OC-41: Opposing Effects of Flow on Sonochemical Activity in a High Frequency Ultrasonic Reactor

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1. Introduction

Previous literature reports have shown conflicting effects of flow on sonochemical activity of ultrasound-induced chemical reactions at high frequencies. Both an increase and a decrease in sonochemical activity due to the introduction of flow into the reacting solution have been reported. For example, the sonochemical degradation of pentachloraphenol was decreased by the introduction of continuous flow at 500 kHz over a power range of 12 to 45 W (Gondrexon et al., 1999) and at 133.2 kHz potassium iodide oxidation was most efficient with no flow in the rectangular-channel type reactor compared to that with flow across the faces of the 14 transducers in series (Matsuoka et al., 2007). However a study at 490 kHz found that an overhead stirrer increased the sonochemical efficiency over an input electric power range of 5 to 50 W (Kojima et al., 2010). This introduces opposing effects of flow in different reactors, however it is difficult to compare the effects of flow in these cases as the flow types and the reactor geometries are all varied. Reactor geometries are known to influence the sonochemical activity (Asakura et al., 2008, De La Rochebrochard et al., 2012, Nanzai et al., 2009) and the type of flow may also influence the effect that it has.

In this work we examine the effect of two different flow types; overhead stirring and upward flow circulated through the reactor in the direction of the propagation of the ultrasonic wave in a high frequency sonochemical reactor. The reaction solution heights were kept constant with a batch and continuous reactor style for the overhead stirring and upward flow experiments respectively. The effect of flow after 30 minutes of sonication was measured through sonochemiluminescence (SCL) imaging and the total yield of hydrogen peroxide using iodide dosimetry in a 995 kHz reactor at two different power settings; A: 5.2×10^{-2} W/cm^2 and B: 7.2×10^{-2} W/cm^2.

2. Results and Discussion

We found that the upward flow was able in some cases to increase the yield of hydrogen peroxide and that an overhead stirrer tended to decrease the hydrogen peroxide yield. These findings demonstrate that we need to consider the physical effect of each type of flow on the ultrasonic field and subsequent bubble population. At high frequencies the small, inactive bubbles are mostly produced (Leighton, 1994) which rely on growth through coalescence to increase the active bubble population and subsequently the yield of hydrogen peroxide (Yasui, 2002). Also at high frequencies travelling waves are thought to be dominant rather than standing waves (Leighton, 1994) and so the main force of coalescence is from secondary Bjerknes forces. The travelling wave structure was evident in the SCL imagery with a fountain being observed at the surface of the solution. The SCL images indicate that the active area is influenced mostly by the introduction of the overhead stirring and that the upward flow does not have a notable effect on the active bubble area. Hence in our reactor we have a travelling wave dominant ultrasonic field which relies on growth of small inactive bubbles through coalescence from secondary Bjerknes forces to active bubble size.

The effect of the overhead stirrer follows the theory and its predictions made previously based on reduced coalescence of bubbles. Hatanaka et al predicted that at high frequencies, the introduction of a flow from an overhead stirrer would reduce the coalescence necessary for small inactive bubbles to grow into the active bubble size range (Hatanaka et al., 2006). At 7.2×10^{-2} W/cm^2 no significant difference in hydrogen peroxide yield was observed upon the introduction of overhead stirring although the trend was for the average yield to decrease with an increase in stirring speed. However at power 5.2×10^{-2} W/cm^2 the yield was decreased with an increase in stirring speed (Figure 1). At the lower power setting in this study the decreasing trend in yield is especially evident where at 600 rpm the hydrogen peroxide yield has decreased from the initial experiment without stirring and at 900 rpm the yield was below the detection limits of the spectrophotometer. At the higher power setting the variation in the results makes the difference between the speeds of circulation indistinguishable, although the SCL images show a lowering of the active bubble area with the introduction of overhead circulation at higher speeds. The difference in the results between the two power settings is most likely to be due to the overhead circulation not being as effective on the ultrasonic field at the higher power setting.

The ability of the upward flow at 34 ml/min through the reactor to increase the hydrogen peroxide yield is predicted to be due to increased coalescence of small inactive bubbles due to an increased proportion of travelling wave in the ultrasonic field. At 7.2 × 10^{-2} W/cm^2 there is no determinable difference in hydrogen peroxide yield between 0 and 414 ml/min whereas the flow at 34 ml/min is able to increase the yield. At 5.2 × 10^{-2} W/cm^2 the faster flow speed decreases the hydrogen peroxide yield compared to the slower speed and still experiments (Figure 1). Since the flow is in the direction of the propagation of the ultrasonic wave the solution is able to travel through without too much disturbance of the ultrasonic field. At the higher flow rate an increase in hydrogen peroxide yield at the higher power is not as evident and at the lower power we see a decrease in hydrogen peroxide yield. This is believed to be due to less coalescence of small inactive bubbles due to a disturbance of the ultrasonic wave field. At the lower power setting there is no difference in the hydrogen peroxide yields between the still experiments and the experiments with 34 ml/min, most likely because there is some disturbance and some support of the travelling wave structure in the ultrasonic field. At high frequencies the dominant ultrasonic field type is the travelling wave and this is the main source of sonochemical activity. We believe that the reason for the increase in hydrogen peroxide yield is because the travelling wave was able to be enhanced without too much disturbance of the ultrasonic field to decrease coalescence at the slower flow rate.

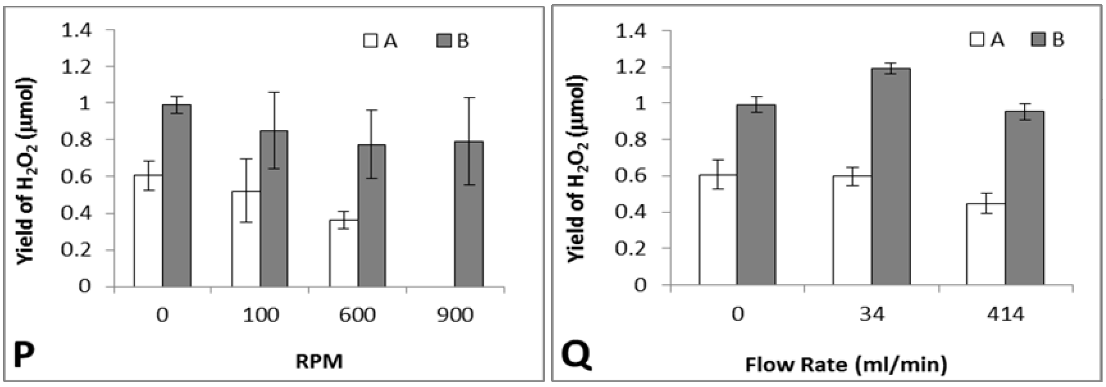

Figure 1:Hydrogen peroxide yield for the two flow types; overhead stirring (P) and upward circulation (Q). The open and shaded bars correspond to power setting A; 5.2 × 10^{-2} W/cm^2 and B; 7.2 × 10^{-2} W/cm^2 respectively.

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