Jurnal
Teknologi

Full Paper

HYDRODYNAMIC CAVITATION USING DOUBLE ORIFICE-PLATES FOR THE GENERATION OF HYDROXYL RADICALS

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Article history
Received
18 January 2016
Received in revised form
14 July 2016
Accepted
18 October 2016

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Graphical abstract

Abstract

The generation of hydroxyl radicals ($OH^{\bullet}$) by hydrodynamic cavitation (HC) using single and double orifice plates was studied. Five orifice plates with different configurations (size and number of orifice, total orifice area) were tested. The formation of $OH^{\bullet}$ was measured by iodide dosimeter method using spectrophotometer at 355 nm wavelength. The effects of plate configurations and double plate arrangements on $OH^{\bullet}$ generation were investigated in 60 minutes of reaction time using an inlet pressure of 45 psi and initial potassium iodide (KI) concentration of 20 g/L. The generation of $OH^{\bullet}$ were expressed in terms of concentration and percentage of increase of iodine liberation. The liberated iodine for single plate ranged from 0.26 to 0.56 g/L (84 to 180% increase). The highest liberation was achieved using plate with the lowest total flow area of orifice, which had the smallest cavitation number. The double plate arrangement produced the highest iodine liberation (1.30 g/L; 420% increase) with the highest cavitational yield (2.9 x 10^{-1} mg/J) as compared to those of single plate arrangement. In double plate arrangement, the enhancement was dependent on the configuration and arrangement of the plates.

Keywords: Hydroxyl radical, hydrodynamic cavitation, dual orifice plates, single orifice plate, advanced oxidation processes, iodide dosimeter

Abstrak

Hidroksil radikal ($OH^{\bullet}$) yang dihasilkan oleh *hydrodynamic cavitation* menggunakan satu plat dan dua plat berlubang telah dikaji. Lima plat berlubang dengan pelbagai konfigurasi (saiz dan bilangan lubang, jumlah keluasan lubang) telah diuji. Pembentukan $OH^{\bullet}$ diukur dengan kaedah *iodide dosimeter* menggunakan *spektrofotometer* pada panjang gelombang 355 nm. Kesan konfigurasi plat dan susunan dua plat kepada pembentukan $OH^{\bullet}$ dikaji dalam masa tindak balas selama 60 minit pada tekanan inlet 45 psi dan kepekatan awal kalium iodida (KI) 20 g/L. Pembentukan $OH^{\bullet}$ dinyatakan dalam bentuk kepekatan dan peratus peningkatan pembentukan iodin. Pembentukan iodin untuk satu plat adalah dalam julat 0.26 hingga 0.56 g/L (peningkatan 84 hingga 180%). Pembentukan tertinggi dicapai dengan menggunakan plat yang mempunyai keluasan lubang aliran terendah dengan *cavitation number* yang paling kecil. Penggunaan susunan dua plat menghasilkan iodine tertinggi (1.30 g/L; peningkatan 420%) dengan *cavitational yield* (2.9 x 10^{-1} mg/J) berbanding dengan penggunaan satu plat. Bagi susunan dua plat, peningkatan pembentukan iodin bergantung kepada konfigurasi plat dan cara susunan antara dua plat.

Kata kunci: Radikal hidroksil, *hydrodynamic cavitation,* dua plat berlubang, plat berlubang tunggal, proses pengoksidaan lanjutan, *iodide dosimeter*

1.0 INTRODUCTION

Hydroxyl radicals (HO•) are the prominent oxidizing agent generated in advanced oxidation processes (AOPs). With oxidation potential second to fluorine, the radicals have been used as the oxidant to destroy pollutants unselectively with reaction rate constants ranging from 10^6 to 10^9 $M^{-1}•s^{-1}$ [1]. Different technologies and systems to generate HO• have been studied, developed and applied [2, 3, 4]. These include ozone-based systems, photo-based systems, hydrogen peroxide based systems, and cavitation systems. As compared to other systems, hydrodynamic cavitation (HC) is getting more attention recently due to its operation simplicity and lower cost [5].

One of the techniques to measure the presence of HO• is through the chemical method known as iodide dosimeter. In this method, iodine is formed via the oxidation of iodide ions by the HO• as shown in Eqns. (1) to (3). Hence, the concentration of HO• is indicated by the concentration of liberated iodine measured in the solution; higher iodine liberated relates to higher concentration of HO• in the solution. Several studies have used iodide dosimetry for the measurement of HO• [6, 7] and this method has also been validated against terephthalic acid dosimetry by Ebrahiminia *et al.* [8].

$$HO^{\bullet} + I^- \rightarrow OH^- + I \quad (1)$$
$$I + I^- \rightarrow I_2^- \quad (2)$$
$$2I_2^- \rightarrow I_2 \; 2I^- \quad (3)$$

Hydrodynamic cavitation has been studied for different applications including cell disruption [9], hydrolysis of canola oil [10], wastewater treatment [11, 12], water disinfection [2, 13, 14], oxidation of alkylarenes [15, 16], synthesis of biodiesel [17, 18, 19], and nanoemulsions formation [20, 21, 22]. Additionally, few studies have been conducted to enhance the capacity of HC in generating the radicals. Wang *et al.* [23] and Ghayal *et al.* [24] have studied the performance of multiple orifice plates in a single plate to generate the radicals. Additional constriction of venturi within multi-hole orifice plates have been studied to extent the degradation of Rhodamine B by Mishra and Gogate [25]. Chakinala *et al.* [26] have used chloroalkanes as additives in improving the efficacy of HC. Similarly, Ambulgekar *et al.* [16] and Wu *et al.* [27] investigated the effect of aqueous potassium permanganate and hydrogen peroxide, respectively in enhancing the performance of cavitation. Despite these efforts, more alternative approaches are needed to enhance the radicals formation in the HC system.

The present study evaluated the use of double orifice plates that are arranged in series to enhance the radicals generation in HC. In a typical single plate arrangement, the cavities are developed and collapsed as the liquid passes through the orifice under high inlet pressure. This causes the formation of very high energy density, local temperature and local pressure at the surface of the cavities resulting in the formation of HO•process. With the residual inlet pressure exerted to the liquid, the same phenomena is expected to take place simply by adding a second plate in series into the cavitation chamber, causing more radicals to be formed. Based on our literature review, this technique has never been tested previously. Therefore, using iodide dosimeter, the study investigated the effects of double orifice plates in enhancing the HO• formation under different plate arrangements.

2.0 METHODOLOGY

2.1 Materials and Methods

Potassium iodide (KI) used was of analytical grade (Sigma-Aldrich). The stainless steel orifice plates with five different configurations were designed based on Vichare *et al.* [28]. The plates are shown in Figure 1 and the detail physical characteristics are given in Table 1. The effective plate diameter is 20 mm.

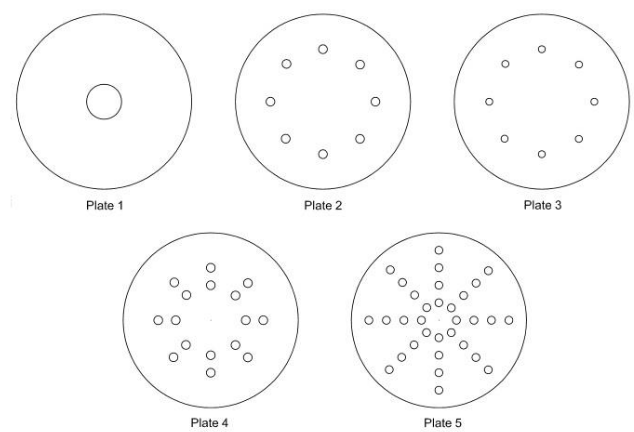

Figure 1 Types of plate and arrangement of holes on the plates

Table 1 Detail specifications of plates used in the study

[a]Plate number	Number of orifice	Diameter of orifice (d_h) (mm)	Total area of orifices (mm^2)	Total perimeter of orifices (mm)
Plate 1 (P1)	1	4.0	12.6	12.6
Plate 2 (P2)	8	1.0	6.3	25.1
Plate 3 (P3)	8	0.8	4.0	20.1
Plate 4 (P4)	16	1.0	12.6	50.3
Plate 5 (P5)	32	0.9	20.4	90.5

[a]Effective plate diameter is 20 mm

2.2 Analytical Method

The concentrations of liberated iodine were determined using HACH spectrophotometer (HACH DR5000) at the wavelength of 355 nm [8]. The absorbance values were then referred to the calibration curve of iodine-absorbance (Absorbance = 812.19 x Concentration + 0.009; R^2 = 0.955).

The extent of iodine liberation was calculated as in Eqn. (4),

Extent of iodine liberation (%) = (B-A) / A x 100 (4)

where A is the initial value of iodine and B is the value of iodine at time t.

2.3 Hydrodynamic Cavitation Reactor

The experimental setup of the HC reactor (Figure 2) was based on Vichare *et al.* [8]. The reactor consists of a horizontal multistage pump (Shanghai East Pump Co., Ltd, China), a cavitation chamber with three flanges provided for the orifice plates and 20 mm inside diameter of hard glass tube (made of acrylic in between flanges for visual observation), control valves (V1, V2, V3, V4) and pressure gauges (P1, P2). The piping system of 20 mm inside diameter was divided into two lines, which were the main line and the by-pass line. The main line was where the cavitation chamber was located for the cavitation to take place. The by-pass line was used for flow control and safety measures of the system. Both discharge points of the main line and the by-pass line ended in the 10 L holding tank. A propeller (30W, WiseStir, HS-30D; Daihan Sci.) was installed in the holding tank to mix the water in the tank with the incoming treated water.

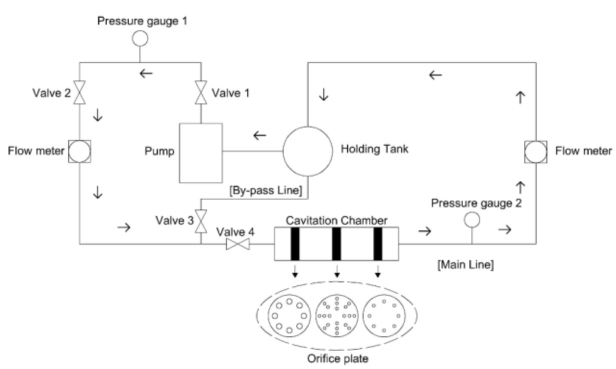

Figure 2 Schematic diagram of hydrodynamic cavitation reactor setup

2.4 Experimental Procedures

The preliminary experiments were conducted to characterize the effect of plate configurations on iodine liberation. These were followed by applying double plates in the cavitation chamber. 10 L of KI solution (20 g/L) was prepared in distilled water [26] for a typical experimental run. An experimental run started with filling the holding tank with KI solution. The solution was mixed using the propeller and an aliquot of sample was then taken out from the holding tank at t = 0 min. Then, under the specified inlet pressure of 45 psi, the solution was pumped from the holding tank to the cavitation chamber in the main line and returned back into the holding tank. During the experimental runs, all valves, except for Valve 3, were opened. Samples were collected from the holding tank at specific intervals. The pressure and flow rate of the solution in the system were measured using the pressure gauge and flow meter, respectively.

3.0 RESULTS AND DISCUSSION

3.1 Preliminary Study

The flow and characteristics of the plates are shown in Table 2. Besides the number and the diameter of the orifice, the plates are characterized according to variable α as given in Eqn. (5) [28]. The characteristics of cavitation in the HC chamber is measured based on cavitation number (C_v) according to Eqn. (6) [29].

α = Total perimeter of orifice / Total area of orifice

$$= \frac{n \times 2\pi(d_h/2)}{n \times \pi(d_h/2)^2} = \frac{4}{d_h} \quad (5)$$

$$C_v = \frac{P_2 - P_v}{0.5\rho v^2} \quad (6)$$

where n is the number of orifice and d_h is the diameter of orifice, P_2 is the recovered downstream pressure, P_v is the vapour pressure of liquid and *v* is the liquid velocity at the orifice, which is determined from the flow rate and area of the orifice.

Table 2 Flow characteristics of single-plate

Plate	Flow rate (L/min)	Orifice velocity (m/s)	Cavitation number (C_v)	α (mm^{-1})
P1	7.6	10.1	4.25	1
P2	4.9	12.9	1.15	4
P3	3.5	14.6	0.55	5
P4	7.7	7.3	6.80	4
P5	8.3	6.8	11.46	4.4

At an inlet pressure of 45 psi, the flow rate passing through the plates ranged from 3.5 to 8.3 L/min. The orifice velocity ranged from 6.8 to 14.6 m/s, while the C_v ranged from 0.5 to 11.5. The profile of iodine liberation for the plates with respect to time is illustrated in Figure 3. Within 60 minutes of reaction, the iodine liberation ranged from 0.26 to 0.56 g/L, which represents 84.2 to 179.4% increase in iodine concentration.

Of the five plates tested, P3 generated the highest iodine, followed by P5, P4, P2 and P1. The one without the orifice plate generated the least amount of iodine (10.6%), representing the liberation of iodine that could be due to other factors within the system. Plate P3 has the smallest total area of orifice and C_v and the highest value of α. These optimum conditions lead to higher number of cavities and increased turbulence pressure fluctuating frequency leading to more efficient collapse of cavities resulting to more $HO^•$ generation [30]. On the contrary, P1 that has the largest hole size and smallest α value is likely to have a much lower turbulence frequency than the natural oscillation frequency of the generated cavities resulting in much lower cavity collapse intensity. As for other plates, the iodine liberation is affected by the combination of the size and the number of orifice on the plate. Plate P5, for example, despite having the highest C_v, its iodine liberation is second after P3. This is anticipated to be caused by the high number of orifice, which provide more sites for cavity generation.

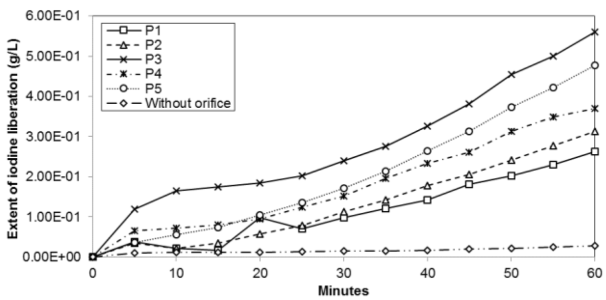

Figure 3 Extent of iodine liberation using different configurations

3.2 Double-plate Arrangement

3.2.1 Effect on Iodine Liberation

The effect of orifice plates on the extent of iodine liberation was further explored with double orifice plates by arranging them in a single line. Two plate arrangements were initially studied, namely P3 followed by P2 (P3P2) and vice versa (P2P3). P3 was chosen due its high iodine liberation in the preliminary study, while P2 was chosen due to its low total area of orifice. The first plate is designated as primary plate, while the following plate as secondary plate.

The results of iodine liberation with double orifice plates for 60 minutes at 45 psi inlet pressure are given in Figure 4. The highest iodine liberation was produced by P3P2 plate arrangement, followed by P3, P2P3 and P2. The iodine liberation by P3P2 was 1.30 g/L, which represents an increase of 420%. It can be observed that the presence of secondary plate enhanced the liberation of iodine, which indicates the additional generation of $HO^•$ possibly based on the same cavitation mechanisms as discussed earlier. The extent of iodine liberation for double plates that was higher than the single plate could be due to the occurrence of back pressure created by the secondary plate. This reduced the velocity of the fluid, which provides more time for the cavities to grow larger before collapsing, similar to the phenomenon postulated in the venturi system [27]. This increased the collapse intensity and thus increased the formation of $OH^•$. However, further study is needed to have a better understanding of this phenomenon.

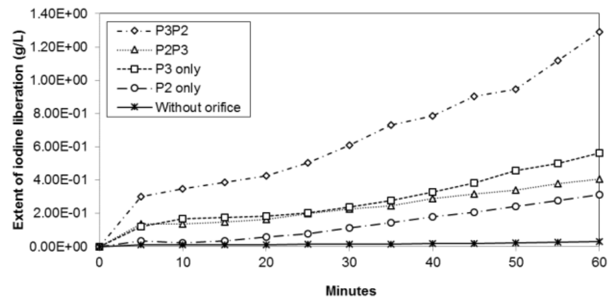

Figure 4 Iodine liberation with different coordination of orifice plates

The arrangement of plates was also observed to influence the extent of iodine liberation. The extent of iodine liberation was higher for the combination of P3P2 as compared to P2P3. The presence of P2 after P3 added more cavitation activities in the system. On the other hand, although the presence of P3 after P2 also provided additional cavitation activities to the system, the cavities produced by P3 following P2 were not as much as when P3 was arranged as the primary plate.

The flow characteristics of the plates at different arrangements are given in Table 3. Due to the double plate arrangement, the flow rate, the primary plate orifice velocity and the C_v were reduced due to additional constriction. It can be clearly observed that P3P2, which liberated the highest iodine has the lowest C_v and it was vice versa for P2. As for P2P3 and P3, the iodine liberation for both were about the same up to 30 minutes of reaction, following which, more iodine were liberated by P3, despite having higher C_v value as compared to P2P3. The addition of P3 after P2 significantly enhanced the iodine liberation as compared to P2 alone. However, there may be some other factors affecting this process (other than the geometry of cavitation device) to enhance the performance of P3 as compared to P2P3.

Further study was conducted to investigate the effect of having other plates as secondary plate following P3. Two further combinations, namely P3P4 and P3P5 were studied at an inlet pressure of 45 psi for 60 minutes. The flow characteristics of the combinations and the results are shown in Table 4 and Figure 5, respectively.

As shown in Figure 5, the pair of P3P2 liberated the highest iodine, followed by P3P4 and P3P5. These pairs have about the same C_v value (i.e. 0.13) and the second plate has about the same orifice diameter; the only difference is the number of orifice of the second plate which resulted in different total area of orifice with P2 having the smallest, followed by P4 and P5. As expected, the second plate with lower total orifice area gave higher velocity. This provided higher cavity activities, and thus generated more radicals to liberate the iodine.

Table 3 Flow characteristics for different plate arrangement

Plate	Flow rate (lit/min)	+Orifice velocity (m/s)	+Cavitation number (Cv)
P2	4.9	12.9	1.15
P3	3.5	14.6	0.55
P2P3	2.8	7.5	0.32
P3P2	2.6	10.6	0.13

+For double plates, calculations were made based on primary plate

Table 4 Flow characteristics of double-plate

Plate	Flow rate (lit/min)	+Orifice velocity (m/s)	+Cavitation number (Cv)
P3	3.5	14.6	0.55
P3P2	2.6	10.6	0.13
P3P4	3.4	14.0	0.13
P3P5	3.4	14.2	0.13

+For double plates, calculations were made based on primary plate

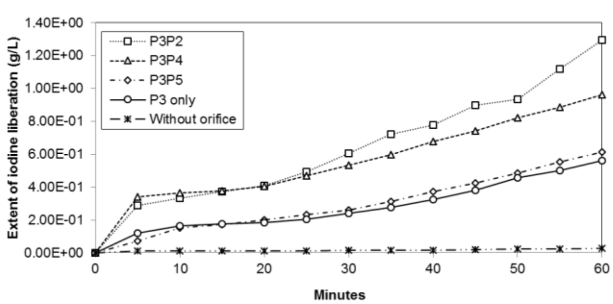

Figure 5 Profile of iodine liberation for five different combination of plates

3.2.2 *Effect on Cavitational Yield*

The formation of OH• generated by HC was also analysed based on cavitational yield; the basic term that is used for measuring cavitation on the basis of total energy consumption and energy efficiency of the system. The cavitational yield is defined as the extent of iodine liberation per unit energy consumed (mg/J) according to Eqn. (7) [31].

$$\text{Cavitational yield (mg/J)} = \frac{\text{Amount of iodine liberation}}{H\rho gQt} \quad (7)$$

where H is pressure head of the flowing liquid, Q is the flow rate in the main line and t is the time of operation. Considering the rating of the pump used in the present work as 1.2 kW, the total power dissipation per unit volume was calculated as 120 W/L.

Figure 6 shows the cavitational yield for single and double plates that were tested in this study. The cavitational yield ranged between 1.9 x 10^{-2} and about 2.9 x 10^{-1} mg/J. The highest cavitational yield was achieved by P3P2, while the lowest was by P1. The cavitational yield observed in the present study is consistent with the results reported in the literature [31]. It can be clearly seen that the yield of cavitation or the energy efficiency of the system was enhanced by the combination of plates.

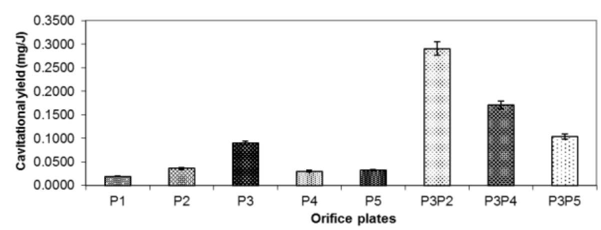

Figure 6 Variation of cavitational yield for different plate configurations and plate arrangement

4.0 CONCLUSION

The effects of plate configuration and double plate arrangement on HO• generation using iodine liberation as indicator were explored. It was observed that the plate with smaller total orifice area generated more radicals due to the high velocity of water passing through the orifices and hence, generated higher cavitational intensity. For plates with the same total orifice area, more iodine were generated by the plate with the more number of orifice, or plate with smaller orifice diameter. For single orifice plate, the plate with the lowest total orifice area (4 mm^2) was found to liberate the highest iodine concentration (180%) after 60-minute of reaction. However, more iodine was generated when double orifice plates were arranged in series. Based on double-plate arrangements, the enhancement was observed to be dependent on the configuration and the arrangement of the plates. Combination of plates with the smallest total flow area liberated the highest amount of iodine. The arrangement of plate was also important as certain arrangement may provide resistance to the flow, hence may reduce the cavitational intensity. The double plate arrangement of P3P2 was found to be the best plate arrangement liberating iodine up to about 420%. The same arrangement also generated the highest cavitational yield as compared to other arrangements.

Acknowledgement

The authors would like to acknowledge Universiti Teknologi Malaysia and Ministry of Education, Malaysia for supporting the study under Research University Grant Scheme (Vot No. Q.J130000.2522.04H81).

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