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HIGH FREQUENCY ULTRASONIC ASSISTED MASS-TRANSFER FOR WATER BATCH REACTOR

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ABSTRACT

Ultrasonic assisted mass transfer is one of the potential alternatives that possess the advantages of high mass transfer enhancement with absence of moving part. A higher frequency ultrasonic irradiation generates a higher streaming turbulent force. However, most of the current studies are performed under low frequency ultrasonic irradiation. In this paper, a high frequency ultrasonic assisted mass transfer was studied in a water batch reactor. The mass transfer coefficient was determined by the dynamic pressure step method. The power of the ultrasonic irradiation was measured using calorimetric method. The result showed that, approximate 28 % power from the electrical dissipation power has been converted to the ultrasonic irradiation power. Besides, based on the experimental result, the mass transfer coefficient has been significantly enhanced by the ultrasonic irradiation up to 20 times faster for CO_2 desorption as compared to the case without ultrasonic irradiation.

Keywords: ultrasonic enhancement, water, mass transfer, high frequency, desorption.

INTRODUCTION

Ultrasonic wave is a traditional technology used to enhance the multiphase reactions, which is mainly attributed to its physical kinetic effect. The promising ultrasonic applications have been developed for solid cleaning, liquid chemical reactions, emulsions, humidifier and others (Cravotto *et al.*, 2005; Thompson and Doraiswamy, 1999; Thompson and Doraiswamy, 1999). Besides, several research studies have been performed on the ultrasonic-assisted study for the mass transfer process (Laugier *et al.*, 2008; Schueller and Yang, 2001; Gondrexon *et al.*, 1997: Kumar, *et al.*, 2005). This is due to the streaming turbulence effect caused by the ultrasonic power, which referring to the acceleration effect of liquid phase under a strong and high frequency ultrasonic waves (Lighthill, 1978; Loh *et al.*, 2002). Higher ultrasonic frequency generates the stronger streaming turbulence effect (Rudenko *et al.*, 1998). The effect can be functioned as an agitator. This has resulted in the advantages of ultrasonic assisted gas-liquid mass transfer over conventional agitation batch reactor by having higher enhancement factor with the absence of moving part. The conventional agitation involves the mechanically moving part which having the disadvantages of complicated system in high pressure condition and difficulty in crystallization process.

As reported by Lighthill (1978), the ultrasonic streaming force can be increased by the ultrasonic frequency. Besides, the ultrasonic frequency has created the "acoustic fountain" effect through atomization and fountain formation (Gondrexon *et al.*, 1997). Acoustic fountain is the phenomena where liquid droplet pinched out by the strong vibration on the gas-liquid interfacial. It is noted that, the generated liquid droplet with smaller diameter size provides a large surface area for the mass transfer process. Meanwhile, the size of the droplet can be varied with the ultrasonic frequency (Yasuda *et al.*, 2011). Therefore, higher frequency generates smaller droplet and thus increasing the surface area for the mass transfer process (Avvaru *et al.,* 2006). To date, most of the current studies are performed under low frequency ultrasonic waves (20 kHz) for the desorption purpose (Gantert and Moller, 2012; Tanaka *et al.*, 2014). Hence, the present study focuses on the effect of high frequency ultrasonic waves of 1.7 MHz in mass transfer process. A custom-made ultrasonic system was designed prior to the gas-liquid mass transfer study for CO_2 desorption. The power of the ultrasonic was measured using calorimetric method. The mass transfer coefficient was determined experimentally using dynamic pressure step method.

THEORY AND BACKGROUND

Ultrasonic power system

A high frequency ultrasonic system was fabricated using a piezoelectric transducer. For the power system, a voltage adjustable power supply has been assembled to suit for the ultrasonic piezoelectric transducer with 1.7 MHz and 14 mm diameter, which functioned as a convertor that convert the electrical power to ultrasonic waves. The ultrasonic piezoelectric transducer was mounted in an ultrasonic holder with direct contact configuration with the distilled water without transmission horn. The ultrasonic total dissipation power was directly measured from the electrical dissipation across the ultrasonic piezoelectric transducer, which is expressed as in Equation (1):

$$P_T = \frac{{V_{rms}}^2}{R} \quad (1)$$

where P_T is the total dissipation power, which is included of ultrasonic irradiation power and the heat loss during the ultrasonic irradiation generation, V_{rms} is the roof mean square of the supplied high frequency alternative voltage to the ultrasonic piezoelectric transducer, which was measured by a high frequency AC voltage meter, and R is the efficient resistance of ultrasonic piezoelectric transducer, which was determined using digital oscilloscope Tektronix TDS 210.

The correlation of the total dissipation power with ultrasonic power can be expressed as in Equation (2):

$$P_{us} = kP_T \tag{2}$$

where P_{us} is the actual ultrasonic power transmitted to the liquid and k is the coefficient of the ultrasonic power to total electrical dissipation power.

The actual ultrasonic power transmitted to the liquid can be determined using calorimetric method. This can be performed by assembling a simple reactor to measure the increase in temperature under ultrasonic irradiation. Following this, the ultrasonic power can be determined from the temperature increasing rate as in Equation (3):

$$P_{us} = V_L \rho_L C_L \frac{dT}{dt} \tag{3}$$

where V_L is the volume of liquid, $\frac{dT}{dt}$ is the initial rate of temperature increase under ultrasonic irradiation, C_L is the heat capacity of liquid and ρ_L is liquid mass density. The experiment was conducted by using different volume of liquid with corresponding to the different height of liquid level in order to take account the ultrasonic attenuation in water.

The calorimetric method was repeated for three different volume of water (200 ml, 300 ml, and 400 ml) in different electric dissipation power up to 32 Watt. The ultrasonic power coefficient was then determined from the gradient of the plot of ultrasonic power versus total electrical dissipation power. The ultrasonic power coefficient was found to be insignificantly affected by the water volume, as shown in Table-1.

Table-1. The coefficient ultrasonic power measured by using calorimetric method.

Volume	200 ml	300 ml	400 ml
k	0.277	0.279	0.280

Based on results, the ultrasonic power coefficient is equal to 0.279 ± 0.001, indicating approximate 28 % dissipation power has been transmitted to ultrasonic power. This transmission is found to be independent on the height of liquid as predicted by the ultrasound attenuation. This might be attributed to the absorption of ultrasonic power by the vibration of the liquid interfacial layer.

For the ultrasonic reactor setup, the ultrasonic transducer is mounted at the bottom of the vessel. By ignoring the ultrasonic energy absorption by liquid, the intensity of ultrasonic irradiation can be calculated through Equation (4):

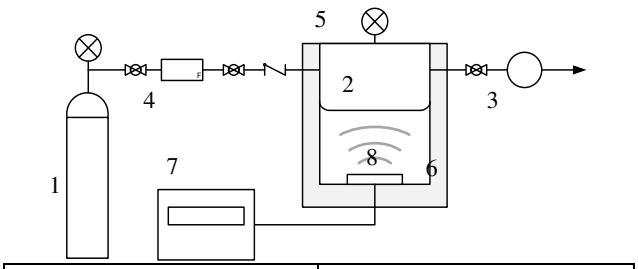
$$I = \frac{P_{us}}{A} \tag{4}$$

where A is the effective area of ultrasonic waves propagation, which also referred as the ultrasonic transducer disc area (approximately 154 mm^2 in this case).

METHODOLOGY

Gas-liquid mass transfer experiment setup

The experiment was set up based on dynamic pressure-step method in determining the mass transfer coefficient for CO_2 desorption. The schematic diagram of the experiment setup is shown in Figure-1. A CO_2 gas tank was connected to the vessel which was installed with 1.7 MHz ultrasonic piezoelectric transducer. The frequency of the transducer was controlled by an ultrasonic power system. Then, the ultrasonic reactor was located in a water bath with the temperature maintained at 30°C. The leakage test was conducted prior to the testing to ensure the accuracy of the experiment.



CO_2 gas tank	Pressure sensor
Ultrasonic reactor	Water bath
Air pressure regulator	Ultrasonic power system
Mass flow controller	Ultrasonic piezoelectric transducer

Figure-1. Experiment setup for the ultrasonic mass transfer study.

The vessel was filled with 100 ml distilled water, which acted as the physical solvent and tightly closed under 1 bar. Before the desorption process, the CO_2 is required to be absorbed into water solvent by increasing the pressure. Therefore, the pure CO_2 gas was compressed into the vessel with the flow rate of 10 SLPM, controlled by a mass flow controller. The pressure of vessel was governed by a back pressure regulator with absolute pressure of 11 bar. The absorption process of CO_2 was maintained until equilibrium. For the desorption process,

the valve was opened to reduce the pressure to 1 bar immediately. The time dependent pressure was manually recorded. The desorption process can be observed from the increasing of the pressure in vessel. The experimental procedures were then repeated for the experiment without ultrasonic waves as comparison.

Mass transfer coefficient determination

The enhancement of CO_2 mass transfer can be justified based on the mass transfer coefficient. The gas-liquid mass transfer coefficient was determined using a physical mass transfer method (Zieverink *et al.*, 2006). The resistance of the mass transfer was mainly located in the liquid layer. Therefore, the mass transfer coefficient can be obtained from the pressure drop. The mass transfer coefficient determination equation is expressed as in Equation (5):

$$\ln\left(\frac{P_m-P_f}{P(t)-P_f}\right)=\left(\frac{P_m-P_0}{P_f-P_0}\right)k_La(t-t_0) \quad (5)$$

where k_La is the mass transfer coefficient, P_f is final steady state pressure, P_m is the targeted pressure (absolute pressure of 11 bar in this case) and P_0 is the equilibrium pressure before expansion. By plotting Equation 5, the gradient of the plot, which is equal to k_La, can be obtained.

The mass transfer coefficient enhancement factor (E_c) is then determined by using Equation (6):

$$E_c=\frac{k_La^{us}}{k_La^0} \quad (6)$$

where k_La^{us} is the mass transfer coefficient with ultrasonic waves and k_La^0 is the mass transfer coefficient without ultrasonic waves.

Mass transfer rate determination

In addition, the ultrasonic enhancement in CO_2 absorption can be determined from the initial mass transfer rate. The initial mass transfer rate ($\dot{n}$) can be expressed as in Equation (7):

$$\dot{n}=\frac{V}{RT}\frac{dP}{dt}\bigg|_{t=0} \quad (7)$$

where V is the gas volume in the vessel and T is temperature. Meanwhile, the mass transfer rate enhancement factor (E_r) can be calculated by using Equation (8):

$$E_r=\frac{\dot{n}^{us}}{\dot{n}^0} \quad (8)$$

where $\dot{n}^{us}$ is mass transfer rate with ultrasonic waves and $\dot{n}^0$ is mass transfer rate without ultrasonic waves.

The difference between mass transfer rate enhancement and mass transfer coefficient enhancement is that, the mass transfer rate enhancement considered the increasing of initial rate of CO_2 amount transfer from water. Meanwhile, the mass transfer coefficient enhancement considered the decreasing of the time taken to reach the steady stage for the ultrasonic system.

RESULT AND DISCUSSIONS

Absorption

Before the desorption study, the absorption process was conducted to absorb the CO_2 gas into the water. The absorption can be observed from the pressure profile. Figure-2 shows the absorption pressure drop profiles in the gas-liquid mass transfer system. The pressure drops from 11 bar to 7.7 bar in 3 hours. There is insignificant pressure change observed after 3 hours. Therefore, P_0 is assumed to be 7.7 bar.

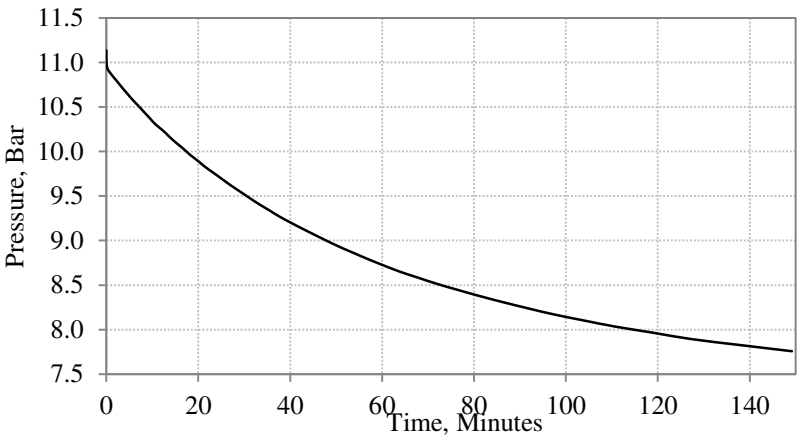


Figure-2. The time dependent pressure profiles during the absorption processes.

Desorption

For the desorption process, the CO_2 desorbed from the water increases the pressure in vessel. Figure-3 shows the CO_2 desorption pressure profiles in the gas-liquid mass transfer system with the presence of ultrasonic power. The results demonstrate that, the increase of pressure which indicated the desorption of CO_2, presents to be significantly faster than the case without ultrasonic power. Besides, it can be seen that, the final steady state pressure is higher with the presence of the total ultrasonic dissipation power. This means that, the CO_2 solubility is decreased under ultrasonic irradiation. This might be attributed to the ultrasonic pseudo-solubility and ultrasonic heating effect. The heat might be caused by the ultrasonic waves, which is generated by the ultrasonic transducer. Therefore, the temperature of the vessel is increased, and thus decreasing the CO_2 solubility in water.

In relation to this, Figure-4 is plotted based on Equation (5) for the mass transfer coefficient determination with presence of ultrasonic power. The mass transfer coefficient can be obtained from the gradient of the graph, which are 0.001 s^{-1} and 0.021 s^{-1} for the case without ultrasonic irradiation and with ultrasonic irradiation, respectively.

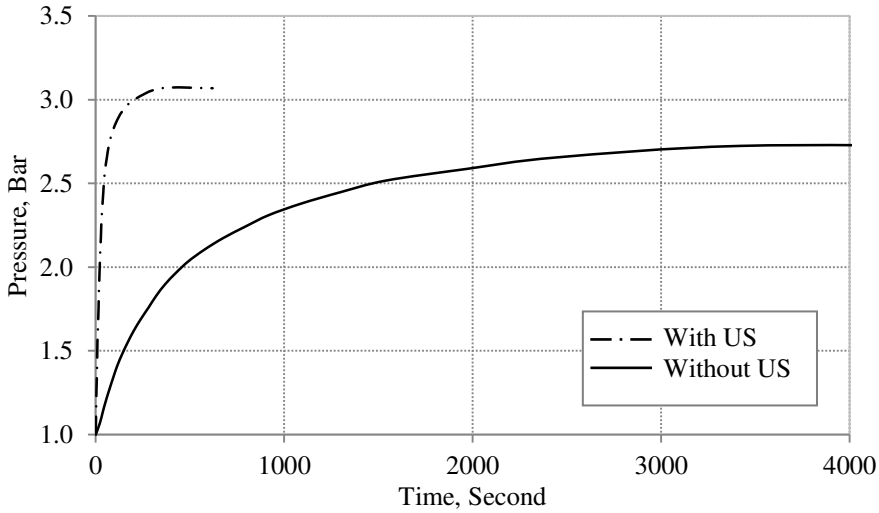


Figure-3. The time dependent pressure profiles during CO_2 desorption processes with the presence of ultrasonic power.

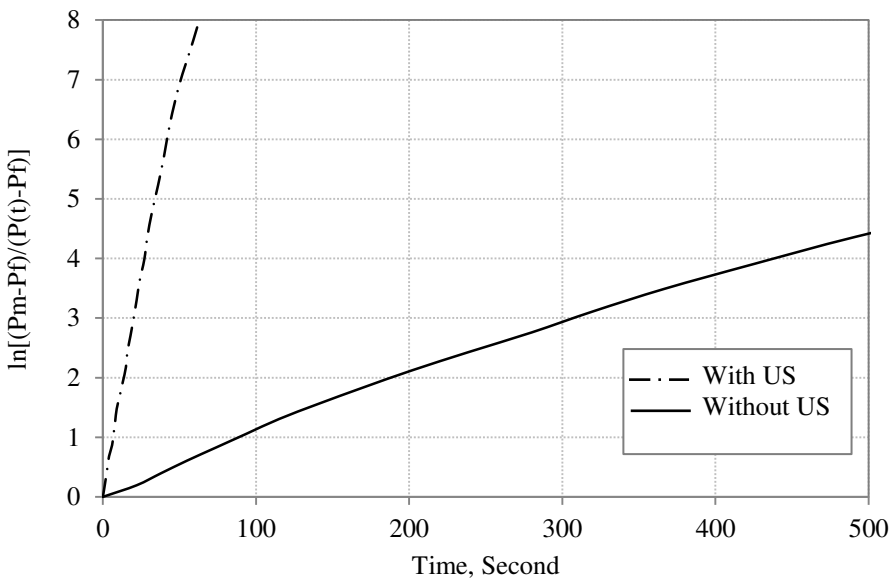


Figure-4. Plots of mass transfer coefficient determination for CO_2 desorption process with the presence of ultrasonic power.

Figure-5 shows the results of the mass transfer rate and the coefficient of enhancement with the presence of ultrasonic power. It is found that, the desorption mass transfer coefficient enhancement factor, which was determined from the time required to achieve steady state using Equation 5 and 6, is up to ~20 times. Meanwhile, the mass transfer rate enhancement factor, which was determined using Equation (7) and (8), is consistent with the desorption mass transfer coefficient result. The experiment enhancement is found to be mainly depended on the ultrasonic streaming effect. The cavitation effects are still unclear for the ultrasonic enhancement.

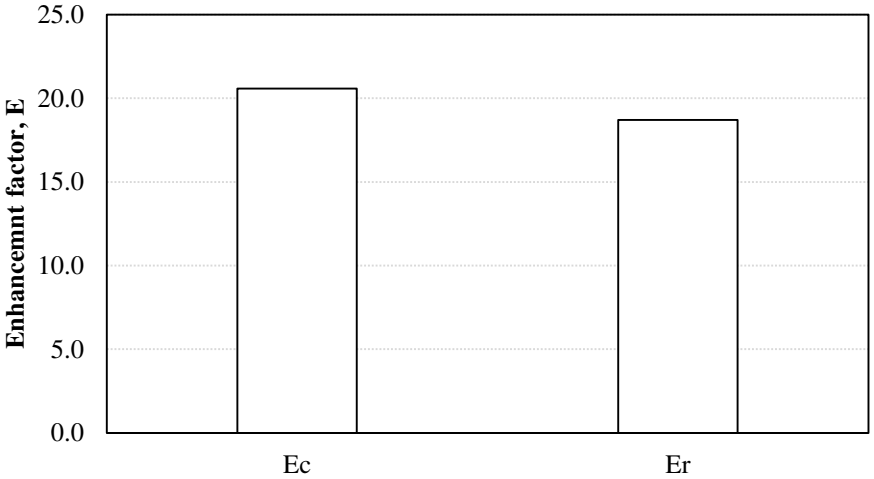


Figure-5. The comparison of mass transfer coefficient enhancement, *Ec* and mass transfer rate enhancement, *Er* for CO_2 desorption with the presence of ultrasonic power.

CONCLUSIONS

In conclusion, the ultrasonic irradiation with high frequency has significantly enhanced the desorption mass transfer coefficient up 20 times faster than the case without ultrasonic irradiation. The total electrical dissipation power is equal to 32 Watt. Meanwhile, the ultrasonic power is equal to 9 Watt. The power efficiency is approximate to 28 %. Besides, the gas solubility is found to be slightly decreased under the ultrasonic waves. However, further investigation is still required to understand the mechanism of the ultrasonic enhancement.

ACKNOWLEDGEMENT

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Nomenclature

P_{us}	Total ultrasonic dissipation power	W
V_{rms}	Roof mean square of supplied high frequency alternative voltage	V
R	Efficient resistance of ultrasonic transducer	Ω
I	Intensity of ultrasonic	W m^{-2}
A	Effective area of ultrasonic waves propagation	m^2
k	Coefficient ultrasonic power	-
$k_L a$	Mass transfer coefficient	s^{-1}
P_f	Final steady state pressure	Pa
P_m	Targeted pressure	Pa
P_0	Equilibrium pressure before compression	Pa
t	Time	s
E_c	Mass transfer coefficient enhancement factor	-
$\dot{n}$	Initial mass transfer rate	kg s^{-1}
E_r	Mass transfer rate enhancement	-
P	Pressure	Pa
V_G	Gas volume in vessel	m^3
R	Ideal gas constant	J mol^{-1} k^{-1}
T	Temperature	K
C_L	Heat capacity	J kg^{-1} k^{-1}
V_L	Liquid volume	m^3
ρ_L	Liquid mass density	kg m^{-3}
P_T	Total electrical dissipation power	W
t_o	Initial time	s
us	Ultrasonic	-