7.15)

The Sh values obtained from experimental k_{SL} values and those calculated using Eq. (7.15) are compared in a parity plot as shown in Fig. 7.3. It can be seen that all data points are situated within the ±10% band. Based on these results, we propose that Eq. (7.15) can be used to estimate k_{SL} values for mixing systems similar to this work for a C_v range of 0.05 to 0.35 (v/v). More experimental work needs to be carried out if Eq. (7.15) is to be used for estimating k_{SL} for C_v values outside the range studied in this work as well as for mixing systems with different tank and/or impeller geometry.
7.3 Mathematical correlation to estimate impeller power consumption to just completely disperse solids, $P_{JCD}$

In a well-dispersed multiphase system, the impeller power consumption can be defined using Eq. (7.1) (Wang et al., 2016). As the two-phase systems investigated in this study are not completely homogenous due to how ‘just completely dispersed’ was defined, a fitting parameter, $k_D$ is incorporated into Eq. (7.1) to account for the axial solids concentration gradient and can now be written as:

$$P_{JCD} = k_D N_p \rho_{sturry} N_{JCD}^3 D^5$$  \hspace{1cm} (7.16)

where $k_D$ is a constant that takes the influence of the solids concentration gradient into account. The values of $k_D$ were obtained in this work using back-solving approach.

The power consumption estimated using Eq. (7.16) were found to be in agreement with experimental data up to a C V range of 0.2 (v/v) in both baffled and unbaffled vessels (Wang et al., 2016). Bubbico et al. (1998) proposed a similar correlation in which the k value was shown to be highly dependent on the impeller type, particle shape and size. Their work investigated the dispersion of very large particles ($d_p = 1.29$ to $4.3$ mm) in a baffled vessel.
with a $C_V$ of up to 0.15 (v/v). As this work aims to investigate power consumption in both baffled and unbaffled vessels at high $C_V$, the equation used by Wang et al. (2016) will be used in this study.

The $P_{ICD}$ values estimated using $k_D$ values shown in Table 7.2 and the experimental $P_{ICD}$ values are shown in Figs. 7.4a to 7.4d for both single- and dual-impeller systems. It can be seen that the estimated values are in good agreement with experimental data but with some deviation at higher $C_V$ for A310 under baffled conditions (Fig. 7.4a). When the experimental and estimated $P_{ICD}$ values are also plotted in a parity plot for single- and dual-impeller systems, respectively (Figs. 7.5a and 7.5b), it can be seen that the majority of data points are situated within the ±15% band. Generally, the $k_D$ values in baffled vessels are ≈1 whereas they are higher in unbaffled vessels. This could be attributed to a non-uniform particle distribution under unbaffled condition which affects the local slurry concentration in the vicinity of the impeller leading to increased impeller power draw.

### Table 7.2 Parameter $k_D$ values used in Eq. (7.16) to estimate $P_{ICD}$

<table>
<thead>
<tr>
<th></th>
<th>RT</th>
<th>A310</th>
<th>RT</th>
<th>A310</th>
<th>Baffled</th>
<th>Unbaffled</th>
<th>Baffled</th>
<th>Unbaffled</th>
<th>Baffled</th>
<th>Unbaffled</th>
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<th>Unbaffled</th>
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</thead>
<tbody>
<tr>
<td>$k_D$</td>
<td>1.1</td>
<td>3.1</td>
<td>1.3</td>
<td>1.1</td>
<td>1.0</td>
<td>1.4</td>
<td>1.0</td>
<td>1.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 7.4a Experimental and estimated $P_{\text{JCD}}$ values as a function of $C_V$: baffled vessel, single-impeller system

Figure 7.4b Experimental and estimated $P_{\text{JCD}}$ values as a function of $C_V$: unbaffled vessel, single-impeller system
**Figure 7.4c** Experimental and estimated $P_{JCD}$ values as a function of $C_V$: baffled vessel, dual-impeller system

**Figure 7.4d** Experimental and estimated $P_{JCD}$ values as a function of $C_V$: unbaffled vessel, dual-impeller system
Figure 7.5a Comparison of estimated and experimental values of $P_{\text{JCD}}$: single-impeller system

Figure 7.5b Comparison of estimated and experimental values of $P_{\text{JCD}}$: dual-impeller system
Summary

A mathematical model to estimate the specific impeller power consumption in solid-liquid agitated vessels fitted with single and dual impellers was developed satisfactorily using experimental data. Estimated values were in good agreement with the experimental data for both baffled and unbaffled conditions for a $C_V$ range of $0.05 - 0.35$ (v/v) with the vast majority of data points situated within a 15% band.

Using experimental values of $k_{SL}$, impeller power consumption, and solids concentration data, a correlation for estimating the solid-liquid mass transfer coefficient $k_{SL}$ was derived. The correlation proposed in this study estimated values that were found to match experimental results for a $C_V$ range of 0.05-0.35 (v/v) for all vessel geometries used in this work. The mean absolute % error between experimental and estimated values of $k_{SL}$ was found to vary from 1.1 to 5.9 %, depending on the impeller and tank geometry. It should be noted that the correlation is applicable to solid-liquid mixing systems similar to the ones used in the study. It is not recommended to extrapolate the results for systems with more impellers or for vessels with diameters other than 0.2 m or with $C_V > 0.35$ (v/v) without further investigations.

Also, a mathematical correlation to estimate power consumption required for particle dispersion was developed from experimental data for both single- and dual-impeller systems for a $C_V$ range of $0.05 - 0.35$ (v/v). Estimated values for both baffled and unbaffled vessels were found to fit experimental data reasonably well within a ±15% band.
Chapter 8

Results and Discussion

Particle influence on cavitation
Introduction

In Chapter 5, the effects of solids concentration, impeller type and baffle arrangement on impeller energy consumption for solids suspension were discussed. It was shown that increased energy efficiency is achieved by operating a solid-liquid agitated at $C_V \geq 0.2$ (v/v). In Chapter 6, maximum $k_{SL}$ values were also obtained when operating a solid-liquid agitated vessel at $C_V \geq 0.2$ (v/v). The results from these chapters suggest that process intensification of solid-liquid agitated vessels can be achieved not only through optimising impeller/vessel geometry, but also by operating at higher $C_V$ ranges than hitherto used in industry. As the use of cavitation shows promising signs of improving solid-liquid mass transfer (Schueller and Yang, 2001; Breitbach et al., 2003; Hamdaouie et al., 2003), it would be valuable to investigate the effect of high solids concentration on cavitation with the aim of using the generated knowledge in intensifying existing industrial processes for improving the throughput. In this chapter, the impact of particle size, type, and concentration on a sonochemical reaction is studied using a reaction in which OH$^-$ radicals produced due to cavitation react with each other to form hydrogen peroxide in a solution of potassium iodide. The hydrogen peroxide then oxidises an iodide ion to form iodine, which subsequently reacts with excess iodide ions to form $I_3^-$. Measuring the rate by which $I_3^-$ is formed provides an indication of cavitation activity (Koda et al., 2003).

The results presented in Chapter 8 have been submitted as part of a manuscript for peer review, and is under review.


8.1 Results and Discussion

8.1.1 Effect of solids concentration on ultrasonic intensity and hydrogen peroxide formation in aqueous potassium iodide

The sonochemical reaction involving the production of iodine from KI solution was conducted to study its dependence on particle concentration. The size and type of particles
(ion-exchange resins) were fixed, and all experiments were conducted at NJS to ensure the availability of maximum surface area for irradiation. UV-Vis spectrometer absorbance values for I$_3^-$ are shown in Fig. 8.1 as a function of C$_V$ (0 to 0.5 (v/v)) at various time intervals. It can be seen that the absorbance of I$_3^-$ increases with increasing irradiation time for all C$_V$. Similar results have previously been reported by other researchers (Mason and Peters, 2002; Koda et al., 2003). It can also be observed that the absorbance decreases with increasing C$_V$ up to 0.1 (v/v) and then increases depending on the sampling intervals. However, the absorbance either starts remaining constant or decreasing after C$_V$ = 0.4. These results indicate that the presence of dispersed particles can affect the inception and growth of cavities in the liquid medium. However, the varying trends of cavitational activity (as indicated by absorbance) with increasing C$_V$ have not been reported in the literature to our knowledge.

The decrease in cavitational activity with increasing solids concentration, as seen in Fig. 8.1 for C$_V$ < 0.1, has been reported by other researchers. Gogate et al. (2004) found that increasing concentrations of TiO$_2$ up to 500 ppm results in a decrease of phenol degradation via cavitation and suggested that this is due to the dominant effect of wave scattering and attenuation. Other researchers found that the addition of particles at C$_V$ < 0.1 has a detrimental effect on cavitation due to the substantial prevention of sound propagation (Lu and Weavers, 2002; Lu et al., 2002). Based on similar considerations, the decrease in absorbance with increasing C$_V$ up to 0.1 (v/v) observed in the present work can be ascribed to the attenuation of ultrasonic wave by particles, despite the increase in the number of nucleation sites with increasing C$_V$.

At C$_V$ > 0.1 (v/v), the increasing absorbance or the enhanced sonochemical yield with increasing C$_V$ can be ascribed primarily to increased number of nucleation sites that are available with a higher number of particles. Apparently, the decrease in the cavitation threshold with increasing C$_V$ leads to a greater number of low energy cavitation. As a result, for 0.1 < C$_V$ < 0.4, the net effect of increased particle concentration on cavitation is positive because the effect of reduced cavitation threshold becomes more dominant than that of wave attenuation.

At all intervals, the highest absorbance values in this work were obtained at C$_V$ = 0.4 (v/v), beyond which the absorbance starts decreasing again. It can be attributed to an increase in the apparent viscosity of the solid-liquid mixture with increasing solids concentration. It is
widely published in the literature that ultrasonic cavitation is affected by a range of parameters including solvent properties (Mason, 1992). Liu et al. (2010) has reported that as the liquid viscosity increases the cavity radius decreases but the oscillation time or bubble life increases, which would together retard cavitation rates. It has also been stated that higher apparent viscosity of the solid-liquid mixture can also cause bubbles to coalesce more frequently in a three-phase mixing system (Tagawa et al., 2011). Even though the size of the bubbles in a three-phase mixing tank are larger compared to cavitation bubbles which are in the range of microns, it has been shown that cavitation bubbles do coalesce (Ashokkumar, 2011). The formation of larger cavitation bubbles could increase the bubble life and thereby decrease cavitation rates. Hence, Fedors’ (1974) correlation (Chapter 5) was used in this work to estimate the apparent viscosity of slurry for a range of CV. The apparent slurry viscosity estimated as a function of CV is shown in Fig.5.2, where it can be seen that there is an exponential increase in apparent viscosity as CV increases. The apparent viscosity at CV of 0.5 (v/v) is an order of magnitude greater than that at CV < 0.1 (v/v) and about three times greater than at CV of 0.4 (v/v). Based on these apparent viscosity values, it can be said that the increase in apparent viscosity at CV = 0.5 (v/v) is the main limiting factor in reducing cavitation rates. Another interesting observation from Fig. 8.1 is that the difference in absorbance for CV of 0.4 and CV of 0.5 (v/v) decreases as irradiation time increases. There is no clear explanation available in the literature about this phenomenon, and therefore further research is required to understand the causes for this result.

Figure 8.1 Impact of solids concentration (CV) on I₃⁻ absorbance, Solid phase: ion-exchange resin, Liquid phase: 0.1M KI solution
Fig. 8.2 shows the SEM images of the ion-exchange particles that have been subjected to ultrasound irradiation. The attenuation of ultrasonic waves and therefore the decrease in the intensity of the propagated wave can be indirectly seen in the figure. At $C_V$ of 0.01 (v/v), a majority of the resin particles that experienced combined agitation and ultrasound irradiation are fractured (Fig. 8.2e). The extent of particle fracturing decreases as solids concentration increases, which can be seen by comparing the number of unfractured particles in Figs. 8.2e, 8.2f and 8.2g. This finding suggests that particle fracturing is the result of high energy cavitation, which occurs at low $C_V$ due to the higher cavitation threshold. At low $C_V$, the cavitation threshold is higher due to fewer available nucleation sites and higher liquid tensile strength, which leads to cavitation events of a higher intensity. In the absence of ultrasound, but under only agitation, the resin particles did not fracture at all $C_V$, which suggests that particle fracturing is solely due to cavitation, not due to particle-particle interaction (Figs. 8.2b to 8.2d).

![Figure 8.2 SEM images of ion exchange resin after agitation or combined agitation and ultrasonic irradiation at different $C_V$. a: Prior to agitation, $C_V$ 1%. b-d: Agitation only, $C_V$ 1, 5 and 30%, respectively. e-g: Combined agitation and ultrasound, $C_V$ 1, 5 and 30%, respectively](image)

To confirm that changes observed in absorbance (formation of $I_3^-$) data shown in Fig. 8.1 are mainly due to ultrasound, experimental results for combined agitation and ultrasonic
irradiation are compared with those for agitation alone in Fig. 8.3. It can be seen that no I$_3^-$ formed at any C$_v$ under silent conditions (agitation only) regardless of the irradiation time.

![Figure 8.3 UV-Vis absorbance by I$_3^-$ under ultrasonically irradiated and silent conditions at different C$_v$](image)

**Figure 8.3 UV-Vis absorbance by I$_3^-$ under ultrasonically irradiated and silent conditions at different C$_v$**

### 8.1.2 Effect of particle size on cavitation rates in an ultrasonically irradiated potassium iodide medium

The influence of particle size on cavitation events was investigated using spherical glass beads with diameters of 206, 551 and 1290 µm. The volumetric solids concentration was fixed at 0.01 (v/v) as the measured absorbance values are relatively high enough allowing for any difference in absorbance to be easily analysed. The transient changes in absorbance values are shown in Fig. 8.4. It can be seen that cavitation activity increases with increasing particle size. The effect becomes significant as irradiation time increases. The effect of particle size on cavitation can be further confirmed by analysing the SEM images shown in Fig. 8.5, which show the surface topography of selected glass particles after 10 minutes of combined agitation and ultrasonic irradiation. The surface of particles remains relatively smooth when only agitation is used (Figs. 8.5a to 8.5c). The crater observed in Fig. 8.5b may have been the result of a particle-particle collision. However, under ultrasound irradiation, the particle surface becomes rougher and the roughness increases with increasing particle size.
(Figs. 8.5d to 8.5f). The types of deformation that occurred on the particle surface changed with increasing particle size. The deformation was in the form of streaks for 206 µm particles, flaky for 551 µm particles, and deep crevices for 1290 µm particles. All these deformations could potentially act as additional nucleation sites.

An increase in particle size corresponds to a decrease in the interfacial surface area, which should result in a reduction in cavitational activity. However, Borkent et al. (2007) showed that the influence of particle type and surface roughness is more important in determining cavitation yield than surface area. It has also been shown that the presence of suspended particles decreases the tensile strength of a liquid and the decrease is greater with increasing particle size (Marschall et al., 2003). The decrease in liquid tensile strength results in higher number of cavitation events due to lowering of the cavitation threshold. It has been stated that particles greater than 150 µm can act as a wall thereby causing asymmetric bubble collapse resulting in microjet formation (Doktycz and Suslick, 1990). The asymmetric collapse leads to the generation of a large number of tiny bubbles that lead to enhanced sonochemical yields. Other researchers have also shown that an increase in particle size leads to a decrease in ultrasonic wave attenuation thereby allowing more cavitation to occur (Zanwar and Pangarkar, 1988; Romdhane et al., 1997). Thus, the decrease in liquid tensile strength and ultrasonic wave attenuation with increasing particle size can be attributed to increased levels of cavitation. This increase in cavitation leads to greater erosion of the particle surface thereby creating an increased number of nucleation sites and subsequent increase in sonochemical yield.
8.1.3 Effect of particle type on cavitation rates

To investigate how the type of particle surface affects cavitation activity, three particle types namely, cation exchange resin, spherical glass beads, and sand were used as the solid
phase in this work. The transient absorbance results for the three types of particle are shown in Fig. 8.6 for a volumetric solids concentration of 0.01 (v/v). The effect of particle size is considered to be negligible as compared to the dominant effect of solid surface type. Clearly, sand is subjected to greater cavitation activity compared to resin and glass particles. This phenomenon can be further confirmed from the SEM images of particle surface shown in Fig. 8.7. The surface of sand is rough and has an undulating topography (Fig. 8.7c). In contrast, the resin and glass particles have relatively smooth surfaces (Figs. 8.7a and 8.7b). These results suggest that surface roughness is a major factor in determining cavitation rates due to its effect on the number of available nucleation sites. Similar results have been reported by Borkent et al. (2007), who suggested that surface roughness is important in facilitating the entrapment and stabilization of gas pockets on the particle surface which act as nucleation sites. Using fluorescence intensity to measure cavitation activity in suspensions containing microparticles ($d_p = 5\mu m$), Haosheng et al. (2009) showed that the fluorescence intensity resulting from irradiated suspensions containing spherical balls was almost the same as that obtained in suspensions containing irregular shaped particles. They suggested that particle shape has little effect on cavitation activity for particles of similar size and that their results may not be applicable for larger or smaller sized particles. Belova et al. (2011) also showed that cavitation activity is dependent on the hydrophobicity of a particle surface with larger number of nucleation sites on hydrophobic surfaces enhancing cavitation. Even though all the particles used in this study can be deemed as hydrophilic, the differences in cavitation activity for these particles could be due to different levels of surface roughness they have.
Figure 8.6 Effect of particle surface properties on $I_3^-$ absorbance at $C_V = 0.01$ (v/v)

Figure 8.7 SEM images of different particles after combined agitation and ultrasonic irradiation. a: Ion exchange resin. b: Glass beads. c: Sand
Summary

The influence of particle size, surface roughness, and high solids concentration on cavitation activity was investigated experimentally in a stirred solid-liquid vessel using cationic exchange resin, spherical glass beads, and sand particles when they are suspended off the tank bottom under ultrasonic irradiation. Experimental results showed that cavitation activity increases with irradiation time for all solids concentration used. The extent of cavitation decreases initially with increasing solids concentration due to wave attenuation but increases after that due to the availability of additional nucleation sites with a higher number of particles. The increase in cavitation activity reaches a maximum at $C_V$ of 0.4 (v/v) before decreasing again probably due to higher apparent viscosities of the suspension, which probably leads to the coalescence of cavitating bubbles and therefore increased oscillation time. Spherical glass particles with diameters of 206, 551 and 1290 µm were used to investigate the effect of particle size on cavitation, and it was found that cavitation activity increases with increasing particle size. It was found that particles with rougher surfaces can initiate greater cavitation levels due to their ability to facilitate the entrapment and stabilization of gas pockets on their surface. These results indicate that the presence of particles at higher concentrations helps to enhance cavitation, and particle size and surface roughness play important roles in enhancing sonochemical yields. This leads us to an investigation on enhancing solid-liquid mass transfer with cavitation, the results of which are reported in Chapter 9.
Chapter 9

Results and Discussion

Solid-liquid mass transfer with ultrasound
Introduction

In Chapters 5 and 6, it was shown that process intensification of solid-liquid agitated vessels can be achieved by operating at higher CV ranges. Additionally, Chapter 8 showed that the extent of cavitation decreases initially with increasing solids concentration due to wave attenuation but increases after that due to the availability of additional nucleation sites with a higher number of particles. Following the results obtained in these chapters, this chapter aims to investigate the effects of combined ultrasound and agitation on solid-liquid mass transfer in 1) an ion exchange system and 2) a desorption system at high CV. The findings will help to evaluate whether process intensification of solid-liquid mixing vessels can be achieved not only by increasing the solids concentration but also by increasing the solid-liquid mass transfer yield through the use of ultrasound.

The results presented in Chapter 9 are to be submitted as part of a manuscript for peer review.


9.1 Results and Discussion

9.1.1 Effect of cavitation on solid-liquid mass transfer rates

9.1.1.1 Ion exchange-NaOH system

Mass transfer experiments were conducted to study the effect of cavitation on ion exchange operations. The solid-liquid system used in this study involves NaOH solution and cation exchange resin as described in section 3.2.8. All experiments were done in an agitated vessel operating at NJS to ensure the entire solids surface area was available for mass transfer. The concentration of sodium ions in the liquid phase as a function of time under silent and irradiated conditions are shown in Fig. 9.1 for CV = 0.03 (v/v). It can be seen that cavitation has no effect on the ion exchange rate. The rate-controlling step of ion exchange is diffusion through the liquid boundary layer around the particle. The results obtained in this study suggest that the contribution of cavitation in decreasing the boundary layer is not significant.
and agitation alone is sufficient. Cheng and Wang (1982) compared the kinetics of ion exchange under mechanical agitation and ultrasonic irradiation. They found that ion exchange rate due to mechanical stirring is greater than that for ultrasound. In a later study, Entezari and Tahmasbi (2009) found that higher ion exchange rate was achieved due to cavitation at lower CV (CV = 0.0016 and CV = 0.004 (v/v)). They found that the rate of mass transfer is higher under both irradiated and silent conditions for CV = 0.004 (v/v) than for CV = 0.0016 (v/v), which they attributed to increased number of sorption sites. Therefore, Entezari and Tahmasbi suggested that the sorbate/sorbent ratio is an important factor in ion exchange. It should be noted that Entezari and Tahmasbi used very low solids concentrations and only ultrasound in contrast to the present work which used both ultrasound and mechanical agitation with high CV.

![Figure 9.1 Sodium ion concentration in the liquid phase as function of time, CV = 0.03 (v/v)](image)

**Figure 9.1 Sodium ion concentration in the liquid phase as function of time, CV = 0.03 (v/v)**

### 9.1.1.2 Phenol desorption from polymeric resin system

The solid-liquid system used in this study involves polymeric resin and water as described in section 3.2.9. Phenol resin particles, called by the trade name as Dowex Optipore resin, were saturated with phenol adsorbate, and added to deionised water and then subjected to mechanical agitation and ultrasonic irradiation. When polymeric resin particles are ‘just
suspended’ off the tank bottom, phenol is desorbed from resin into water. The rate at which the phenol was desorbed from the solid phase into the liquid phase was determined by removing liquid samples at regular intervals and analysing them using UV-Vis spectrometer (Thermo Scientific Helios Zeta) at 270nm. Fig. 9.2 shows the change in phenol concentration in the liquid phase as a function of time for $C_V = 0.03$ (v/v) for agitation plus irradiation and agitation only. The rate of increase in phenol concentration in the liquid phase for combined agitation and irradiation is greater than that for agitation alone. The increase in phenol concentration is rapid at the start of the experiment for both conditions. Similar observations were made by Rege et al. (1998) who found that phenol desorption rate is greater for ultrasonic irradiation compared to agitation. They also observed the rapid initial increase in phenol concentration in the liquid phase and suggested that this was caused by the diffusion of phenol retained in the macropores of solids into the bulk liquid. To determine the role of cavitation, Rege et al. (1998) performed a desorption experiment with degassed water and found that the desorption rate was higher than that obtained using agitation alone but lower than that obtained when using deionised water. Based on this experiment, they suggested that cavitation plays a significant role in desorbing phenol from polymeric resin because the number of cavities produced due to ultrasonic irradiation will be substantially lower in degassed liquids. Li et al. (2006) have also found that, under irradiated condition, the rate of phenol desorption from polymeric resin increases and the time required to reach equilibrium decreases.
9.1.2 Effect of solids concentration on solid-liquid mass transfer rates in solid-liquid agitated systems irradiated with ultrasound

9.1.2.1 Ion exchange- NaOH system

The influence of solids concentration on ion exchange was investigated under combined agitated and irradiated condition to determine how it affects the mass transfer at high solids loadings. The volumetric solids concentrations used in this study were \( C_V = 0.03, 0.1, 0.15 \) and 0.2 (v/v). The mass of sodium ions exchanged from the liquid phase to solid phases is plotted in Fig. 9.3 as mass of \( Na^+ \) per mass of resin as a function of time. The mass of \( Na^+ \) ion in the liquid phase was obtained by measuring the conductivity of the liquid phase and converting it into \( Na^+ \) ion concentration using a pre-determined calibration constant. The mass ratio decreases rapidly with increasing time and reaches zero eventually regardless of \( C_V \) for combined agitation & irradiation and agitation only. It can be seen that irradiation does not have any significant influence either on the trend of the mass ratio curve or on the final steady state value of the mass ratio. However, the initial value of the mass ratio and the time required to reach the final steady state value are different for different \( C_V \). The initial mass ratio at \( t=0 \) decreases with increasing \( C_V \) and these values are 15.5, 4.3, 2.7 and 1.9 for \( C_V = 0.03, 0.1, 0.15 \) and 0.2 (v/v), respectively. Similarly, the times at which the mass ratio reaches the final steady state value are 140, 40, 23 and 16 seconds for \( C_V = 0.03, 0.1, 0.15 \) and 0.2 (v/v), respectively. The decrease in time required in reaching the final equilibrium value (≈zero) with increasing \( C_V \) can be attributed to increased number of ion exchange sites due to increased number of particles while the NaOH concentration in the liquid phase remains constant. It is clear from these results that including irradiation in addition to agitation does not improve the rate of mass transfer noticeably compared to agitation only condition and therefore unnecessary.
Figure 9.3 Mass of sodium ions exchanged per mass resin as a function of time. a: $\text{CV} = 0.03$ (v/v). b: $\text{CV} = 0.10$ (v/v). c: $\text{CV} = 0.15$ (v/v) d: $\text{CV} = 0.20$ (v/v)

9.1.2.2 Phenol desorption from polymeric resin

The mass of phenol desorbed into the liquid phase per mass of resin is plotted as a function of time in Fig. 9.4 for combined agitation & irradiation and agitation only conditions. During desorption, the concentration of phenol in the liquid phase increases with increasing time until it reaches a steady value, which is a function of $\text{CV}$. The equilibrium mass ratio values are 1.1, 0.39, 0.33, and 0.33 for $\text{CV} = 0.03$, 0.1, 0.15 and 0.2 (v/v), respectively under agitation only condition. It can be seen that equilibrium is not reached until after 10 minutes under combined agitation and irradiation. The mass ratio values under combined agitation and irradiation after 10 minutes are 1.6, 0.6, 0.5 and 0.4 for $\text{CV} = 0.03$, 0.1, 0.15 and 0.2 (v/v), respectively. It can be seen that, for all $\text{CV}$, the mass ratio values are higher for combined irradiation and agitation showing clearly the additional influence of irradiation on the rate and extent of mass transfer.

The time required for the mass ratio to reach the equilibrium value decreases with increasing $\text{CV}$. The % increase in the amount of phenol desorbed per mass of resin under combined agitation and irradiation compared to agitation can be calculated using equation 9.1.
\[ E(\%) = \frac{(MR_{US} - MR_{Silent})}{MR_{Silent}} \times 100\% \quad (9.1) \]

where \( E(\%) \) is the percentage increase or enhancement in mass ratio, \( MR_{US} \) is the amount of phenol desorbed per mass of resin in the presence of ultrasound and \( MR_{Silent} \) is the amount of phenol desorbed per mass of resin under silent conditions. \( E(%) \) is plotted as a function of time in Fig. 9.5 for \( C_v = 0.03, 0.1, 0.15 \) and \( 0.2 \) (v/v). \( E(%) \) decreases slightly with increasing time initially and then starts increasing for all \( C_v \). The highest \( E(%) \) value is observed for \( C_v = 0.15 \) (v/v) for most of the times. The \( E(%) \) values for \( C_v = 0.10 \) (v/v) are nearly equal to those for \( C_v = 0.15 \) (v/v) and those for \( C_v = 0.03 \) (v/v) are slightly lower than those for 0.15 (v/v). The lowest \( E(%) \) values are obtained for \( C_v = 0.2 \) (v/v) at all times. It is interesting to see that \( E(%) \) values for \( C_v = 0.2 \) (v/v) are negative between the time intervals 0 and 180 seconds, and they show increasing trend with time after that. These results indicate that higher \( E(%) \) values for mass transfer are achieved using irradiation with agitation for \( C_v < 0.15 \) (v/v). Since \( E(%) \) is dependent on \( C_v \), the optimisation of the mass transfer process needs to take into consideration the dependence of \( E(%) \) on \( C_v \).

![Figure 9.4 Mass phenol desorbed per mass resin as a function of time. a: Agitation only. b: Combined agitation and ultrasonic irradiation](image-url)
Figure 9.5 The percentage increase in the amount of phenol desorbed per mass of resin in the presence of ultrasonic irradiation.

9.2 Discussion

Solid-liquid agitated vessels are widely used in industry where there is a strong demand to intensify operations due to increasing financial constraints. Process intensification can address this issue by increasing product throughput. Process intensification can be achieved by operating solid-liquid agitated vessels at higher solids concentration or by achieving higher yields. However, to increase the rate and the extent of solid-liquid mass transfer beyond what can be achieved just by agitation alone, an intensive and local energy release method such as ultrasound needs to be used. This increase, however, is dependent on many factors including the physical properties of the solid and liquid phases. This is clear from the results for ion exchange and polymeric resin systems discussed above. While ultrasound has made a significant impact on the mass transfer (up to 55% enhancement) in the case of polymeric resin, hardly any enhancement was found in the case of ion exchange system.

Compared to other solids such as polymeric resin, activated carbon, and metal ores, ion exchange resin is less porous which could restrict the phenomenon of vortex microstreaming within the pores and therefore minimise its influence on solid-liquid mass transfer. Vortex microstreaming has been suggested as one of the mechanisms that contribute to increased mass transfer rates in some solid-liquid systems under irradiated conditions (Rege et al.,
The exchange of ions between ion exchange resin and a free ion in the liquid phase is a diffusion-controlled physisorption process identified to have a low heat of adsorption (approx. 8 kJ/mol) and activation energy (24-40 kJ/mol) values (Inglezakis and Zorpas, 2012). These values are less than those estimated for sorption processes involving polymeric resin and phenol (activation energy value of 40-50 kJ/mol) (Rege et al., 1998). Therefore, it can be suggested that, due to the low energy required to break the affinity between the adsorbent and adsorbate in an ion exchange system, the impact of cavitation is not apparent and the energy transferred due to agitation is sufficient to achieve the mass transfer. The impact of cavitation is therefore more apparent in solid-liquid mass transfer systems with higher activation energies or heat of adsorption such as the polymeric resin/phenol system. As the bonding forces between phenol and activated carbon are higher than those between phenol and polymeric resin, the effect of cavitation would be apparent also in sorption processes involving phenol and activated carbon (Caetano et al., 2009).

The size distribution of ion exchange resin particles used in this study did not change with the application of ultrasound (Chapter 3). This is in contrast to some previous studies in which the average particle size decreased after ultrasonic irradiation thereby increasing the surface area available for interphase mass transfer (Hagenson and Doraiswamy, 1998; Lim and Okada, 2005; Avvaru et al., 2006; Durbha and Aravamudan, 2012). The decrease in particle size would result in increased mass transfer rates due to increased particle surface area.

During the leaching of minerals from ore, a product layer can form on particle surface which could decrease the contact area between the mineral surface and the leaching agent. It has been suggested that cavitation is able to renew or remove the passivation (product) layer thereby leading to increased leaching rates (Hagenson and Doraiswamy, 1998; Okur et al., 2002). It has also been proposed that the shockwaves and microjets produced by cavitation can cause cracks on the particle, through which the leaching agent can enter the interior of the particle by capillary action and thereby improve the mass transfer rate (Narayana et al., 1997).

Unlike previous studies, this work investigated the effect of ultrasound on solid-liquid mass transfer at higher solids concentrations. The presence of large amount of particles can either promote or suppress cavity formation and in turn can influence any ultrasonic enhancement on mass transfer. The substantial increases in solid-liquid mass transfer due to ultrasound cavitation shown by previous authors were obtained in small vessels operated at very low $C_V$. 

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As a consequence, the ultrasonic power per unit volume of slurry is very high and therefore the ultrasonic intensity propagating through the liquid is also high. In such case, a majority of power input does not increase the mass transfer but goes to increase the temperature of the medium. This is in contrast to the experimental procedure adopted in this study, which may explain the results shown in this chapter.

9.3 Addition of energy into a solid-liquid mixing vessel via mechanical mixing and cavitation

The concept of increasing materials throughput using existing mixing tanks whilst also achieving improved economic returns was first introduced by ICI in the early 1980s (Ramshaw, 1985). This concept is known as ‘process intensification’ and is a way of reducing capital and operating costs in a chemical production plant. Wu et al. (2010b) redefined process intensification as increasing production yield per unit volume and per unit time and per unit cost. Process intensification in mixing tanks is an attractive approach to enhance the productivity of mixing vessels because slurry vessels used for mineral processing are typically large and energy intensive.

Process intensification can be achieved by increasing the solids throughput or by increasing the extraction yield through efficient mixing or by supplying additional energy. The addition of energy into a solid-liquid mixing vessel discussed in this work is in the form of mechanical mixing and cavitation via ultrasound. Investigating which form of energy input is more efficient at achieving process intensification will determine the advantages of one method over the other.

Using the methods outlined in Chapter 3, the ultrasonic power consumption and the impeller power consumption at $C_{V(OP)}$ obtained for different impeller and vessel geometries are shown in Table 9.1. The ultrasonic transducer emitted 131W of energy into the solid-liquid mixing vessel, whereas the impeller power consumption ranged from 1.2 - 8.7 W at $N_{JS}$ and $C_{V(OP)} = 0.2 (v/v)$. It is clear from the results shown here that the addition of energy via cavitation is an order of magnitude greater than that of mechanical mixing alone without any proportional benefit in mass transfer. This was the case under both baffled and unbaffled conditions and all impeller configurations. Based on these results, it may be concluded that process
intensification is achieved more efficiently through adding additional energy via mechanical mixing.

Though the addition of ultrasound has shown that it can enhance solid-liquid mass transfer, there still lie inherent limitations to its widespread use in industry. The primary limitation is the utilisation of only a small fraction of the acoustic power in producing cavities. This inevitably results in higher energy input. Coupled with low ultrasonic wave propagation, much of a large agitated vessel remains void of sonication and therefore unaffected by cavitation. This has made the method of delivery of ultrasound into a solid-liquid vessel very important. Though there are custom-made agitated vessels fitted with multiple ultrasonic transducers to increase the volume irradiated with ultrasound, undoubtedly much remains to be done before ultrasound is employed in industry to enhance solid-liquid mass transfer.

Table 9.1 Comparison between the energy consumption of ultrasonic irradiation and mechanical agitation

<table>
<thead>
<tr>
<th></th>
<th>Ultrasound</th>
<th>RTRT unbaffled</th>
<th>RT unbaffled</th>
<th>A310PBT baffled</th>
<th>RT baffled</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power consumption (W)</td>
<td>131</td>
<td>1.3</td>
<td>1.2</td>
<td>8.7</td>
<td>6.5</td>
</tr>
</tbody>
</table>
Summary

Solid-liquid mass transfer under combined agitation and ultrasonic irradiation was investigated experimentally using an ion exchange and a phenol desorption system. Results show that ultrasonic waves at a frequency of 20 kHz and an intensity of 131 W did not affect the average Sauter-mean diameter of resin particles used in both systems (Chapter 3). The results also showed that combined agitation and ultrasound had no effect on mass transfer rates in the ion exchange system for a CV range of 0.03 – 0.20 (v/v). However, the rate and extent of phenol desorbed into water increased in the presence of ultrasound. The amount of phenol desorbed per unit mass of resin (mg/g) decreased with increasing CV under both irradiated and silent conditions. The enhancement in mass transfer E (%) in the phenol-water system with the addition of ultrasound over agitation increased with time and was dependent on CV. Greater E (%) values were obtained at CV = 0.15 and 0.10 (v/v). These results suggest that increased solid-liquid mass transfer in the presence of ultrasound is dependent on the physical and chemical properties of the solid and liquid phases as well as CV.

The ultrasonic power consumption and the impeller power consumption at CV(OP) obtained for different impeller and vessel geometries were compared to determine which method was more efficient at achieving process intensification of solid-liquid agitated vessels. The ultrasonic transducer emitted 131W of energy into the solid-liquid mixing vessel, whereas the impeller power consumption ranged from 1.2 - 8.7 W at NJS for CV(OP) = 0.2 (v/v). From these results, it was clear that mechanical agitation was more efficient at intensifying solid-liquid agitated vessels.
Chapter 10
Conclusions and recommendations
10.1 Conclusions

In many mineral processing industry operations, it is desired to intensify solid-liquid agitated vessels in order to process more materials per unit volume and per unit time and per unit cost. This can be achieved by the addition of energy in form of mechanical agitation or cavitation.

This study, through extensive experimentation, systematically evaluated the effect of solids concentration on specific impeller power consumption and solid-liquid mass transfer coefficient in mechanically agitated vessels fitted with dual impellers. The study focused on dual-impeller agitated systems operating at just suspended conditions for solids concentrations up to 0.35 (v/v), which is higher than values hitherto used in industry. An objective of this study was to also determine the best baffle and impeller configuration to achieve solids suspension, dispersion, and solid-liquid mass transfer. Another aim of this work was to determine if the application of the Zwietering correlation could be extended to predict $N_{JS}$ reliably in dual-impeller systems operating at higher solids concentrations and under different baffle and impeller configurations. Another objective of this work is to develop mathematical correlations for estimating the specific impeller power consumption, the solid-liquid mass transfer coefficient, and the impeller power consumption to disperse solids in agitated vessels over a wider range of solids concentration for different impeller/vessel geometries.

One of the main aims of this work is to determine if cavitation could increase solid-liquid mass transfer in agitated vessels with the expectation that it will be used to intensify existing processes. As there is growing literature on the benefits of operating solid-liquid agitated vessels at higher solids concentrations, another objective of this study is to determine how cavitation is affected by the presence of particles at high solids concentrations and solids of different size and type. This work is beneficial to industry because it can provide useful information in the design of solid-liquid agitated vessels fitted with dual impellers that can handle high solids concentrations. Also this work would be of interest to a broad range of engineers and scientists, including those interested in acoustic cavitation and how it is influenced by solids at high concentrations. A schematic flow chart is presented to show the main outcomes and sequential organisation of the thesis.
Figure 10.1A schematic flow chart of main outcomes and sequential organisation of this work
The following conclusion can be drawn from this study:

- The Zwietering correlation for predicting $N_{JS}$ in solid-liquid agitated systems can be reliably applied to vessels fitted with dual impellers up to a $C_V$ of 0.35 (v/v). This is true for unbaffled as well as baffled vessels.
- Impeller energy efficiency improved by operating both single- and dual-impeller systems at higher solids concentration ($\geq 0.2$ (v/v)).
- The impeller power consumption to just disperse solids increases with increasing $C_V$ under baffled conditions while it remained largely constant under unbaflled condition.
- The removal of baffles helps to improve impeller energy efficiency regardless of impeller type and concentration used.
- Of the vessel/impeller geometries used, the dual RT impeller configuration outperformed the other configurations in terms of impeller energy efficiency.
- The solid-liquid mass transfer coefficient at $N_{JS}$ increased with increasing $C_V$ up to $C_V = 0.2$ (v/v) and decreased thereafter for both single- and dual-impeller systems.
- The removal of baffles led to a decrease in $k_{SL}$ for RT and an increase in $k_{SL}$ for A310PBT. Baffling had no effect on $k_{SL}$ when other impeller configurations were used.
- The differences in $k_{SL}$ values obtained for the mixing systems used in this work were marginal for a $C_V$ range of 0.05 to 0.35 (v/v). Therefore, based on the specific impeller power consumption, the RTRT in an unbaffled system was shown to be the best baffle and impeller configuration.
- A mathematical correlation was developed to estimate $\varepsilon_{JS}$ in single- and dual-impeller systems. Also a correlation based on Sherwood number and Kolmogoroff’s theory of isotropic turbulence was developed to estimate $k_{SL}$ as a function of $C_V$. Also, a mathematical correlation to estimate the impeller power consumption for particle dispersion was developed. The estimations of all mathematical correlations were found to agree with experimental data well with an overall average error of 15% for a $C_V$ range of 0.05 – 0.35 (v/v).
- Cavitation activity in a solid-liquid slurry decreases with increasing $C_V$ up to 0.1 (v/v) and increases thereafter under combined agitation and ultrasonic irradiation.
- Cavitation activity increases with increasing particle size. Surface roughness plays an important role in enhancing sonochemical yields.
Combined agitation and ultrasonic irradiation did not have an effect on mass transfer rates over agitation in the ion exchange system. However, the rate of phenol desorption into water from polymeric resin increased in the presence of ultrasound. The enhancement of mass transfer due to ultrasound was the greatest at $C_V = 0.1$ and $0.15 \, (v/v)$. This suggests that increased solid-liquid mass transfer in the presence of ultrasound is dependent on the physical and chemical properties of the solid and liquid phases as well as $C_V$.

Process intensification is achieved efficiently through the addition of energy via mechanical agitation rather than cavitation.

### 10.2 Recommendations

The following are the recommendations for possible further work:

- The study can be extended to determine whether the Zwietering correlation can be used in solid-liquid systems with different rheology such as systems with more viscous or non-Newtonian liquids.
- The present study increased liquid height and kept the vessel diameter constant. Further investigations can be performed on wider vessels fitted with dual impellers to determine its viability in achieving process intensification.
- The study used a single transducer to irradiate the slurry. Further investigations into enhancing mass transfer with ultrasound can be performed using vessels fitted with multiple transducers as this may help to alleviate the effect of ultrasonic wave scattering.
- This study used a model system to replicate ultrasonic addition into solid-liquid slurry. Further investigations can be done using materials commonly encountered in industry, such as mineral ore and its corresponding lixiant as the solid and liquid phases, respectively.
References


Cline, H. B. 1978. A correlation for predicting the mass transfer coefficient in agitated vessels that accounts for the effect of solids concentration on the particle slip velocity. University of Maryland, Maryland, USA.


Eilers, H. 1941. The viscosity of emulsions of highly viscous substances as function of the concentration. Colloid Zh. 97, 313-321.


Appendix

Figure A.1 \( \ln([C_{Na}] / [C_{Na}]) \) versus t curve used for determining mass transfer coefficient, \( k_{SL} \).

Figure A.2 Liquid temperature change with and without ultrasonic irradiation used to determine ultrasonic power input.
Table A.1 Raw experimental $M_S$, $M_L$, $N_{JS}$, $P_{JS}$, $N_{JCD}$, $P_{JCD}$ and $k_{SL}$ data for RTRT

| CV (v/v) | $M_S$ (g) | $M_L$ (L) | $N_{JS}$ (rpm) | $P_{JS}$ (W) | $N_{JCD}$ (rpm) | $P_{JCD}$ (W) | $k_{SL}$ (mm/s) | CV (v/v) | $M_S$ (g) | $M_L$ (L) | $N_{JS}$ (rpm) | $P_{JS}$ (W) | $N_{JCD}$ (rpm) | $P_{JCD}$ (W) | $k_{SL}$ (mm/s) |
|----------|-----------|-----------|----------------|--------------|-----------------|--------------|----------------|----------|-----------|-----------|----------------|--------------|-----------------|--------------|----------------|     |
| 0.05     | 502.2     | 8.0       | 300            | 1.8          | 310             | 1.6          | 0.094          | 0.05     | 516.0     | 8.2       | 295             | 0.49          | 350             | 1.0          | 0.075          |
| 0.10     | 1004.4    | 7.5       | 360            | 2.7          | 370             | 2.5          | 0.097          | 0.10     | 1032.0    | 7.7       | 340             | 0.69          | 370             | 1.2          | 0.083          |
| 0.15     | 1506.6    | 7.1       | 400            | 3.5          | 420             | 3.4          | 0.098          | 0.15     | 1548.0    | 7.3       | 390             | 0.99          | 400             | 1.4          | 0.094          |
| 0.20     | 2008.8    | 6.7       | 450            | 4.7          | 440             | 3.9          | 0.112          | 0.20     | 2064.0    | 6.9       | 430             | 1.29          | 390             | 1.4          | 0.114          |
| 0.25     | 2511.0    | 6.3       | 500            | 6.1          | 470             | 5.0          | 0.109          | 0.25     | 2580.0    | 6.5       | 470             | 1.68          | 380             | 1.3          | 0.112          |
| 0.30     | 3013.2    | 5.9       | 540            | 7.4          | 490             | 5.5          | 0.103          | 0.30     | 3096.0    | 6.0       | 510             | 2.17          | 365             | 1.3          | 0.100          |
| 0.35     | 3515.4    | 5.4       | 600            | 9.9          | 510             | 6.1          | 0.101          | 0.35     | 3612.0    | 5.6       | 560             | 2.99          | 345             | 1.2          | 0.099          |

Table A.2 Raw experimental $M_S$, $M_L$, $N_{JS}$, $P_{JS}$, $N_{JCD}$, $P_{JCD}$ and $k_{SL}$ data for A310PBT

| CV (v/v) | $M_S$ (g) | $M_L$ (L) | $N_{JS}$ (rpm) | $P_{JS}$ (W) | $N_{JCD}$ (rpm) | $P_{JCD}$ (W) | $k_{SL}$ (mm/s) | CV (v/v) | $M_S$ (g) | $M_L$ (L) | $N_{JS}$ (rpm) | $P_{JS}$ (W) | $N_{JCD}$ (rpm) | $P_{JCD}$ (W) | $k_{SL}$ (mm/s) |
|----------|-----------|-----------|----------------|--------------|-----------------|--------------|----------------|----------|-----------|-----------|----------------|--------------|-----------------|--------------|----------------|     |
| 0.05     | 502.2     | 8.0       | 570            | 3.1          | 630             | 3.7          | 0.084          | 0.05     | 516.0     | 8.2       | 560             | 1.5          | 540             | 1.9          | 0.089          |
| 0.10     | 1004.4    | 7.5       | 710            | 5.2          | 730             | 5.5          | 0.094          | 0.10     | 1032.0    | 7.7       | 640             | 2.1          | 570             | 2.0          | 0.095          |
| 0.15     | 1506.6    | 7.1       | 780            | 6.7          | 800             | 7.1          | 0.104          | 0.15     | 1548.0    | 7.3       | 730             | 3.0          | 600             | 2.3          | 0.118          |
| 0.20     | 2008.8    | 6.7       | 860            | 8.7          | 880             | 9.2          | 0.127          | 0.20     | 2064.0    | 6.9       | 810             | 3.8          | 610             | 2.6          | 0.130          |
| 0.25     | 2511.0    | 6.3       | 920            | 11.0         | 930             | 11.1         | 0.115          | 0.25     | 2580.0    | 6.5       | 930             | 5.2          | 590             | 2.3          | 0.127          |
| 0.30     | 3013.2    | 5.9       | 1060           | 16.3         | 950             | 11.9         | 0.106          | 0.30     | 3096.0    | 6.0       | 1070            | 6.6          | 560             | 2.1          | 0.124          |
| 0.35     | 3515.4    | 5.4       | 1110           | 17.3         | 920             | 12.3         | 0.102          | 0.35     | 3612.0    | 5.6       | 1160            | 8.1          | 500             | 1.8          | 0.121          |
Table A.3 Raw experimental $M_S$, $M_L$, $N_{JS}$, $P_{JS}$, $N_{JCD}$, $P_{JCD}$ and $k_{SL}$ data for RT

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<th>$P_{JS}$ (W)</th>
<th>$N_{JCD}$ (rpm)</th>
<th>$P_{JCD}$ (W)</th>
<th>$k_{SL}$ (mm/s)</th>
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<th>CV (v/v)</th>
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<th>$M_L$ (L)</th>
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<th>$P_{JS}$ (W)</th>
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<th>$k_{SL}$ (mm/s)</th>
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Table A.4 Raw experimental $M_S$, $M_L$, $N_{JS}$, $P_{JS}$, $N_{JCD}$, $P_{JCD}$ and $k_{SL}$ data for A310

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