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ARTICLE *in* ULTRASONICS SONOCHEMISTRY · NOVEMBER 2003

Impact Factor: 4.32 · DOI: 10.1016/S1350-4177(03)00104-4 · Source: PubMed

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Ultrasonics Sonochemistry 10 (2003) 331–335
Ultrasonics SONOCHEMISTRY
www.elsevier.com/locate/ultsonch

Mapping of an ultrasonic horn: link primary and secondary effects of ultrasound ☆

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Received 17 March 2003; accepted 17 March 2003

Abstract

The erratic behaviour of cavitational activity exhibited in a sonochemical reactor pose a serious problem in the efficient design and scale-up; thus it becomes important to identify the active and passive zones existing in the reactor so as to enable proper placement of the reaction mixtures for achieving maximum benefits. In the present work mapping of ultrasonic horn has been carried with the help of local pressure measurement using a hydrophone and estimation of amount of liberated iodine using the Weissler reaction and a quantitative relationship has been established. The measured local pressure pulses have been used in the theoretical simulations of the bubble dynamics equations to check the type of cavitation taking place locally and also estimate the possible collapse pressure pulse in terms of maximum bubble size reached during the cavitation phenomena. Relationship has been also established between the observed iodine liberation rates and the maximum bubble size reached. The engineers can easily use these unique relationships in efficient design, as the direct quantification of the secondary effect is possible.

Keywords: Sonochemical reactors; Mapping; Local pressure; Cavitational yield; KI oxidation

1. Introduction

Cavitation results in generation of very high pressures (1000–50,000 bars) and temperatures (in the range of 1000–5000 K) and these effects are observed at millions of locations in the reactor. There are large illustrations where these spectacular effects have been successfully harnessed for a variety of applications worldwide. Few of the important applications can be given as chemical synthesis (in both homogenous and heterogeneous systems by way of increase in the rate and selectivity of many chemical reactions), wastewater treatment (degradation of many of the biorefractory/complex chemicals), textile processing, biotechnology (cell disruption and foam control in bioreactors), crystallization etc. However it should be noted that, inspite of extensive research and vital potential applications, there is hardly any chemical processing carried out on an industrial scale owing to the lack of expertise required in diverse fields such as material science, acoustics, chemical engineering etc. for scaling up successful lab scale processes and also due to the lack of suitable reactor design and scale-up strategies.

The major problems identified in designing large scale reactors are local existence of cavitation events very near the irradiating surface, wide variation of the energy dissipation rates in the bulk volume of the reactor and the erosion of the sonicator surfaces at the high power intensities required for industrial scale operations. In some of the earlier works [1,2], we have tried to address the first problem by using a new geometric arrangement of the transducers on the irradiating surface. The second problem is mainly attributed to the current state of the transducer technology and the attenuation of the acoustic field by cavitation events at and near the sonicator surface. It is of paramount importance to understand the effect of these changes in the cavitational

☆ This paper was originally presented at Applications of Power Ultrasound in Physical and Chemical Processing (Usound3) Paris, December 2001.

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1350-4177/$ - see front matter
doi:10.1016/S1350-4177(03)00104-4

activity, which results in active and passive zones in the reactor. Hence it becomes necessary to characterize the cavitational activity and to study the spatial variation of the same. The third major problem identified in the efficient scale-up of the ultrasound-based reactors is more dependent on the available knowledge base from the field of material science. Aim should be at developing new transducers with material of construction giving minimal erosion rates at higher power dissipation levels and at the same time with similar levels of energy transfer for cavitation events (similar acoustic properties).

In the present work, the variation in the cavitation intensity has been studied in a conventional immersion type reactor i.e. ultrasonic horn. Attention has been focused on local measurements of the pressure amplitudes using hydrophone, measurement of chemical effect of cavitation i.e. extent of decomposition of aqueous KI solution and relating the observed cavitational yields with the measured pressure fields and also with the theoretical predictions of the bubble dynamics equations.

2. Experimental

Ultrasonic horn used in the present work (Dakshin Ltd. Mumbai, India, maximum power rating of 240 W and driving frequency of 20 kHz) has a tip diameter of 2 cm and was always dipped to 2 cm below the liquid level. The experiments were done in a beaker of 2000 ml capacity (diameter = 13.5 cm, height = 17.5 cm) completely filled with distilled water.

The local pressure amplitudes were measured using a hydrophone (Bruel & Kjaer Ltd. Type 8103, Denmark) and a charge amplifier. The dimensions of the hydrophone used were; length of 50 mm and diameter of 9.5 mm. The signal from the hydrophone was amplified through a charge amplifier and fed to a fast Fourier transform (FFT) based spectrum analyzer (Lecroy Ltd. Model 9310 M, USA). The resultant output was recorded in terms of mV for the corresponding frequencies. From the FFT, it was observed that peaks were obtained at the driving frequency and multiples of the driving frequency (i.e. at 40, 60 and 80 kHz). Using the hydrophone calibration details supplied by the manufacturer, the amplitudes of the pressures were calculated at these peaks. As cavitation is a very random phenomenon, two consecutive readings at the same conditions may yield completely different results in terms of the measured pressure amplitudes. Therefore 60 pressure pulses were collected for a particular frequency at a particular location. The sixty readings ensured that statistically significant number of events have been measured which reduces the random variation in the measured pressure pulse due to the occasional and random cavity collapse. The measurements were made in axial direction with increasing distance from the tip of the horn and also some measurements were made in the plane of the horn tip in the radial direction.

The model reaction considered for the quantification of secondary effects is the decomposition of potassium iodide liberating iodine. The details about the chemical reaction and quantification of liberated iodine have been already discussed earlier [1].

The measured pressure pulse at the driving frequency has been used in the bubble dynamics equations for checking the type of collapse as well as the maximum cavity radius reached during the growth phase. Simulations were carried out to find out how the variation in the local pressure amplitude affects the bubble dynamics. The measured values of pressure amplitude were substituted for P_A in the equation for local fluctuating component of pressure given below:

$$P_\infty = P_0 - P_A[\sin(2\pi f t)] \quad (1)$$

where, f is the frequency of irradiation (20 kHz) and P_0 is the initial pressure inside the bubble.

Rigorous and more realistic equation [3] considering the compressible nature of the liquid medium has been used in the present case for numerical simulations. The details about the simulation conditions, the solution methodology and assumptions involved in these equations have been already described in the earlier work [4] and hence not repeated here. The simulations were performed for the frequency showing maximum amplitude in the frequency spectrum with an initial cavity size of 2 μm. For each value of the driving pressure, the maximum radius reached by the growing bubble during growth stage of the cavitation phenomena was obtained from the solution of the bubble dynamics equation. The maximum bubble radius gives an indication of the nature of cavitation i.e. stable or transient; hence the final pressure/temperature pulse that is likely to be generated during the cavity collapse resulting in the desired effects and the number of free radicals generated as well as the zone of cavitational influence on the surrounding liquid.

3. Results and discussion

3.1. Variation in the local intensity

The FFT response curves for two sample positions viz. at a distance of 1 cm from the tip of the horn (A) and at distance 4.5 cm away from the tip (G) have been shown in Fig. 1. It can be seen from the figure that peaks are observed at the driving frequency (20 kHz) and also at multiples of the driving frequency (i.e. 40, 60 and 80 kHz) as well as the sub-harmonic frequency. Moholkar et al. [5] have shown that these peaks are absent for the non-cavitating liquids. Non-cavitating liquids show only one

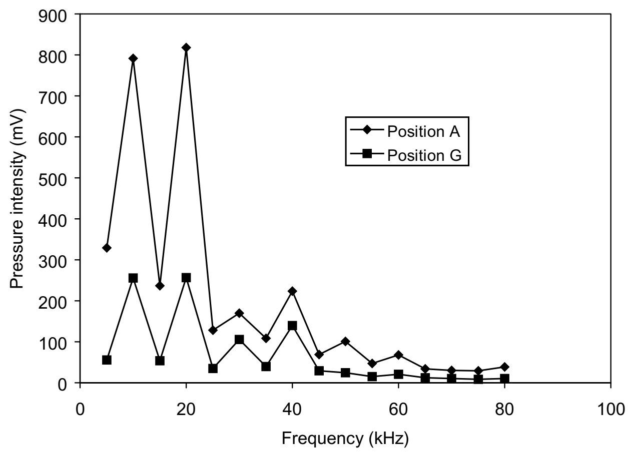

Fig. 1. FFT response curves for two sample positions.

peak at the driving frequency. This is due to the fact that in the case of water, the acoustic emission spectra comprises of ultrasound waves with the pressure pulses due to the bubble oscillation/collapse superimposed on it. However for the non-cavitating liquids such as silicon oil, cavitation phenomena is not observed resulting in only the fundamental frequency peak. It can be also seen from the figure that the intensity of the peak at the driving fundamental frequency is greater for the position A as compared to the position G (A is nearer to the horn tip and intensity decreases as one moves away from the source). Thus the FFT spectrum can be used as criteria in deciding whether cavitation is occurring in the chosen liquid medium, if visual observation is not permissible.

The complete profile for variation of the local cavitational activity and the resulting pressure field with axial distance from the horn tip has been shown in Fig. 2. Overall, a decreasing trend in the cavitational activity has been observed with an increase in the distance from the horn tip. It can be also seen that the cavitational activity and the resulting pressure field shows intermediate peaks i.e. increase in the intensity with the distance between two peaks approximately equal to $\lambda/2$. Pugin [6] and Romdhane et al. [7,8] have also obtained similar results. The intermediate increase in the local ultrasonic activity can be attributed to the prominent subharmonic peaks (observed at frequency = 10 kHz) at these locations. The FFT response at position G (distance = 4.5 cm) shows a dominant peak in pressure intensity at frequency of 10 kHz, which is equal to $f/2$ where f is the driving frequency for the horn (refer Fig. 1). Such a peak was not observed at the earlier location, F, at a distance 4 cm from the tip of the horn along the axis. Esche [9] has shown that sub-harmonically oscillating bubbles evolve into transient cavities. Transient cavitation is more efficient and results in relatively more violent collapse of cavities as compared to stable cavitation resulting into higher local pressure.

Experiments were also performed at different radial locations to check for the radial homogeneity of the ultrasonic activity. The results have been shown in Fig. 3. It can be easily seen from the figure that the ultrasonic intensity measured in terms of local pressure decreases with an increase in the distance away from the axis passing through the centre of the irradiating surface. The observed results are in accordance with the results obtained by Romdhane et al. [7] who have shown that the ultrasonic activity diminishes at a distance of 3 cm away from the centre axis.

At all these locations, experiments were also performed using KI decomposition as the model reaction. One to one correspondence was observed between the measured pressure amplitudes using the hydrophone and the extent of iodine liberation. The trends obtained with both the methods (measurement of pressure or decomposition of KI) were similar indicating that either of the method can be used for mapping of ultrasonic reactors. Faid et al. [10] have also obtained same results

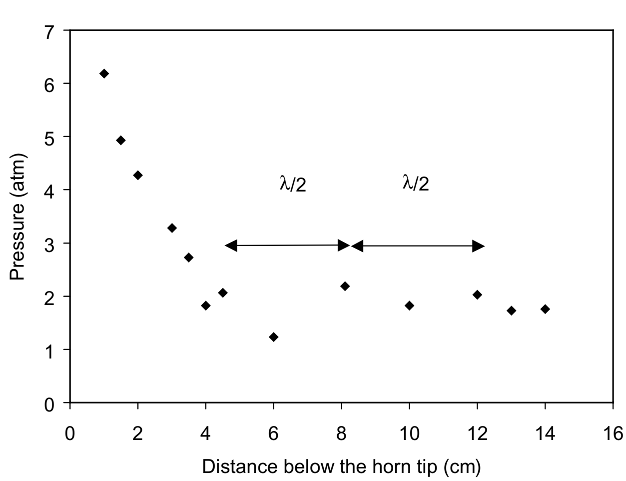

Fig. 2. Variation of the ultrasonic activity in the axial direction for ultrasonic horn.

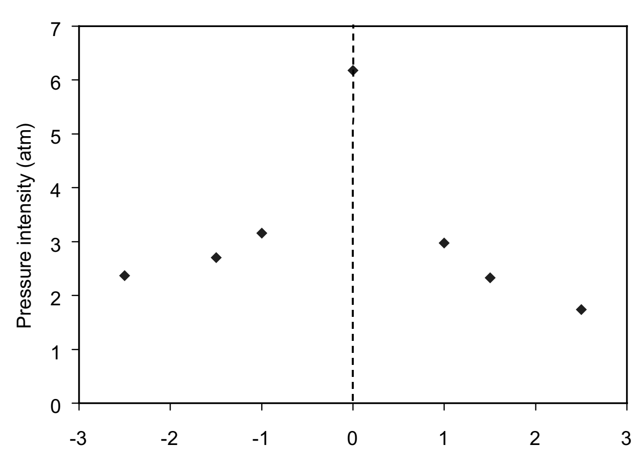

Fig. 3. Variation of ultrasonic activity in the radial direction (- - - indicates axis of the horn).

with different methods used in their work (measurement of cavitational activity with the help of electrochemical, thermoelectrical and chemical probes). The details about the dependency of KI decomposition rates with the driving measured pressure fields have been discussed later.

The observed results conclusively establish that the ultrasonic activity is maximum at the zone very near to the irradiating surface and drastically decreases as one moves away from the source both in axial as well as radial direction.

3.2. Quantification of iodine yield in terms of measured pressure amplitude

Fig. 4 shows the variation of the amount of iodine liberated with the total pressure amplitude (summation of pressure amplitudes at driving as well as sub-harmonic and harmonic frequencies) in the ultrasonic horn reactor. It can be easily seen that there exists a linear relationship between the two given by the following equation, which is obtained by curve fitting exercise with an R^2 value of 0.92 indicating good fitting:

$$\text{Iodine yield } (\mu\text{g/lit}) = 1.146 \times \text{total pressure amplitude (atm)} - 1.05 \quad (2)$$

A quick analysis of the equation also gives that the threshold pressure required for the start of iodine liberation i.e. the positive effects of cavitation phenomena as equal to 0.92 atmospheres (as measured by the hydrophone). The existence of threshold pressure for iodine liberation can be attributed to the fact that certain minimum number of free radicals are required for the release of measurable amount of iodine. The radicals will be generated only when the conditions are favourable for the pyrolysis of water i.e. only above certain threshold pressure amplitude resulting into the collapse with some minimum violent conditions (collapse pressures and temperatures).

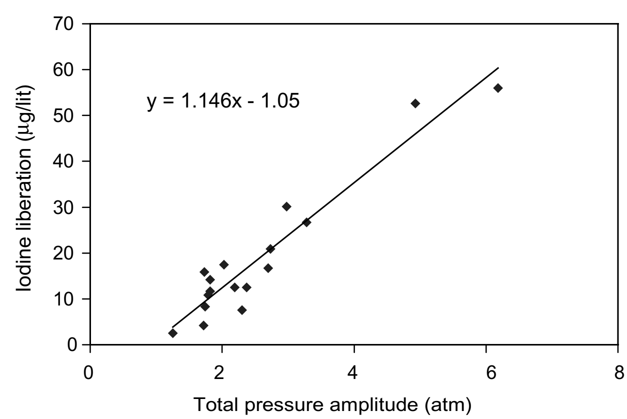

Fig. 4. Correlation fitting for iodine liberation as a function of total pressure amplitude.

3.3. Variation of iodine yield with the maximum bubble size

As explained earlier the values of measured pressure pulses at the driving frequency were used in the bubble dynamics simulations for the estimation of the maximum bubble size reached during the cavity life cycle. The simulated values of maximum bubble size give an indication about the type of cavitation taking place at the different locations. Cavitation bubbles are classified as stable or transient according to their radial motion under the influence of the acoustic field. Flynn [11] has characterized the stable and transient cavities as follows; if the ratio of R_{max}/R_0 during the radial motion exceeds the minimum value given by $(7.48P_{go}/P_0)^{1/3}$, the resulting cavitation will be transient. For air bubbles in water, this ratio is found to be equal to 2. In the present work, numerical simulations result in an R_{max}/R_0 ratio of greater than 2 (typical range obtained is 5–6 depending on the operating conditions) for all the measured pressure amplitudes indicating the occurrence of transient cavitation at all the considered locations, which is also confirmed by the release of measurable amount of iodine. It should be also noted that in the case of stable cavitation, the cavities just oscillate without releasing significant amount of energy and hence the chemical reactions do not occur.

The variation in the amount of iodine liberated with the maximum radius ($[R_{max}/R_0]^3$ is considered as the volume of the collapsing bubble is relative to the amount of the free radicals released into the reactor) has been depicted in Fig. 5. Following equation has been obtained with the curve fitting exercise of the data:

$$\text{Iodine liberated } (\mu\text{g/lit}) = 2.9688 \times [(R_{max}/R_0)^3]^{0.4891} \quad (3)$$

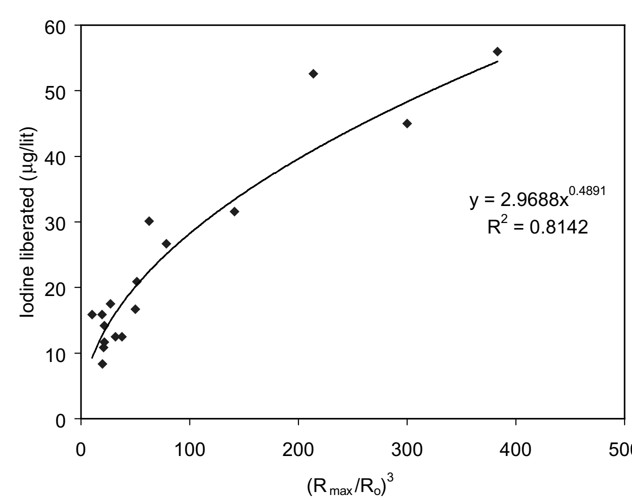

Fig. 5. Variation of the iodine liberation with the ratio $(R_{max}/R_0)^3$ obtained from numerical simulations.

It has been observed that the R^2 value for the fitted equation is 0.8142 indicating that the equation does not fit that well with the experimental data; still is indicative of the type of data analysis that needs to be done in the design procedure. More work is being done with mapping at increased number of locations for developing an accurate relationship. It can be also seen that the three points at very high values of $(R_{max}/R_0)^3$ i.e. >200 show relatively same values of iodine liberation indicating that even if the number of free radicals generated due to the collapse of cavities are significantly higher (as predicted by theoretical simulations and growth of the cavity), the actual number of free radicals attacking the KI molecules are nearly the same (as given by similar iodine liberation). This can be also attributed to the fact that at higher amounts of free radicals, the recombination reaction of the free radicals becomes significant.

It is clear that the rate of iodine liberation will be higher at higher R_{max} values (only till an optimum value), which in turn depends on the local pressure amplitude (direct variation has been observed). Thus for higher local power dissipation i.e. higher ultrasound dissipation intensities (but again till an optimum value of intensity), the amount of iodine liberated will be higher. Entezari and Kruus [12] have investigated the effect of the increase in intensity (and hence the pressure amplitude) on reaction rates of decomposition of KI and reported an increase in the same with an increase in the power dissipated. Though the observations of Entezari and Kruss [12] are based on the global rates, the comparison is just to indicate the correctness of the trend observed in our measurements. It must also be kept in mind that there exists an optimum power dissipation beyond which the beneficial effects of dissipating more power are not observed [13].

It should be noted that the developed correlation, unique of its kind, along with the earlier correlation of cavitational yield in terms of developed collapse pressure [1] are only trend setting and by no means, generalized one. There is ample scope for further work, which may include establishing the validity of the equations over a wider range of operating parameters, exactly quantifying the number of free radicals generated during the collapse (more importantly estimating the actual number taking part in the reaction, by considering the number of free radicals generated, time of collapse of the cavities and the lifetime of the free radical), estimating the size of the nuclei/cavity that will be strongly dependent on the type of equipment used for generation of cavitation, combining the effects of collapse pressure generated and the maximum bubble size reached etc. Attempts in this direction are also being made in this department.

4. Conclusions

The nature of the sound/cavitational field existing in any ultrasonic reactor is substantially non-uniform with spatial variations in the axial as well as radial directions. In general the activity show a decreasing trend away from the transducer and is maximum in the vertical plane of the emitting area. The type of the sensor used (hydrophone for pressure measurements and chemical source in terms of measuring rates of chemical reaction) for mapping the local field do not affect the trends observed and one to one correspondence is observed between the two.

Correlations have been developed for the prediction of iodine liberated as a function the pressure amplitude and the maximum bubble size reached predicted using theoretical simulations. These correlations, unique of its kind, should be taken as the initial step, which will lead us to a generalized correlation for the estimation of the sonochemical yield in terms of the operating and geometric parameters of the reactor. Such generalized correlations will be very helpful in effective design and scale up of the sonochemical reactors.

Acknowledgement

Authors would like to acknowledge the funding of the Indo-French Center for Promotion of Advanced Research (Centre Franco-Indien Pour La Promotion de La Recherche Avancee), New Delhi, India for the collaborative research work.

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