



# Destruction of Rhodamine B using novel sonochemical reactor with capacity of 7.5 l

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## Abstract

Sonochemical reactors offer excellent promise in the wastewater treatment applications due to the creation of local hotspots with overall ambient operating conditions and release of highly reactive free radicals. However, its application in the actual industrial practice is hampered by the fact that the existing available information related to the optimization of operating parameters is restricted mainly to small scale operations with reported experiments with capacity in the range of few milliliters to 1 l. With this information, efficient scale up and successful operation of industrial scale reactors is almost impossible. Thus, design of novel large-scale sonochemical reactors and subsequent testing of the same for destruction of different pollutants is the need of the present hour. In an attempt to move one step ahead in the design process, a novel sonochemical reactor with a capacity of 7.5 l has been developed and tested using destruction of Rhodamine B as a model reaction. Effect of various operating parameters such as frequency of irradiation, use of multiple frequencies and power dissipation into the system on the extent of degradation has been studied. Experiments have also been performed with the KI decomposition (model reaction typically used in investigating cavitation effects) so as to establish the dependency of the trends in the variation of the extent of degradation with the operating parameters on the cavitation intensity requirements for a specific application. For the degradation of Rhodamine B, power dissipation into the system was found to be the controlling parameter and the extent of degradation is directly proportional to the power dissipation with a coefficient of 74.1.

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## 1. Introduction

Cavitation can be defined as the phenomena of formation, growth and subsequent violent collapse of microbubbles or cavities occurring in extremely small intervals of time releasing large magnitudes of energy

over a very small location but at millions of places in the reactor simultaneously. The end result is the generation of local hotspots and also the formation of reactive hydroxyl and hydroperoxyl radicals, which are strong oxidizing agents. These conditions are suitable for the oxidation of a variety of organic compounds and can be used in the wastewater treatment schemes, if properly designed for the large-scale application. Among the various modes of generation of cavitation, use of ultrasound results in maximum intensity

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of cavitation as confirmed by the quantification of collapse pressure pulse presented in the earlier work [1,2]. The application of acoustic cavitation in the area of liquid effluent treatment is not new to the researchers and there have been many illustrations in the literature dealing with the application of ultrasound for the destruction of variety of contaminants [3–9]. Excellent reviews are also available on the use of sonochemical reactors depicting in details the mechanism of generation of cavitation as well as its action, types of reactors used, optimum operating conditions and different applications [10–13]. It should be noted here that the majority of the studies are on a laboratory scale with the capacity of the reactor of the order of few milliliters. It is unreliable to use this knowledge and design large-scale reactors for wastewater treatment applications due to very large scale-up ratios and also non-suitability of ultrasonic transducers to operate at high frequencies and high power dissipation rates required for large scale operation. Thus development of an efficient sonochemical reactor with operating capacity of few liters, checking the efficacy of the designed reactor for variety of pollutants and understanding the controlling operating parameters for the specific reactions seems to be the need of the hour and the present work focuses on this aspect.

Based on the extensive analysis of the literature and our earlier work (theoretical predictions of the bubble dynamics equations [1,14] and experimental work [15–17]), following important points appear to be important for large-scale sonochemical reactors and also have been considered in the design of the novel reactor developed and used in the present work:

- 1) Multiple frequencies result in higher growth and subsequently more violent collapse of the cavities and hence higher intensities of cavitation as compared with the single frequency operation.
- 2) It is better to dissipate same power through larger areas of dissipation, which also results in lower operating intensity of irradiation and hence more violent collapse of the cavities.
- 3) Multiple transducers give better cavitation effects due to increased cavitationally active volumes, in terms of more number of cavitation locations.
- 4) Parallel plate reactors are more efficient in generation of the standing waves and hence should result in more intense cavitation events though the exact

mechanism of generation of standing waves and its effect on the reaction yields is still far from being understood.

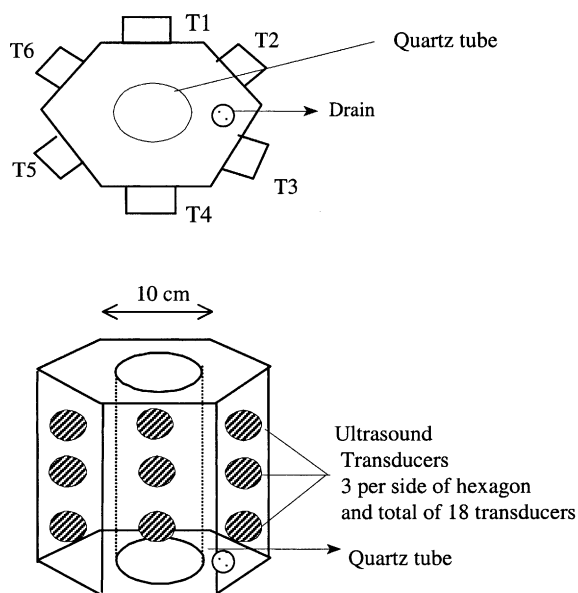
- 5) Hexagonal geometry of the reactor results in near-uniform distribution of the cavitation activity minimizing the existence of the dead zones.

Degradation of Rhodamine B (a typical pollutant observed in the effluent stream of dye industry) has been studied in this novel triple frequency flow cell at an operating capacity of 7 l. First, experiments have been performed with KI solution so as to investigate the efficacy of the novel reactor (Weissler reaction is generally used for investigating the efficacy of cavitation reactors). Aim was also to validate whether the trends observed for the model reaction in terms of the variation of operating parameters are equally applicable to the actual pollutants and also to identify the controlling operating parameters in deciding the extent of degradation.

## 2. Experimental section

### 2.1. Sonochemical reactor

The hexagonal triple frequency flow cell has a total capacity of 7.5 l and can be operated in batch (in the present work, operation has been restricted only to batch mode with operating volume of 7 l) as well as continuous mode (the flow cell can also be operated in continuous mode with the solutions entering at the bottom of the reactor and leaving from the top of the cell). Schematic representation of hexagonal flow cell has been given in Fig. 1. Transducers (three transducers per side; hence a total of 18 transducers in the reactor) having equal power rating of 50 W per transducer (i.e. 150 W per side) have been mounted on the each side of the hexagon. The two opposite faces of the flow cell have the same irradiating frequency. The operating frequency of transducers is 20, 30 and 50 kHz (all the transducers on a particular side emit same frequency; the details have been depicted in the Fig. 1). There is a possibility of controlling the mode of operation and there are seven different configurations of frequencies (20, 30, 50, 20 + 30, 30 + 50, 20 + 50 and 20 + 30 + 50 kHz), which can be used in the reactor. The maximum rated power dissipation is 900 W when all the transducers with a combination of 20 + 30 + 50



Effluent is taken in batch mode (Volume = 7L)  
 Power supply = 150 W/side  
 T1 and T4 operate at 20 kHz  
 T2 and T5 operate at 30 kHz  
 T3 and T6 operate at 50 kHz

Fig. 1. Schematic representation of triple frequency flow cell used in the experimental work.

kHz frequencies are functional. The actual power dissipation into the system will be different and will be dependent on the transfer efficiency of the transducers.

## 2.2. Energy efficiency analysis

Energy efficiency gives an indication of the quantity of energy effectively dissipated in the system, a fraction of which is utilized for the generation of cavities, whereas the remaining fraction appears as the mechanical/heat energy and both should be as high as possible for the particular cavitating equipment. Calorimetric method is used to determine the energy efficiency of the sonochemical reactor and has been described in details in the earlier work [15].

## 2.3. Degradation of potassium iodide

Weissler reaction i.e. decomposition of potassium iodide liberating free iodine has been widely used as a

model reaction to check the efficacy of the cavitation equipment or to investigate the effect of different operating parameters [15]. It is, however, important to check whether the results/trends obtained with this model reaction are equally applicable with the actual effluents/other complex chemicals and to the best of our knowledge, no earlier work has concentrated at this effort.

The extent of iodine liberated during the reaction has been estimated with the help of UV/VIS spectrophotometer by measuring its absorbance at 355 nm. Pre-calibrated charts were used for the estimation of the concentration of the pollutant from the absorbance values. Experiments have been performed using 1% KI solution for 30 min of irradiation time.

The total irradiation time had to be divided into equal intervals of 10 min each due to the limitations of introduction of a direct cooling apparatus into the system and due to the fact that the transducers cannot be used continuously for extended periods. The temperature rise achieved during 10 min operation was 1–3 °C depending on the power input to the system which indicates that the operation may be considered as isothermal. After operation of 10 min, the reactor was kept off for about 15 min for natural cooling of the transducers. In this gap of 15 min, the solution was also removed through a drain, circulated through an external heat exchanger for cooling of the reaction mixture (cooling was achieved using chilled water at 10 °C) and refilled into the reactor after achieving cooling. Once the temperature of the reaction mixture was achieved at ambient conditions, the next cycle of irradiation was started. It should be also noted that during the re-circulation, the system is not hermetically sealed and some natural aeration might occur during the transfer of solution from the reactor to the cooling bath and back. Aeration, if occurring only helps in enhancing the intensity of cavitation and hence the extent of degradation, though at the present instance its quantification is not possible.

## 2.4. Degradation of Rhodamine B

Removal of color (arising from the use of synthetic dyes) from the wastewater streams of the textile/dyes industry is a well-investigated problem over the years [18]. The conventional methods such as the aerobic and anaerobic biological oxidation, wet air oxidation,

which are otherwise much successful in the removal of color due to most of the dyes, fail in the case of aromatic amine dyes. These circumstances lead to a need for developing an alternative technology for the decolorization and use of sonochemistry can be one such alternative. Sivakumar and Pandit [9] have studied the decolorization of aqueous solution of Rhodamine B (tetraethylrhodamine also known as Basic Violet 10) using the laboratory scale ultrasonic reactors (standard immersion reactor i.e. ultrasonic horn, cleaning tank type reactor i.e. ultrasonic bath and dual frequency flow cell) and reported that the technique of acoustic cavitation indeed can be used for the decolorization purpose. The free radicals as well as the conditions of high temperature and pressure generated in the cavitation phenomena result in the breakage of the chromophores leading to decolorization of the effluent, even though the total reduction in the COD was not significant indicating only the breakage of the chromophores rather than complete mineralization, also indicating that the sonochemical

treatment might only be effective in decolorization of the textile/dyes industry effluents.

In the present work, degradation of Rhodamine B have been studied in the large scale reactor i.e. the triple frequency flow cell with an initial concentration of  $\sim 4.5 \mu\text{g/ml}$  and irradiation was continued for 30 min. At regular time intervals, solution was withdrawn and analyzed for absorbance at 553 nm. The operating procedure for the maintenance of temperature was similar to that explained earlier.

### 3. Results and discussion

#### 3.1. Energy efficiency calculations

The energy efficiency for the triple frequency flow cell was found to be in the range of 60–85% depending on the mode of operation i.e. single or multiple frequency operation (Fig. 2). It must be noted that the change in the energy efficiency is solely attributed

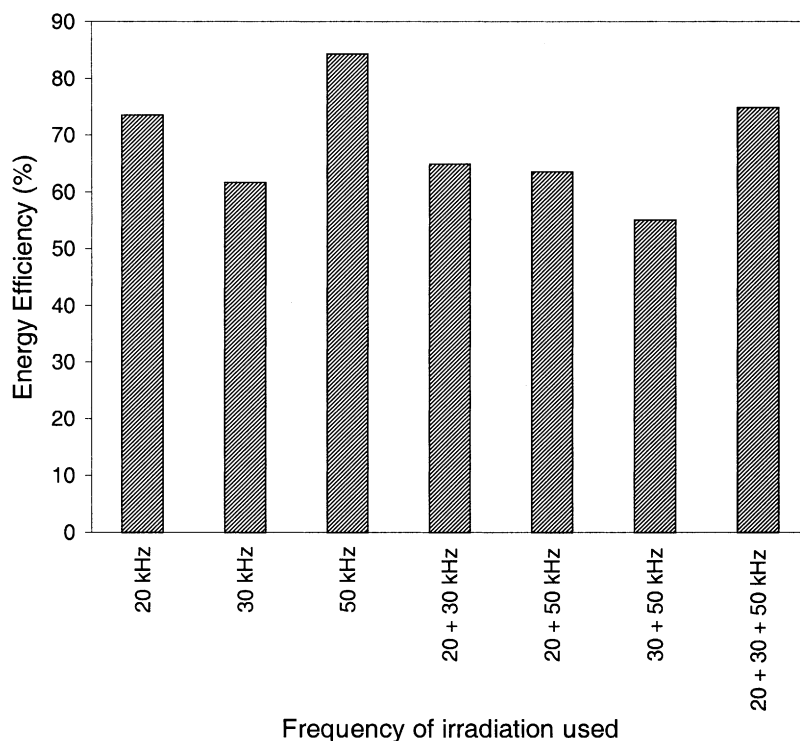


Fig. 2. Variation of energy efficiency with the operating mode in the reactor.

to the inefficiency of the multiple transducers on the particular side of the hexagonal reactor and have no dependency on the frequency of irradiation. Nevertheless, comparison with the earlier work, where similar energy efficiency analysis has been done for the conventional reactors [15] indicates that the energy efficiency is almost eight to ten-times higher as compared with the ultrasonic horn and about 100% more as compared with the ultrasonic bath where three transducers are attached at the bottom of the reactor as well as dual frequency flow cell where three transducers are attached on the two opposite faces of the reactor with rectangular cross-section. It may also happen that the position of the transducers i.e. on parallel plates facing each other, improves the fluid coupling and circulation due to the acoustic streaming resulting in better utilization of the supplied energy. This can be also confirmed by the fact that dual frequency flow cell (transducers arranged on two parallel faces) has more energy efficiency as compared with ultrasonic bath (transducers only at the bottom) as obtained in the earlier work [15]. Thus it can be said that multiple transducers positioned on parallel plates facing each other and with larger areas of dissipation are much better in transforming the input electrical energy into the useful energy available for cavitation events.

Further, it was also observed that the energy efficiency values are almost unaltered by the pollutant used in the reactor i.e. experiments with potassium iodide and Rhodamine B resulted in similar transfer of the supplied electrical energy indicating that operation with real industrial effluents will also result in similar amount of energy being available for the generation of cavitation and hence similar cavitating conditions in terms of magnitudes of local temperatures/pressures and the amount of free radicals are expected to be generated.

### 3.2. Degradation of KI

Fig. 3 shows the variation of iodine released as a function of time for the 30 kHz operation; similar trends have been obtained for the operations at other frequencies as well. It can be seen from the figure that iodine liberation is observed within 2 min of irradiation time indicating that there is no induction time required for the decomposition of potassium iodide. This can be attributed to lower intensity requirements

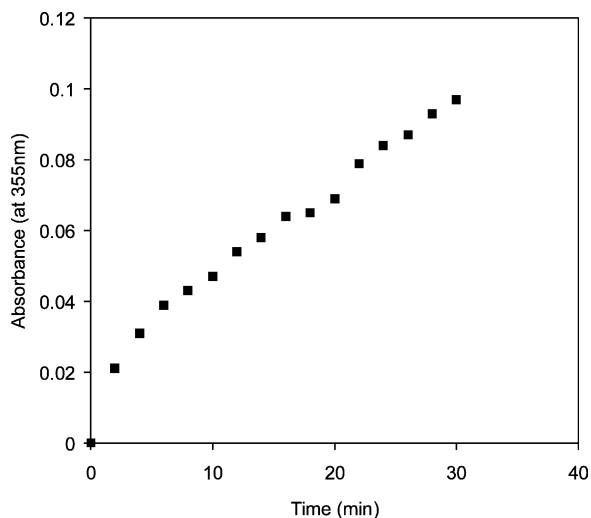


Fig. 3. Variation of the iodine concentration with time at operating frequency of 30 kHz.

for the initiation and propagation of the reaction i.e. as soon as the cavitating conditions (high temperature, pressure pulse and release of free radicals) are generated, degradation of potassium iodide starts. Fig. 4 gives the extent of iodine liberated in 30 min of irradiation time for all the combinations of frequencies of irradiation (seven in total). It can be observed that the extent of liberation of iodine increases with an increase in the frequency of irradiation for operation with single frequency; though the increase from 30 operation to 50 kHz operation was not substantial. This is in accordance with the predictions of bubble dynamics equations, which indicate that the collapse temperature, pressure and also the number of free radicals generated increases with an increase in the frequency of irradiation [1,14]. For multiple frequency irradiation, combination of 20 + 30 + 50 kHz gives maximum liberation of iodine followed by 30 + 50, 20 + 50 and 20 + 30 kHz combinations in the same order. Also the extent of iodine liberated was more for the dual and triple frequency combinations as compared with the single frequency operation. This is again in accordance with the bubble dynamics predictions [19], which indicated that cavitation is more intense for the combination of frequencies as compared with the single frequency irradiation.

It is also important to check whether the obtained increase in the extent of iodine liberated is only

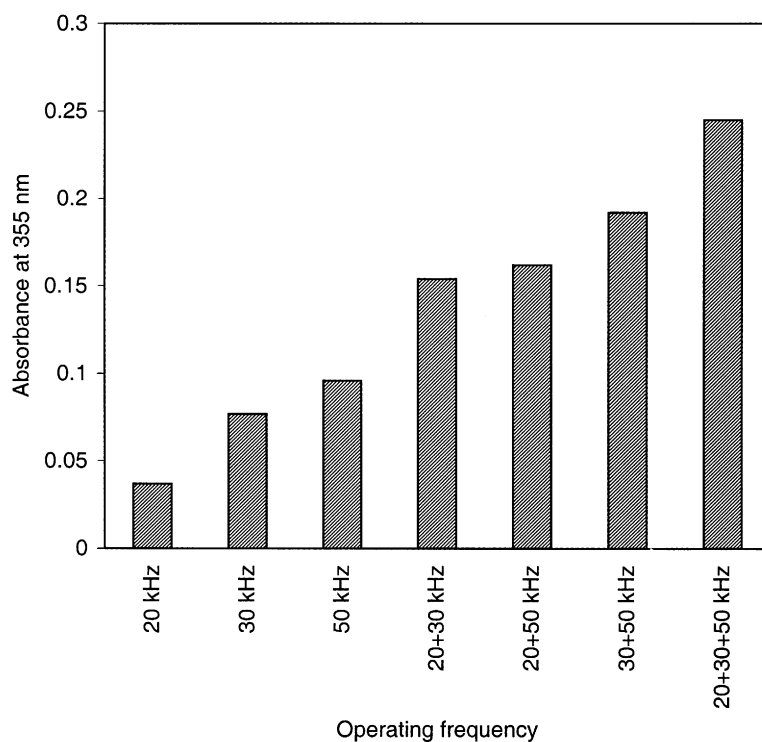


Fig. 4. Effect of the operating frequency on the extent of degradation of KI for complete irradiation time span.

due to the combination of the cavitation fields produced by the multiple transducers or a synergism also exists. For the case of combination of 20 and 30 kHz frequencies, the observed iodine release was  $1.286 \times 10^{-4}$  g/l whereas for the individual operations of 20 and 30 kHz, the iodine liberation was found to be  $3.091 \times 10^{-5}$  and  $6.433 \times 10^{-5}$  g/l, respectively. Thus, the extent of iodine liberation for the combination operation was about 35% more as compared with the addition of the iodine released in the individual operations, indicating that the use of multiple frequencies is indeed synergistic. Further, when similar analysis was done for the other combinations of higher frequencies or the triple frequency operation, it was observed that the advantage of synergism is only to the tune of 10–20% increase over the additive values. Hence it can be said that it is better to have combination of lower frequencies rather than combining the higher frequencies of irradiation through multiple transducer operation to get maximum synergism.

It is interesting to compare the amount of iodine liberated as obtained in the present case with that

obtained in the conventional reactors e.g. ultrasonic horn. In the earlier work [15], experiments were done using 50 ml of 1% KI solution with ultrasonic horn operating at 20 kHz frequency of irradiation and supplied electric power as 240 W. The amount of iodine liberated was estimated at  $5.1 \times 10^{-7}$  g/W. In the present case, for the multiple frequency operation (20 + 30 + 50 kHz), the amount of iodine liberated is  $2.278 \times 10^{-6}$  g/W, which is about four-times more than that obtained with the ultrasonic horn. A similar analysis of comparing the efficacy of the present reactor with ultrasonic bath type of reactor available in the department (capacity of reactor is 1500 ml, operating frequency of irradiation as 20 kHz and power input as 120 W) indicated that similar levels of power dissipation into the system results in better results for the triple frequency flow cell, at least for the reaction of degradation of potassium iodide. It is also important to note that the volume that is being used here is also 10 to 15-times more than being treated in ultrasonic bath type reactors (three transducers arranged in triangular pitch at the bottom of the reactor).

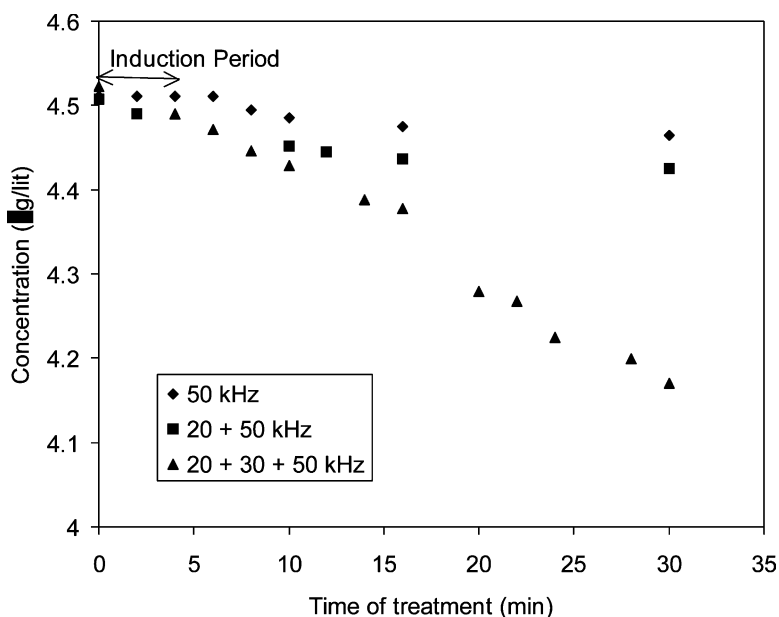


Fig. 5. Variation of concentration of Rhodamine B with time of treatment for different operating modes of irradiation frequencies.

Thus the efficacy of the novel triple frequency hexagonal flow cell is well established as compared with the conventional reactors (immersion and cleaning tank type of reactors) considering the model reaction and also looks promising for large-scale applications.

### 3.3. Degradation of Rhodamine B

Fig. 5 gives the variation of the concentration of Rhodamine B with time for the various operations (one each from single and dual operations has been shown for clarity). An induction period can be seen for the single frequency operation (50 kHz has been depicted in the figure), beyond which there is a decrease in the concentration of Rhodamine B, indicating that a certain minimum number of free radicals and cavitation intensity is required for the onset of the degradation process. Similar induction periods were also observed for the other individual frequency (20 and 30 kHz) operations. It was also observed that the induction period decreases with an increase in the frequency of operation (induction period of 14 min for 20 kHz operation, 10 min for the 30 kHz operation and 6 min for the 50 kHz operation; all the concentration profiles are not shown in the figure for clarity). Further the induction period was absent for the dual

and triple frequency operations (Fig. 5). This can be attributed to the fact that collapse is more violent (and hence the free radical generation rate is higher) for higher frequencies or for the combination of frequencies. Also the number of cavitation events is more for the dual and triple frequency operations due to the increased power dissipation for the multiple transducers. Thus the overall intensity of cavitation (number of cavitation events multiplied by the cavitation intensity obtained by the collapse of single cavity) is higher at higher and multiple frequency operation resulting in the initiation of the degradation process for this complex pollutant almost instantaneously.

The extent of degradation for the single frequency operation for the complete irradiation time was found to be in the order  $20 > 30 > 50$  kHz (Fig. 6) which is quite contrary to that observed for the KI decomposition and to the predictions of numerical simulations of bubble dynamics equations for single cavity collapse. This can be attributed to the fact that overall intensity of the cavitation phenomena depends on two factors viz. violent collapse of the single cavity (due to an increase in the frequency, the collapse pressure increases) and number of cavitation events (an increase in the power dissipation increases the number substantially). The power dissipation into the liquid

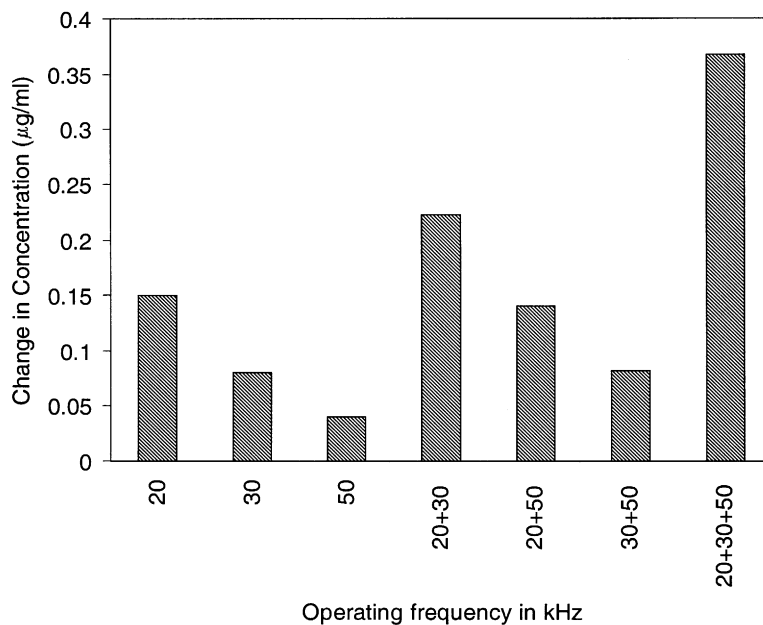


Fig. 6. Effect of the operating frequency on the extent of degradation of Rhodamine B for complete irradiation time span.

was found to be the maximum for the 20 kHz transducers followed by 30 kHz and least for the operation using 50 kHz transducers. Thus though the collapse of single cavity is becoming more violent, the number of cavitation events decreases due to a decrease in the power dissipated into the system and hence overall intensity of cavitation decreases. Due to the fact that Rhodamine B degradation requires intense conditions, the extent of degradation also decreases. In the case of potassium iodide decomposition, however, due to lower cavitation intensity requirements, the increase in the collapse pressures due to an increase in the operating frequency alone is sufficient to increase the extent of iodine liberation even though there is a decrease in the number of cavitation events. It must be also noted here that extent of increase in the degradation observed for an increase from 30 to 50 kHz was marginal due to strong negative contribution of decreased power dissipation. Thus it can be said that the number of cavitation events (decided by the total power dissipation into the system) is the controlling factor in the case of Rhodamine B degradation whereas any one factor i.e. either the collapse intensity of single cavity or the change in the number of cavities is sufficient to cause an increase in the extent of degradation of potassium

iodide. These results also emphasize the fact that the trends obtained for studies with the model reaction are not necessarily applicable to all the reactions requiring different intensities of cavitation. Thus, it is important to perform laboratory scale studies for establishing the optimum set of operating parameters for the application in question unless data are available in the open literature with similarity in the range of operating parameters including reactor configuration.

For the dual and triple frequency operation, the extent of degradation was found to be more as compared with the single frequency operation. This increase, however, is mainly due to the fact that the power dissipation into the liquid increases thereby increasing the number of cavitation events. The trend in dual frequency operation was found to be  $30 + 50 < 20 + 50 < 20 + 30$  kHz which is again attributed to the decrease in the power dissipation into the system in the same order, though there is also a simultaneous increase in the intensity of the cavitation for the single cavity (due to an increase in the net frequency of irradiation). These results also confirm the controlling factor as the number of cavitation events and hence the power dissipation into the system. Thus, in the case of Rhodamine B degradation, it can be said



that dissipating more power by using multiple transducers is more beneficial as compared with increasing the frequency of irradiation or using multiple and different frequencies. It should be also noted that there will not be an indefinite increase in the number of cavitation events and also with a large number of cavities present in the system, there is likelihood of coalescence of cavities resulting into an increase in the size of the nuclei (and hence lower intensity of cavitation). Also at higher power dissipation and hence higher intensities of irradiation, there exist a large number of gas bubbles or cavities in the solution, which scatter the sound waves to the walls of the vessel or back to the transducer. Thus lesser level of energy focussing or concentration occurs although the vessel is exposed to higher and higher intensities. Thus there may be an optimum value of power dissipation beyond which the beneficial effects in terms of increase in the extent of degradation will not be present. Ondruschka et al. [20] have shown that the rate constant for sonolytic degradation of MTBE increases linearly till an intensity of  $5 \text{ W/cm}^2$  corresponding to a total power input of 100 W beyond which there is only a marginal increase in the rate constant. Gutierrez and Henglein [21] have also obtained similar results with the decomposition of aqueous potassium iodide solution.

As synergism was observed in the case of potassium iodide degradation for the combination of frequencies, similar analysis was done in the case of Rhodamine B degradation studies also. For the 20 + 30 kHz operation, the combined operation gave a concentration change of  $0.138 \mu\text{g/ml}$  whereas the individual operations resulted in concentration change of  $0.043 \mu\text{g/ml}$  for 20 kHz operation and  $0.1 \mu\text{g/ml}$  for 30 kHz operation. Thus the concentration decrease observed for the combined operation is marginally less as compared with the addition of the individual operations. Hence, it can be said that synergism is not observed for the degradation of Rhodamine B, requiring higher intensity of cavitation. Similar results have been obtained for the other combination operations i.e. 30 + 50, 20 + 50 and 20 + 30 + 50 kHz, showing similar to marginally less degradation as compared with the individual operation results (when added for overall effect). The observed results may be attributed to the fact that when the multiple frequency operation is compared with the additive effects, the total power dissipation remains the same in two cases and as the

power dissipation is the controlling factor, the net degradation also remains the same or is marginally less. Thus it is important to check the controlling operating parameter for the reactor before deciding the reactor configuration.

### 3.4. Correlation for the percentage degradation of Rhodamine B

As for the case of Rhodamine B degradation, actual power dissipation into the system is observed to be the controlling factor; a correlation was developed for the prediction of percentage degradation of Rhodamine B (at the end of 30 min) as a function of power dissipation per unit volume (Fig. 7) and is given as:

Percentage Degradation

$$= 74.1 (\text{Power dissipation in W/ml})$$

The equation is valid only over the range of power density as used in the present work (0.025–0.1 W/ml) and should not be generalized as discussed earlier. Comparison with earlier work [9] with single frequency/transducer reactor indicates that the same power dissipation is better transformed into useful cavitation effects in the case of this triple frequency hexagonal flow cell. The exponent over power dissipation was found to be 0.64, which is less as compared with the present case (equal to 1). The constant

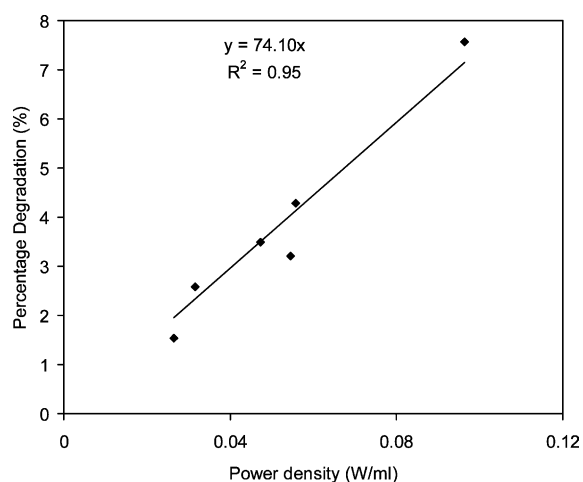


Fig. 7. Equation fitting for the prediction of percentage degradation of Rhodamine B solution at the end of 30 min.

in that equation (75.9) is marginally different from the present case; it is strongly dependent on the time of operation and initial concentration of the pollutant, which was the same for the two cases. Thus the multiple transducer reactors with large irradiating surface are more effective for wastewater treatment as compared with conventional single frequency/single transducer reactors.

### 3.5. Process intensification for degradation of Rhodamine B

The extent of degradation for the case of Rhodamine B was found to be quite less and hence some studies for enhancing the intensity of cavitation events were aimed at, using aeration and presence of solid particles.

#### 3.5.1. Aeration

Senthilkumar et al. [22] have reported that the rates of degradation of potassium iodide were substantially higher in the initial irradiation periods due to the presence of dissolved gases and decreased as time progressed due to the degassing action of the ultrasound. This can be attributed to higher number of cavitation events due to the presence of gas bubbles in the initial period. Thus, if a continuous source of nuclei in the form of continuous aeration is provided, the overall intensity of cavitation events may increase resulting in an increase in the extent of degradation. Experiments with aeration (at constant rate of  $1.02 \text{ cm}^3/\text{s}$  using a multipoint sintered sparger) indicated that the extent of degradation increased by about 30% over entire 30 min of operation (Fig. 8 shows a sample representation of the data obtained for 20 kHz operation).

It was also observed that the extent of enhancement is more or less the same for all the other operating modes of single (30 and 50 kHz as operating frequency) and dual frequency irradiations (20 + 30, 20 + 50, 30 + 50 kHz). Further, when the operation was switched to triple frequency operation (20 + 30 + 50 kHz), the extent of enhancement (comparison of rate of degradation in the presence and absence of aeration) was found to be 25% which is marginally less as compared with the single and dual frequency operations. This observed result can be possibly attributed to the fact that too much cavitation results in decoupling effect though the extent

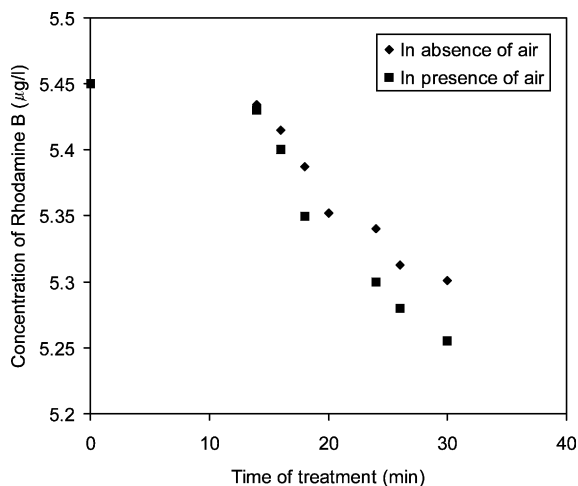


Fig. 8. Effect of continuous aeration on the extent of degradation at operating frequency of 20 kHz.

of effect will be lower due to the multiple transducer irradiation as compared with the conventional horn system (in the earlier work with formic acid degradation [23], it has been shown that aeration results in a decrease in the extent of degradation in the case of ultrasonic horn). Thus aeration can be used as one of the methods for intensifying the cavitation activity in the large-scale sonochemical reactors.

It should be also noted that more work is indeed required for understanding the detailed effect of aeration in terms of parameters such as flow rate of air, using separate sparging of air followed by ultrasonic irradiation (simultaneous operation results in a decrease in the extent of dissolved air due to degassing effect) and experiments with different dissolved gases such as argon, argon/air mixture, ozone etc. before firm recommendations about aeration/gassing as a process intensification step can be made.

#### 3.5.2. Presence of solid particles

Presence of solid particles affects the cavitation phenomena in two different and opposing ways. On one hand, it increases the intensity of cavitation by providing additional nuclei for the generation of cavities as well as by surface cavitation; on the other hand it also acts as a barrier for the propagation of sound waves thereby decreasing the energy transmitted into the system. The negative contribution of the scattering of sound waves might be much lower at

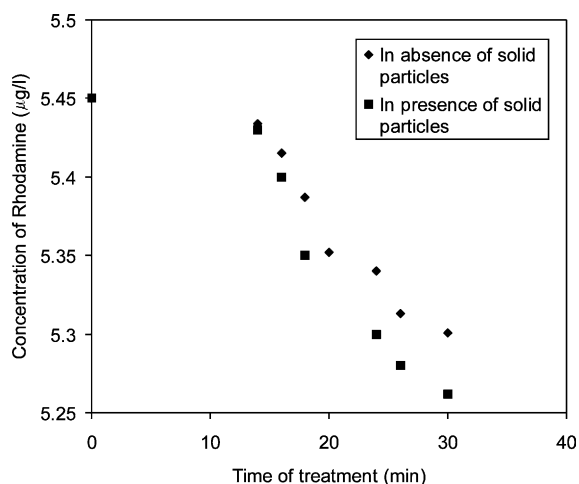


Fig. 9. Effect of the presence of solid particles at loading of 300 ppm on the extent of degradation.

larger scales of operation as compared with the operation using ultrasonic horn (volume treated is just 50 ml) and hence degradation of Rhodamine B was studied in the presence of  $\text{TiO}_2$  particles. It is also important to check the adsorption of the pollutant on the solid particles as adsorption acts as a technique for the physical removal of the pollutant from the solution and will affect the percentage degradation values if not accounted for [7]. Adsorption studies of Rhodamine B on  $\text{TiO}_2$  particles indicated that the absorbance values of the solution were not affected even after 2 h of treatment time ( $\text{TiO}_2$  particles kept in suspension in the aqueous solution of Rhodamine B using mechanical agitation) indicating that the pollutant is not adsorbed on the  $\text{TiO}_2$  particles.

Ultrasonic irradiation studies indicated that the percentage degradation increases by about 25% at the end of 30 min of irradiation for a loading of 300 ppm of  $\text{TiO}_2$  and for 20 kHz operation (Fig. 9). In this case, the extent of enhancement obtained in all the operating modes of frequencies of irradiation was similar i.e. within the error limits associated with standard experimentation. Gogate et al. [24] have also reported that the presence of  $\text{TiO}_2$  particles increases the extent of degradation of formic acid in the same reactor and at similar loading of  $\text{TiO}_2$  particles as used in the present work. Thus it can be said that using multiple transducers for irradiation as used in the present novel reactor gives intensification in the ultra-

sonic degradation process as the negative contribution of scattering of sound waves is minimized.

It should be again noted that the results of the studies using solid particles are only preliminary and just to indicate that enhancement in the extent of degradation can be obtained using solid particles at large scale operation. A more detailed study for studying the effect of concentration and type of the solid particles (solids acting as catalyst with or without adsorption of the pollutants can also be considered), combined effect of solid particles and aeration (may eliminate the use of stirrer for keeping the solids in suspension leading to more uniform propagation of the sound waves in the system) can be taken to get a better insight into the intensification phenomena using solid particles.

#### 4. Conclusions

Destruction of pollutants can be achieved using acoustic cavitation on a large scale of operation (7 l) using multiple frequency/multiple transducer reactors. These reactors are more efficient in the transfer of the supplied electric energy into the system as compared with the conventional immersion or cleaning tank type ultrasonic reactors. The extent of degradation is also similar as compared with ultrasonic horn but most importantly at much lower power density into the system, at least for the two reactions considered in the present work.

It has been also clearly established in the present work that the controlling factor in deciding the overall degradation strongly depends on the requirement of cavitation intensity for the particular reaction and can be either the frequency of irradiation (for the case of potassium iodide decomposition requiring lower cavitation intensity) or the power dissipation/intensity of irradiation (for the case of Rhodamine B degradation requiring higher cavitation intensity). Thus a generalized scale up criteria based on the above two important operating parameters cannot be recommended without laboratory scale experiments for the specific application in question.

The extent of degradation achieved depends strongly on the cavitation intensity requirements of a particular application. Aeration and the presence of solid particles can also be used effectively for enhancing the rates of degradation, where acoustic cavitation alone

does not give significant rates of degradation, more specifically in the case of multiple frequency/multiple transducer reactors.

Overall, it can be said that design and successful application of large scale multiple frequency/multiple transducer reactors is the need for the future and the present work can be considered as useful starting point in this direction.

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