



# Destruction of phenol using sonochemical reactors: scale up aspects and comparison of novel configuration with conventional reactors

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## Abstract

Acoustic cavitation is known to produce conditions extremely suitable for the destruction of pollutants but the industrial use is hampered by the lack of suitable scale-up/design strategies and lack of studies at pilot scale of operation. In the present work, the efficacy of acoustic cavitation for the destruction of phenol has been investigated using a novel triple frequency flow cell with a maximum capacity of 7.5 l. The results obtained have been compared with the conventional laboratory scale ultrasonic horn, which is most commonly used for the applications based on acoustic cavitation and another side entering sonochemical reactor with capacity of 400 ml again based on irradiation with single transducer at frequencies similar to the conventional horn. In the novel sonochemical reactor, the effect of frequency of irradiation on the rates of degradation has also been investigated. Some process intensification studies have also been carried out using surface cavitation in the large-scale reactor as well as the ultrasonic horn with an aim of increasing the rates of degradation.

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## 1. Introduction

The inability of the conventional methods to meet the demands of the newer stringent environmental regulations mainly due to the development of newer molecules refractory towards the biological action, resulted in more inclination towards the development of alternative techniques for effluent treatment either

for complete treatment of the wastewater or with an aim of reducing the toxicity of the effluent stream to a particular level beyond which biological oxidation can be effectively used. Cavitation is one such technique, which can be effectively used for the destruction or modification of complex organic/inorganic chemicals, bio-refractory materials, etc. In general there are four types of cavitation, i.e. acoustic, hydrodynamic, particle and optic, but acoustic cavitation (use of ultrasound for the generation of cavities and the reactors based on this principle are described as sonochemical reactors) has been known to generate intensities

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suitable for the degradation of complex chemicals or mixture of chemicals. There have been many illustrations in the open literature where acoustic cavitation has been used for the wastewater treatment applications [1–8] but majority of the work is restricted to laboratory scale operation. As said earlier, as the existing information is not conducive to the design and successful operation of the large scale reactors, it was decided to concentrate on the scale-up of the sonochemical reactors. In the earlier work [9], a novel large scale sonochemical reactor (triple frequency flow cell) with a capacity of 7.5 l (pilot scale) was designed and tested using a model reaction of decomposition of potassium iodide and another reaction of decolorization of Rhodamine B solution (breakage of the chromophores) requiring relatively lower intensities of cavitation. Comparison with the conventional reactors [10,11] indicated that the triple frequency flow cell gives higher values of energy efficiency as well as cavitation yield as compared with the ultrasonic horn for these reactions. With an aim of further validating the results and extending them to other chemicals, the present work has been undertaken and concentrates on the comparison using destruction of phenol as the model reaction, which requires much higher intensities of cavitation as compared with the earlier two reactions. The present work also evaluates the possibility of using ultrasonic horn for large-scale operations by performing experiments at two different scales of operation.

## 2. Experimental

### 2.1. Sonochemical reactors

#### 2.1.1. Conventional ultrasonic horn

The ultrasonic horn is a conventional immersion type single transducer reactor and was procured from a local manufacturer M/s. Dakshin (Mumbai, India). The operating frequency of irradiation is 22.7 kHz whereas the rated power dissipation is 240 W through a transducer area of 81 mm<sup>2</sup>. The radiating body is made of stainless steel. Ultrasonic horn can be operated in continuous mode for 15 min. However, this results in heating of the equipment and hence, it should be cooled for about 15 min before the next operation. Schematic representation of the experimental setup

has been depicted in Fig. 1. Experiments with ultrasonic horn were performed with 50 ml of the solution. A preliminary experiment was also performed with 500 ml of the solution just to check the applicability of ultrasonic horn for large-scale operations.

#### 2.1.2. Side entering sonochemical reactor

The schematic representation of the experimental setup used for side entering sonochemical reactor with a capacity of 400 ml has been shown in Fig. 2. The reactor is agitated with standard turbine impeller for uniform mixing of the contents and irradiated with a single transducer operating at 20 kHz frequency of irradiation (Sonics and materials, USA) and the maximum power dissipation into the system is 120 W. To begin each sonochemical experiment, 400 ml of the stock solution was charged to the reactor. Then it was aerated for 20–25 min to saturate it with air keeping the temperature at 20 °C. Constant operating temperature was maintained by circulating coolant liquid through the outer jacket provided to the reactor. The sonication was continued for 2 h at a power output of 120 W. Samples were removed at regular intervals for analysis.

#### 2.1.3. Triple frequency hexagonal flow cell

The hexagonal triple frequency flow cell has a total capacity of 7.5 l and was operated in a batch mode (7 l of solution was used). The reactor operates at multiple frequencies of 20, 30 and 50 kHz individually or in combination (seven different modes of individual/combined operation are possible), with energy supply of 300 W per frequency of irradiation, i.e.

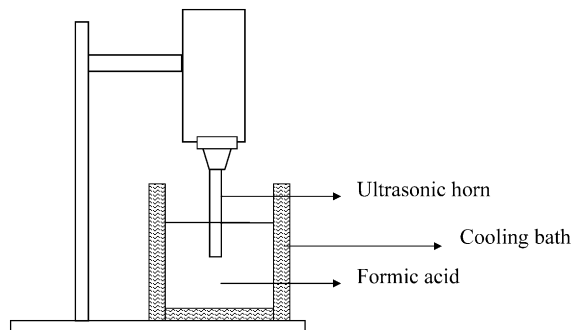
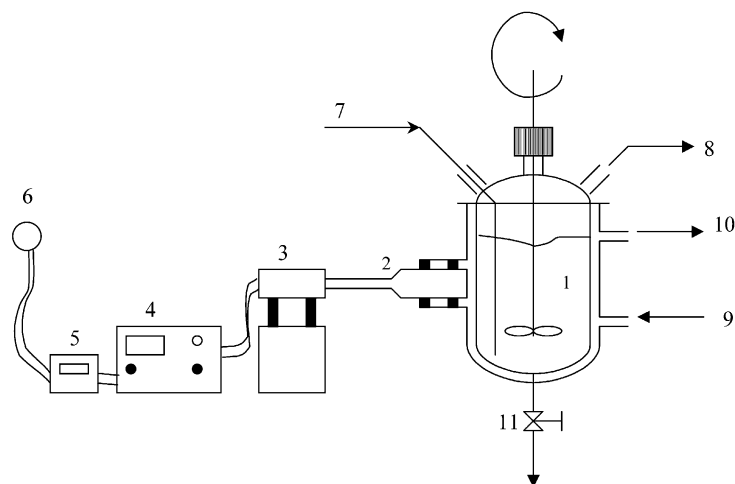


Fig. 1. Schematic representation of the experimental setup using ultrasonic horn.



1. Jacketed, agitated glass reactor with PTD impeller and baffles, 2. Ultrasonic probe, 3. Transducer (20kHz) with Housing, 4. Generator, 5. Wattmeter, 6. Mains supply, 7. Air supply, 8. Sample port, 9. Coolant in, 10. Coolant out, 11. Bottom valve

Fig. 2. Schematic representation of the experimental setup using side entering ultrasonic reactor.

maximum power dissipation is 900 W when all the three frequencies are operational. The detailed discussion about the construction of the reactor including a schematic representation has been presented in the earlier work [9,12].

## 2.2. Experimental methodology

Experiments were performed with varying initial concentrations of the pollutant (phenol in this case) in the range 100–500 ppm at constant temperature of  $30 \pm 1^\circ\text{C}$  with total irradiation time being 90 min unless specified otherwise. Some experiments were repeated in the presence of solid particles  $\text{TiO}_2$  (anatase grade powdered form with concentration of 300 ppm) to study the possible intensification due to an increase in the intensity of cavitation. As the aim of the work was to investigate the extent of possible intensification in the cavitation activity due to surface cavitation, only powdered form of  $\text{TiO}_2$  was used in the experiments (it offers maximum surface area for surface cavitation).

The amount of residual concentration of phenol in the solution was measured using spectrophotometric analysis (measuring the absorbance at wavelength of 270 nm using a UV Visible Spectrophotometer [Chemito, Mumbai, India]). Firstly, pre-calibrated

charts were prepared by measuring the absorbance of different aqueous solutions of phenol with known concentrations. Once the sample was withdrawn during the treatment, measurement of the absorbance of the sample yielded the actual concentration from the pre-calibrated charts.

In the case of experiments with the presence of solid particles ( $\text{TiO}_2$  particles), the solution was filtered a number of times to remove all the solid particles ( $\text{TiO}_2$  particles) before spectrophotometric measurements. It should be noted here that phenol concentration remains unchanged during the filtration operation.

Experiments for studying the adsorption characteristics of the pollutant on the solid particles were also performed as adsorption of pollutant interferes in the determination of concentration of the residual pollutant [13]. It was observed that the adsorption of phenol on the  $\text{TiO}_2$  particles was negligible.

## 3. Results and discussion

### 3.1. Conventional ultrasonic horn

#### 3.1.1. Effect of initial concentration

The variation in the percentage degradation of phenol with the initial concentration of the pollutant (in

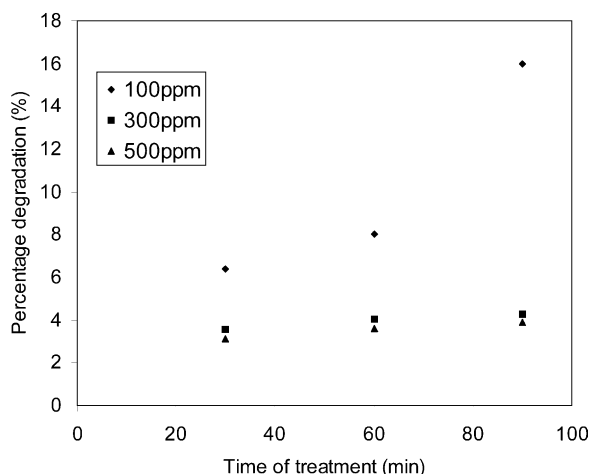


Fig. 3. Dependency of the percentage degradation of phenol on the initial concentration in the ultrasonic horn reactor.

the range 100–500 ppm) in the ultrasonic horn has been shown in Fig. 3. The percentage degradation is found to be inversely proportional to the initial concentration, a trend much similar to that observed in the literature for different pollutants [14–18]. It can be also seen from the figure that for lower concentrations used in the experiments, not much reduction in the rate of degradation has been observed even after 90 min of irradiation indicating that the cavitation is still functional to decrease the concentration of pollutant further. This is very important point to consider, as there may exist an equilibrium concentration of the pollutant beyond which ultrasonic irradiation will not be able to destroy the pollutant with significant rates of degradation (important considering the overall cost of the treatment scheme) and this concentration can be considered as the optimum concentration till which cavitation can be used as a pretreatment stage. It should be also noted that the trend of significant reduction in the rate of degradation with time was also not observed for the case of higher initial concentration of phenol, i.e. 300 and 500 ppm.

In the case of degradation of formic acid [10], significant reduction in the rate of degradation has been observed with the time of treatment (after 90 min of irradiation, the concentration of formic acid was found to be around 90 ppm with the rate of destruction nearly equal to zero) indicating the limiting concentration up to which cavitation can be successfully used. It can be

inferred from these results that the limiting concentration is dependent on the type of the pollutant (reactor operating conditions are exactly identical) and needs to be established by laboratory scale studies for the pollutant in question unless data is available in the literature with similarities in the operating conditions.

### 3.1.2. Scale up aspects

As it was observed that the maximum extent of percentage degradation for the ultrasonic horn was obtained with 100 ppm as the initial concentration, further studies for scale-up and process intensification using surface cavitation (discussed later in Section 3.5 devoted to process intensification) were carried out for this particular initial concentration of phenol. In the same experimental setup and under similar operating conditions, an experiment was performed with 500 ml aqueous solution of phenol so as to check the applicability of the ultrasonic horn for large-scale destruction. It has been observed that no degradation takes place even after 2 h of treatment time. This can be attributed to the fact that the zone of influence of the ultrasonic horn with cavitation intensities necessary to degrade the phenol molecule is restricted very near to the surface which is just a small volume fraction as compared with the capacity of the reactor (500 ml) and hence the cavitation intensity is not sufficient so as to produce enough number of free radicals so that destruction of phenol can take place in the given time of irradiation. It should be also noted that even though some destruction may be taking place in the vicinity of the transducer surface due to very high local cavitation intensity, its contribution to the overall degradation would not be felt due to poor mixing conditions existing in the reactor. Thus the conventional ultrasonic horn based on the principle of single transducer irradiation fails to induce any destruction at larger scale of operation and hence it can be said that the applicability is restricted only to laboratory scale operations.

### 3.2. Side entering sonochemical reactor

With an aim of evaluating the applicability of ultrasonic horn type reactors for large scale wastewater treatment applications, degradation of phenol was studied in the novel configuration as described earlier (initial concentration of phenol is 100 ppm, operating

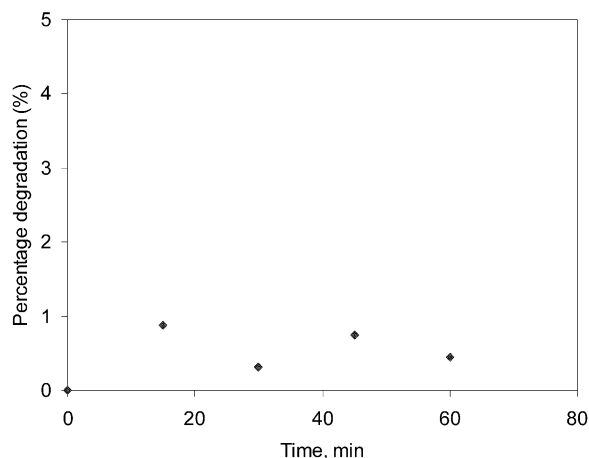


Fig. 4. Degradation of phenol in side entering sonochemical reactor with capacity of 400 ml.

temperature is 20°C). The reactor provides uniform mixing conditions due to continuous agitation as well as accurate maintenance of lower temperatures (degradation rate is inversely proportional to operating temperature [5]) and air saturation (aeration or dissolved gases also increases the rates of degradation [5]). The results obtained for studies in this reactor are shown in Fig. 4. It can be seen from the figure that the percentage degradation is mostly independent of the time of operation (slight deviation in the values of percentage degradation may be attributed to the experimental errors) and is always less than 1%. Thus it can be said that even after increasing the intensity of cavitation and achieving uniform mixing conditions as compared with the conventional configuration of ultrasonic horn, single transducer based reactors are not able to induce any significant destruction for the model pollutant considered in the present work. Since the reactor was not able to give any destruction for the lowest initial concentration of the pollutant, further studies with varying concentrations of phenol were not undertaken in this reactor.

Two important aspects need to be considered in the non-applicability of these reactors for large scale operation. Firstly, the zone of the maximum cavitation activity is always restricted very near to the transducer surface and even continuous recirculation of the liquid packets (by way of agitation) does not improve the destruction efficiency. Thus there is a need of using

multiple transducers for irradiation with increased areas of irradiation (this also helps in increased rates of acoustic streaming [19] and hence additional agitation may not be required for uniform mixing). Second aspect is that the intensity of cavitation produced by the given set of operating frequency of irradiation as well as power dissipation is not enough for achieving the destruction. The intensity of cavitation can very well be increased by increasing the frequency of irradiation as well as the power dissipation into the system [20,21] but the problems with operation using high frequency high power dissipation are well known [22,23]. An alternative way of increasing the intensity of cavitation is the use of multiple frequency irradiations where the individual operating frequencies may be lower in magnitude and differing with each other again by a small magnitude [24]. The novel reactor used in the present work considers multiple frequency irradiations (individual operating frequencies are 20, 30 and 50 kHz) using multiple transducers (3 × 6 transducers) located on each face of the hexagon. The design also enables the formation of standing waves, which are reported to enhance the intensity of cavitation and hence the degradation rates [25].

### 3.3. Triple frequency flow cell

#### 3.3.1. Effect of initial concentration

As the triple frequency flow cell was found to give appreciable destruction for the 100 ppm initial concentration of phenol (results shown in Fig. 5), the effect of initial concentration was studied in details by varying the concentration in the range of 100–500 ppm. Fig. 5 shows the variation of percentage degradation with the initial concentration of the pollutant in the flow cell operated at 20 kHz as the frequency of irradiation. It can be seen that the percentage degradation is inversely proportional to the initial concentration, a trend much similar to that observed in the experiments with the conventional reactor. It can be also seen from the figure that for the lower concentrations used in the experiments, not much reduction is observed in the rates of degradation even after 90 min of irradiation indicating that the cavitation intensity is again sufficient to decrease the concentration of the pollutant further. Similar trends were also observed for the remaining operating modes of irradiation, i.e. single frequency operation with 30 and 50 kHz as the frequency and

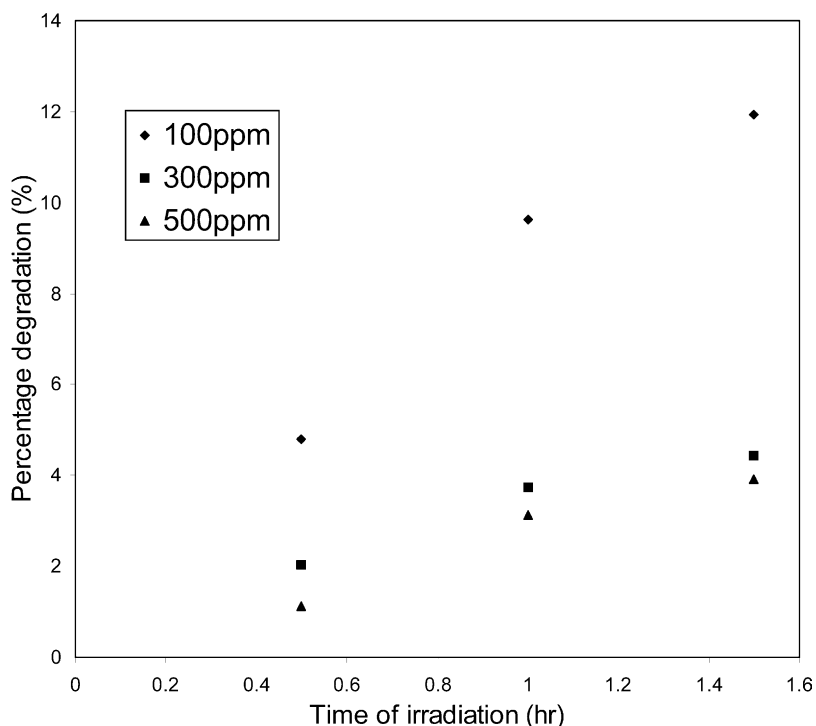


Fig. 5. Dependency of the percentage degradation of phenol on the initial concentration in triple frequency flow cell (operation at 20 kHz of frequency).

dual and triple frequency combinations. Thus it can be said that the mode of operation of the triple frequency flow cell does not affect the trends in the variation of the pollutant degradation rate with the operating parameters.

### 3.3.2. Effect of frequency of irradiation

The effect of the frequency of irradiation on the extent of degradation of phenol has been shown in Fig. 6. It can be seen from the figure that the extent of degradation increases with an increase in the frequency of irradiation for the single frequency operation. Further, the percentage degradation is more for the triple frequency operation as compared with dual and single frequency operations. The observed variation can be explained on the basis of the variation in the cavity collapse pressures and temperatures with the frequency of irradiation. In the earlier work [20,24], we have shown that at a constant intensity of irradiation and for a fixed initial radius of the nuclei, the magnitude of pressure pulse generated at the end of

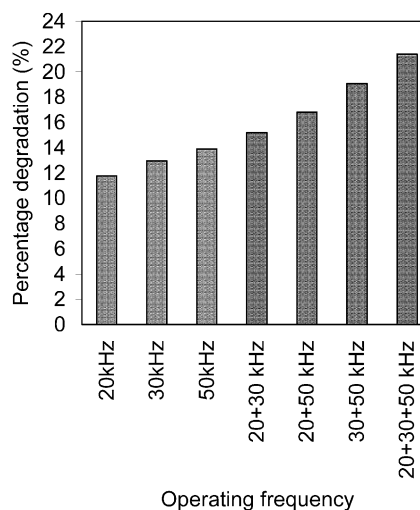


Fig. 6. Dependency of the percentage degradation of phenol on the operating frequency of irradiation in the triple frequency flow cell (90 min of treatment time and the initial concentration of phenol is 100 ppm).

collapse of a single cavity increases with an increase in the frequency of irradiation and also is more for the multiple frequency operation as compared with the single frequency operation. Moreover as the event of the collapse is adiabatic, increased collapse pressures, naturally increases the maximum temperatures reached in the cavity and also the number of free radicals generated at the end of cavitation phenomena are expected to be higher. Thus in accordance with the hot-spot theory as well as the free radical theory (majority of the sonochemical reactions are attributed to these two theories and the controlling mechanism of the reactions depends on the operating conditions and type of the pollutant which decides its location, i.e. in the cavity, on the surface of the cavity or in the liquid bulk), the rate of chemical reaction will be higher for the multiple frequency operation. A further confirmation of enhanced cavitation activity at higher/multiple frequencies of irradiation can also be obtained from the energy analysis presented in the earlier work [21]. Similar experimental results regarding the effect of frequency of irradiation can be found in the literature. Francony and Petrier [26] have shown that the degradation rate of carbon tetrachloride are higher at 500 kHz as compared with 20 kHz operation (single frequency operation) whereas Sivakumar et al. [5] have shown that the rate of sonochemical degradation of *p*-nitrophenol in a dual frequency rectangular flow cell is higher for the dual frequency operation as compared with the single frequency operation.

It is also imperative to check whether the multiple frequency operation is synergistic or not. The synergy is defined in terms of more than additive destruction, i.e. increase in the cavitation yield per unit power consumption. It can be easily seen from Fig. 5, that the triple and dual frequency operations are not synergistic for the present reaction as the magnitude of destruction for these operations are always less as compared with the additive values for the individual single frequency operation, e.g. the extent of degradation for the triple frequency operation is 21.4% whereas for individual 20, 30 and 50 kHz operations, the percentage degradation is 11.7, 13 and 13.9%, respectively, i.e. total additive destruction is equal to 38.6%. In the earlier work [9] destruction of two different pollutants, i.e. potassium iodide and Rhodamine B have been studied in the triple frequency flow cell. It was observed that the multiple frequency operation was

synergistic for the case of degradation of potassium iodide whereas for Rhodamine B degradation, individual frequency operation was found to be more suitable in terms of power dissipation analysis. Also for formic acid degradation, the use of multiple frequency irradiations gives more degradation as compared with individual frequencies but the operation is not synergistic [14]. Thus it appears that the multiple frequency operation is more suitable for reactions requiring lower intensity of cavitation whereas multiple transducer irradiation, with same or different frequency is more suited for reactions requiring higher intensities of cavitation (In this case, multiple frequency operation is not synergistic).

#### 3.4. Comparison of the novel reactor with conventional reactor

Cavitation yield was used to compare the efficacy of the two reactors (Conventional ultrasonic horn and Triple frequency flow cell; Side entering sonochemical reactor does not result in any degradation) in degrading phenol. Cavitation yield has been defined as:

$$\text{Cavitation yield} = \frac{\text{Moles of phenol degraded}}{\text{Power supplied into the system}} \quad (1)$$

Cavitation yield for the case of flow cell (all the conditions remaining same; initial concentration of phenol is 100 ppm, operating temperature of 30 °C, time of treatment is 90 min, nearly same operating frequency, viz. 20 kHz for the flow cell and 22 kHz for the ultrasonic horn) has been found to be  $2.786 \times 10^{-2}$  mol/W whereas for the ultrasonic horn, it is  $1 \times 10^{-3}$  mol/W. Thus, the triple frequency flow cell operating at large scale of capacity gives 25 times more cavitation yield as compared with the conventional reactor. Similar results have been obtained for the destruction of other pollutants as well (potassium iodide and Rhodamine B [9]; formic acid [14]). It should be noted that all these reactions are studied only for single contaminant model solutions and more work is indeed required for studying the efficacy of this reactor as well as the conventional reactors for real industrial effluents containing phenol, formic acid or any of the complex chemicals as the main constituents before firm conclusion for the applicability of the multiple frequency

multiple transducer reactors in actual practice can be made.

### 3.5. Process intensification using surface cavitation

Another critical aspect of the scale up of reactors is that the rates of degradation achieved in the laboratory/pilot scale and for simulated effluents may not be exactly replicated in the actual practice. Thus it is important to maximize the rates of degradation or in other words process intensification is another important aspect that needs to be considered for efficient scale up. Also the rates of degradation achieved in the novel large scale reactor are still less (though more efficient as compared with conventional reactors) and hence the treatment times will be substantially higher, possibly leading to an uneconomical operation. With this background, process intensification studies were carried out using solid particles.

In the case of flow cell, presence of solid particles (300 ppm powdered  $\text{TiO}_2$  particles) was indeed found to be beneficial similar to that observed for the degradation of formic acid in the earlier work [14]. The presence of solid particles affects the cavitation activity in two different and opposing ways; firstly it intensifies the process by providing additional nuclei due to the discontinuities in liquid medium and hence the number of cavitation events increase, but at the same time due to the scattering of incident sound waves, the net energy dissipation into the system decreases. In the case of triple frequency flow cell, due to multiple transducers the negative effect of scattering is not much felt and hence overall process intensification is observed (ca. 40% increase for 100 ppm initial concentration of phenol, 20 kHz frequency of irradiation and  $\text{TiO}_2$  loading of 300 ppm).

For the case of ultrasonic horn, presence of solid particles at concentration of 300 ppm was found to be detrimental resulting in lower extent of degradation as compared with that observed in the absence of solid particles. Thus in this case the scattering of the incident sound waves appears to be the predominant factor as compared with the enhancement in the number of cavitation events. It was decided to perform additional experiments to understand the mechanism of the effect of solid particles and to conclusively establish the hypothesis for the controlling effect of the presence of solid particles.

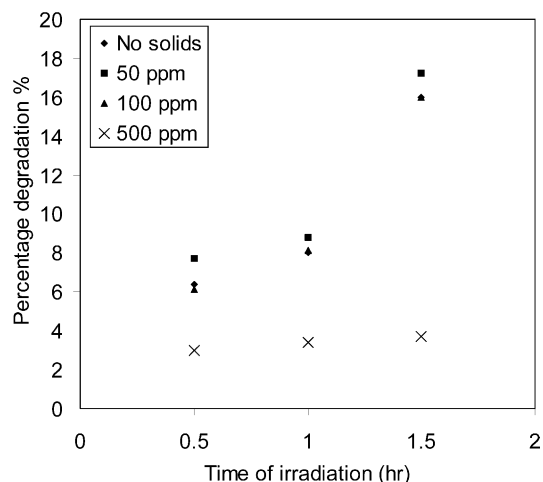


Fig. 7. Effect of concentration of solids on the extent of degradation of phenol (initial concentration of 100 ppm) in ultrasonic horn.

Fig. 7 gives the variation of percentage degradation with the time of treatment (for initial concentration of phenol as 100 ppm) for different concentrations of the solid particles. It can be seen that at very low concentration of solid particles, i.e. 50 ppm, the surface cavitation seem to dominate the negative effects of scattering of sound waves and hence a small increase in the percentage degradation is observed. As the concentration of the solid particles increases, the negative effect of sound scattering becomes dominating resulting in lowering of the extent of degradation. At 100 ppm concentration of the solid particles, the contribution of the two effects seem to be exactly identical resulting in an identical extent of degradation as compared with that observed in the absence of solid particles. At very high concentrations (500 ppm) of solids, the scattering of incident sound waves becomes so high that the extent of degradation is almost five times lower. Thus, these experiments with varying concentration of solid particles allow us to conclusively establish the two mechanisms by which the presence of solid particles is affecting the overall intensity of cavitation (defined as single cavity collapse pressure multiplied by the number of cavitation events) and hence the extents of degradation. As discussed earlier, in the case of flow cell, due to large volumes and multiple transducers, the scattering effect of the solid particles is much lower as compared with the generation of additional cavities and hence



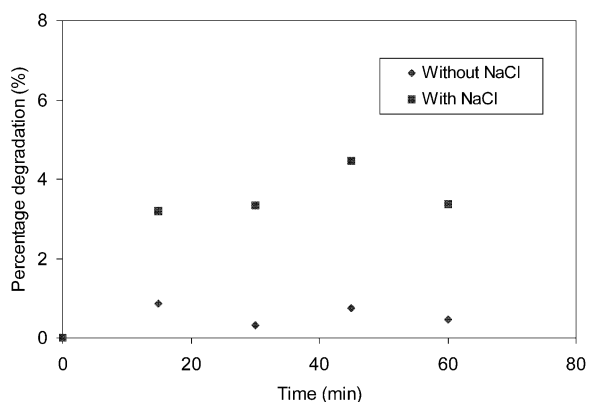


Fig. 8. Effect of presence of NaCl (8% by weight) on the degradation of phenol in ultrasonic horn.

intensification in the rate of degradation has been observed.

It should be noted here that though the presence of solid particles is not beneficial in the case of ultrasonic horn due to the scattering effects, presence of other constituents in dissolved state (scattering effect will not be there) altering the liquid phase physicochemical properties and providing additional nuclei for cavitation may result in the enhancement in the rates of degradation. To evaluate this idea, some preliminary experiments were carried out in the side entering ultrasonic horn using sodium chloride in dissolved state (8% NaCl loading was used at 100 ppm initial concentration of phenol). The results for this set of studies have been depicted in Fig. 8. It can be seen from the figure that the use of NaCl increases the rates of degradation by almost five times. Seymore and Gupta [27] have also reported similar results of enhancement in the rates of degradation of potassium iodide using sodium chloride (three times more degradation is observed due to the presence of NaCl indicating that the extent of the enhancement is also dependent on the type of the pollutant). The enhancement in the presence of NaCl can be attributed to an increase in the partitioning in the presence of NaCl leading to accumulation of the pollutant species at the cavitation implosion sites. In addition to the partitioning enhancement, presence of salt decreases the vapor pressure and increases the surface tension (this will help in promoting a more violent collapse of the cavitating bubble). Thus use of sodium chloride or any other salt such as  $\text{NaNO}_2$  altering the medium physicochemical

properties and the aqueous/organic partitioning of the target pollutant should result in process intensification. More work in this direction to establish the effect of the concentration and the type of the salts, type of the reactor is currently being done in the department.

#### 4. Conclusions

Ultrasonic horn results in the destruction of phenol at laboratory scale (lower initial concentration results in higher extent of degradation) whereas application at larger scale results in cavitation activity insufficient to produce any destruction. Both the configurations of ultrasonic horn as studied in the present work were found not to be suitable for the large scale applications. It appears that increased cavitation activity by use of multiple transducers with or without different frequencies of irradiation is the key factor in efficient scale up of sonochemical reactors.

The novel multiple transducer/multiple frequency reactor can be used effectively for the destruction of phenol at a large scale of operation. The cavitation yield obtained with this reactor is order of magnitude higher as compared with the conventional reactor and also the scale of operation is about 140 times more as compared with conventional ultrasonic horn. The extent of degradation has been found to increase with an increase in the frequency or by using combinations of frequencies, however, no synergism is observed in using combination of the frequencies for the reaction considered in the present work. It can be said that the relative rates of reaction between the generated free radicals and the pollutant and the recombination reaction of the free radicals control the efficacy of the multiple frequency operation and detailed kinetic analysis is a must for the effluent stream in question.

The exact mechanism of process intensification in the degradation of phenol (another critical aspect of scale up and successful application of sonochemical reactors for wastewater treatment) and its dependency on the type of the reactor has been conclusively established in the present work. In the case of ultrasonic horn, addition of solid particles as a source of additional nuclei results in detrimental effects, possibly due to single transducer operation coupled with lower capacity of the reactor resulting into large scale scattering of the incident sound waves. On the other

hand, triple frequency flow cell gives expected intensification of the degradation process due to multiple transducer operation and higher capacity. Use of dissolved salts has been found to result in substantial enhancement in the rates of degradation though detailed investigation of the effect with studies using different salts and concentration of salt is recommended before any firm conclusion can be drawn.

The triple frequency flow cell used in the present work is one of the very few illustrations depicting the successful use of sonochemical reactors for wastewater treatment applications at a pilot scale and process intensification can be efficiently carried out in this reactor as compared with the conventional reactors. Thus, it can be said that the design of large scale reactors should be based on the principle of multiple transducer/multiple frequency irradiation with wider areas of irradiation for the transducers influencing larger volumes of the pollutant streams directly.

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