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Theoretical prediction of cavitational activity distribution in sonochemical reactors

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ABSTRACT

Cavitational activity distribution is usually non-uniform in the sonochemical reactor and this hampers the successful design and operation of large scale sonochemical reactors. Theoretical prediction of the cavitational activity can aid in easy optimization of geometry and operating parameters as it replaces use of rigorous and expensive experimental mapping techniques for understanding the cavitational activity distribution. In the present work, an attempt has been made to predict the cavitational activity in terms of pressure field distribution by solving the wave equation in two different geometries of sonochemical reactors. Numerical simulations have been carried out by using Comsol Multiphysics software. The results are also compared with the experimental investigations reported in the earlier literature illustrations. It has been observed that cavitational activity in the case of ultrasonic horn is concentrated only near the transducer surface and is much more non-uniform as compared to the ultrasonic bath reactor with large longitudinally vibrating transducer. Comparison with experimental results has clearly established the correctness of the theoretical simulations in predicting the cavitational activity in the sonochemical reactors and hence its importance in possible scale up and optimization strategies for large scale operation.

1. Introduction

Use of ultrasound as an effective tool to intensify the rates of physical and chemical processing has been successfully harnessed over the last few years for a variety of applications [1–8] such as chemical synthesis (in homogenous and heterogeneous reaction system), waste water treatment (degradation of complex chemicals), biotechnology (cell disruption), atomization, extraction, leaching, crystallization, initiation of polymerization reaction and digestion of cellulose. Enhancement in the processing rates is mainly due to the fact that, when ultrasound is passed though liquid medium it can generate cavitation phenomena due to alternate compression and rarefaction cycles. The main effects of cavitation phenomena include generation of very high temperature (few thousand Kelvin) and pressure (few hundred atmospheres) pulse locally. In addition, cavitation also results in local turbulence, intense circulation currents (acoustic streaming), high shear stress in the vicinity of the bubble and formation of micro-jet near the solid surface depending on the specific application.

A careful analysis of the literature [2] however indicates that, inspite of having immense potential in intensification of majority of physical and chemical processing applications, very few applications are actually carried out on an industrial scale, probably due to the lack of design and scale up strategies and due to the fact that cavitational activity distribution in the reactor is not uniform. Understanding of the spatial distribution of cavitation activity in sonochemical reactor and possible elimination of the non-uniformity at large scale can lead to some success in application of sonochemical reactors at industrial scale [9]. The distribution of cavitational activity can be understood by measuring the primary effects (the effects which are generated directly due to the bubble collapse such as generation of temperature and pressure pulse or liquid circulation currents) and secondary effects (primary effects lead to the generation of the secondary effects such as chemical decomposition, aluminum foil erosion, and change in electrochemical properties). These experimental techniques, though used widely by researchers [10–14], are not always feasible due to the following reasons:

- Experimental techniques are usually quite expensive and time consuming.
- Cavitation medium gets disturbed due to the presence of external instrument such as thermocouple, hydrophone, aluminum foil,

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and test tube and hence we may not get a realistic picture of the cavitational activity distribution.

- Obtained results may not be reproduced due to the dynamic nature of cavitation phenomena.

Numerical prediction of cavitational activity distribution based on the fundamental bubble dynamics analysis avoids tedious experimentation and also can enable easy optimization of the geometry of reactor, location and geometry of the transducers and ultrasound operating parameters. Based on theoretical analysis, one can obtain the pressure field distribution in any new sonochemical reactor with different geometries and operating conditions, which can aid in optimization for maximum/uniform cavitational activity. The modeling studies can be extended to the quantification of other useful parameters such as distribution of temperature and mass transfer coefficient, which can be controlling parameters depending on the type of application. Such a detailed analysis can be used to identify the regions with maximum pressure fields in a large scale reactor and then may be small reactors can be placed strategically at these locations in order to get maximum benefits. It might happen that the threshold required for certain application is obtained at these locations but if considered globally these effects will be marginalized resulting into much lower yields from cavitation reactors. Thus the location of the transducers on the irradiating surface and the location of micro-reactors will also depend on the type of application, which decides the required cavitational intensity. It should be noted that one to one correspondence between the simulated pressure fields and experimental reactions must be established before reaching to any firm conclusions. This clearly establishes the requirement of a sound strategy based on the theoretical simulations for establishing efficient design and scale up strategies for large scale operation of sonochemical reactors. The present work aims at prediction of the cavitational activity in terms of pressure field distribution by solving the wave equation (Helmholtz equation, considering the effects of location and type of horn) by finite element method, in two different geometries of sonochemical reactors. Pressure field distribution is obtained using COMSOL Multiphysics version 3.3. The obtained results have been compared with earlier experimental investigations of Kanthale et al. [15] and Kumar et al. [16]. Kanthale et al. [15] investigated the cavitational activity distribution in an ultrasonic horn having diameter of 2 cm, maximum power rating as 240 W and driving frequency of 20 kHz, whereas the work of Kumar et al. [16] was based on an ultrasonic bath type reactor with dimensions as 33 cm × 20 cm × 15 cm, operating frequency of 36 kHz and maximum power rating of 150 W.

2. Model formulation

2.1. Three-dimensional wave equation

Wave equations describe the propagation of any waves, i.e. sound, light, etc. through the liquid medium. The sound wave equation is a second order partial differential equation. This equation generally describes the evolution of acoustic pressure p as a function of space r and time t. The propagation of sound wave in one dimension (x direction) can be described by the following equation as:

$$\frac{\partial^2 p}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2 p}{\partial t^2} = 0 \quad (1)$$

where p is acoustic pressure (N/m^2) and c is the speed of sound in medium (m/s). This equation can be solved by assuming constant speed of sound and expressing the acoustic pressure as a function of the ultrasound frequency as follows:

$$p = p_o \sin(\omega t \pm kx) \quad (2)$$

where ω is angular frequency and k is wave number.

Extending the same logic (Eq. (1)), the wave equation considering all the three dimensions in homogenous liquid medium can be written as:

$$\nabla^2 p \frac{1}{\rho} - \frac{1}{c^2 \rho}\frac{\partial^2 p}{\partial t^2} = 0 \quad (3)$$

where $\nabla^2 = \frac{\partial^2 p}{\partial x^2} + \frac{\partial^2 p}{\partial y^2} + \frac{\partial^2 p}{\partial z^2}$

Following assumptions have been made for specific application of Eq. (3) to the present case of propagation of ultrasound wave in the liquid medium:

1. Linear propagation of sound wave through medium.
2. Shear stress is negligible.
3. Density and compressibility of liquid medium are constant.
4. Pressure is time harmonic, i.e. $p(r,t) = p(r)e^{i\omega t}$.

Based on these assumptions, we obtain the following equation, which is described as Helmholtz equation:

$$\frac{\nabla^2 p}{\rho} - \frac{\omega^2}{\rho c^2} p = 0 \quad (4)$$

By applying suitable boundary conditions, the Helmholtz equation has been solved by using a three-dimensional acoustic module in the COMSOL Multiphysics software, which considers the finite element method.

2.2. Consideration of damping effects

In the case of sonochemical reactors, during the propagation of ultrasonic wave through a bulk of liquid away from the transducer, the intensity of wave decreases. This is described as the damping effect. The damping of sound wave occurs due to the reflection, refraction and scattering of the incident sound wave. Eq. (4) has been modified by considering the damping effect (by virtue of modification of density and speed) as follows:

$$\frac{\nabla^2 p}{\rho_c} - \frac{1}{\rho_c c_c^2}\frac{\partial^2 p}{\partial t^2} + d_a \frac{\partial p}{\partial t} = 0. \quad (5)$$

where ρ_c and c_c are the complex density and complex speed respectively (in the case of no damping, $\rho_c = \rho$ and $c_c = c$).

The parameters ρ_c and c_c have been considered to be dependent on the complex wave number (k_c) and complex impedance (z_c) as follows:

$$\rho_c = \frac{z_c k_c}{\omega}; \quad c_c = \frac{\omega}{k_c}$$

The complex wave number and complex impedance have been estimated in terms of the bulk viscosity of liquid medium (μ) as follows:

$$k_c = \frac{\omega}{c}\frac{1}{\sqrt{1 + (i\omega\mu/\rho c^2)}}, \quad z_c = \rho c \frac{1}{\sqrt{1 + (i\omega\mu/\rho c^2)}}$$

It should be noted here that ultrasonic waves have very complex behavior with reflection, attenuation and amplification of different wavelengths. In the present work, we have tried to consider some degree of complexity (with emphasis on the cavitation events) by considering the damping factor and its dependence on the frequency of irradiation, intensity of irradiation, speed of sound and

Table 1
Different types of geometry simulated with total number of degree of freedom, total number of element, dimension and frequency of ultrasound.

Sr no.	Type of geometry	Dimension of bath (m)	Frequency of ultrasound (kHz)	Dimension of horn (m)	Degree of freedom	Number of elements of mesh	Reference
1	Vertical horn	$D = 0.135$, $H = 0.175$	20	$d = 0.02$, $h = 0.02$	150,164	106,309	Kanthale et al. [15]
2	Bath with longitudinal horn	$L = 0.33$, $B = 0.2$, $H = 0.15$	36	$d = 0.03$, $l = 0.27$	64,837	45,310	Kumar et al. [16]

density of the liquid medium (parameters which are important considering the growth and collapse of the generated cavities which finally affects the collapse intensity). In the simulations, temperature has not been considered as a variable as it has negligible effect on damping. The same is true for the wavelength—it was water medium constantly with specified constant acoustic pressure. For the range of intensities considered in the present work, damping is also not dependent on the generated cavitation events. Generation of cavities might interfere in the passage of ultrasonic energy due to shielding effect but this is true at very high acoustic intensities.

2.3. Boundary conditions

While simulating the different geometries, following boundary conditions have been applied:

1. At the tip of transducer, we consider $p = p_o$ (entire ultrasound energy is entering into the reactor through the tip of transducer). For time harmonic analysis, p_o is the initial amplitude of the harmonic source.
2. At the wall of reactor, we consider, $p = 0$ (pressure amplitude vanishes near the wall).

The consideration of boundary condition as $p = 0$ at the wall is justified considering the fact that at the wall of the reactor, the normal component of the acceleration is zero, which means that pressure amplitude vanishes near the wall and corresponds to a total reflection of ultrasound: $p = 0$, hence $-n(1/\rho_0)(\nabla p - q) = 0$, which is true only when the reactor wall thickness is negligible compared to wavelength. For ultrasonic operations where frequency is 20 kHz or 36 kHz, this is indeed true and it is not dependent on the material of the reactor wall.

2.4. Grid generation

Finite element method requires breaking of the computational domain (i.e. entire reactor area) into simple geometry such as triangle and tetrahedrons elements, which are described as mesh/grids. This breaking can be achieved by using mesh generation tool to have either structured mesh or an unstructured mesh. In the present work, unstructured mesh has been used, because in structured mesh generation, the average resolution of mesh differs between direction parallel to the grid line and it requires direction of propagation of wave which is difficult to be characterized.

Table 1 gives the different types of geometries simulated in the present work with total number of degrees of freedom, total number of grid elements, dimension of reactors and frequency of ultrasound [15,16].

2.5. Parametric sensitivity analysis

Parametric sensitivity analysis yields information about the controlling parameters in deciding the final predicted values. The obtained results were also analyzed using statistical analysis methodologies for establishing the controlling operating parameters. The analysis was done using Sigma Stat version 3.5 software supplied by Cranes Software International Limited. Statistical analysis of the obtained variations for damping effects considered in terms of frequency and intensity of irradiation, density of liquid medium, velocity of sound in liquid, etc. and total wall reflection by considering pressure at the wall is equal to zero, was done in order to establish the contributing independent variables to the dependent variable as the cavitational activity. It was observed that both the factors contributed equally to the overall cavitational activity and passed the statistical tests.

3. Results and discussion

3.1. Predication of cavitational activity in ultrasonic horn

Ultrasonic horn with diameter as 2 cm, maximum power rating 240 W and driving frequency of 20 kHz has been considered to be immersed 2 cm below the liquid level in a cylindrical beaker with a diameter of 13.5 cm and height of 17.5 cm as used in the earlier experimental mapping work of Kanthale et al. [15]. The cavitational activity distribution has been predicted based on the model simulations and the complete profile for distribution of pressure field in the sonochemical reactor has been depicted in Fig. 1. It can be easily seen from the figure that active zone with maximum pressure field is concentrated near to the transducer surface and majority of the reactor zone shows minimal activity.

Fig. 2 shows the variation of the cavitational activity with vertical distance away from the transducer surface along the center of the reactor/horn. It can be again seen from the figure that as the distance from the horn tip increases, intensity of cavitational activity in terms of predicted sound pressure decreases. Maximum pressure field intensity (4.2×10^5 Pa) can be found in the region up to 0.02 m distance from the ultrasonic horn and then it decreases exponentially. Also the pressure field shows some intermediate peaks, i.e. marginal increase in the intensity, with the distance between two peaks approximately equal to half the distance of wavelength ($\lambda/2$). The results are in accordance with the earlier experimental measurements done in the same reactor [15,17,18].

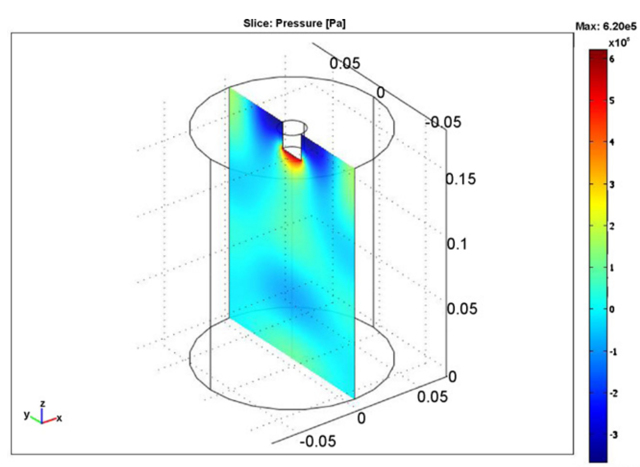

Fig. 1. The pressure field distribution in vertical horn (driving frequency $f = 20$ kHz) type of reactor of dimension (D) = 13.5 m, height (H) = 17.5 m and horn with diameter (d) = 2 cm, height (h) = 2 cm.

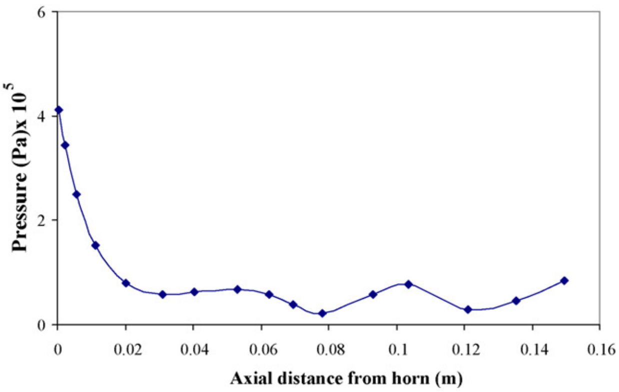

Fig. 2. Variation of pressure amplitude in the axial direction of ultrasonic horn.

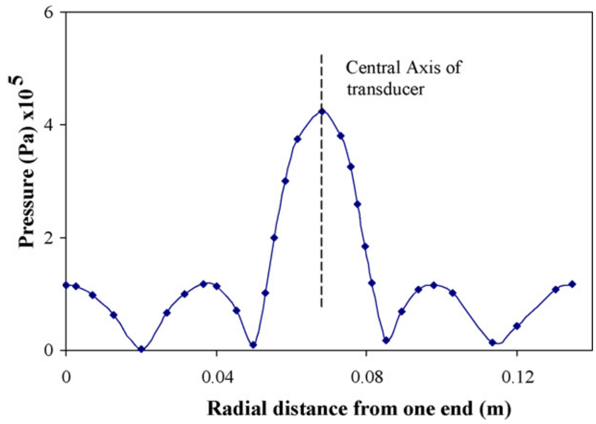

Fig. 3. Variation of pressure amplitude in the radial direction at height $z = 0.15$ m from bottom of the reactor.

Kanthale et al. [15] carried out experimental investigation to determine cavitational activity in terms of local pressure amplitude with the help of hydrophone (Bruel and Kjaer 8103) and by carrying Weissler reaction at different locations. It has been reported that as the distance from horn increases cavitational activity decreases exponentially and some peaks (slight increase in the activity) are observed at distances of 0.045 m, 0.08 m and 0.12 m with the difference between 2 consecutive peaks being equal to half the wavelength. This intermediate increase in the cavitational activity is due to the pressure generated by sub-harmonically oscillating bubbles at a frequency corresponding to multiples of the driving frequency. It should be also noted here that though the trends in variation of cavitational activity are exactly matching, the absolute values differ by about 30%. This can be attributed possibly to effects of changing temperature in experimental investigations and possible presence of dissolved gases which have not been considered in the model.

Romdhane et al. [17] have carried out thermometric investigation in an ultrasonic horn reactor with two different operating frequencies of 20 kHz and 40 kHz and maximum power rating of 160 W. The reactor has dimensions of 0.32 m × 0.23 m × 0.20 m and the height of immersion of ultrasonic horn in the liquid level was 4 cm and 2 cm for the frequency 20 kHz and 40 kHz respectively. Since the present work was carried out only for 20 kHz frequency, we have considered results obtained using 20 kHz frequency horn for comparison purpose. The results indicate that cavitational activity in the reactor decreases exponentially up to 2 cm from emitter surface along the central axis with some intermediate peaks at distances 3 cm and 8 cm, very similar to that obtained in the present work theoretically.

Simulations were also performed along radial direction at a fixed axial distance to check the radial distribution of the cavitational activity. The obtained results have been shown in Fig. 3. It is observed that the cavitational activity measured in terms of local pressure decreases with an increase in the distance away from the central axis towards the wall. The maximum pressure intensity (4.2×10^5 Pa) is found to be very near to the horn at a distance $y = 0.0675$ m from side of reactor at a height of 0.02 m from the transducer surface. An increase in the activity near to the wall starting from a distance of 0.04 m from the central axis is observed possibly due to the reflection of sound wave at the wall. The magnitude of intensity is 1.2×10^5 Pa. The obtained results are again in accordance with the experimental investigations obtained by Kanthale et al. [15]. It has been reported that the cavitational activity decreases in a distance away from central axis (with maximum pressure amplitude of 6×10^5 Pa) up to 0.02 m and then remains constant 2×10^5 Pa near wall.

3.2. Simulation of ultrasonic bath with longitudinal vibrating transducer

Simulations have also been performed for ultrasonic bath type reactor with longitudinally vibrating transducer at the bottom. The reactor has dimensions of 0.33 m × 0.2 m × 0.15 m and has a driving frequency of 36 kHz and maximum power rating of 150 W. Kumar et al. [16] have investigated the cavitational activity distribution using a hydrophone for the measurement of pressure field. In the current work, simulations were performed considering an active volume equal to 7 L corresponding to 90% capacity similar to that used in the earlier experimental investigations. Fig. 4 shows the cavitational activity distribution in the ultrasonic bath reactor with longitudinally vibrating transducer at one cross-section. Comparing Fig. 4 with Fig. 1, it can be said that there are large number of active zones in the reactor as compared to the ultrasonic horn reactor.

The variation in the cavitational activity along the length of the horn at the height of $z = 0.095$ m from bottom of reactor has been shown in Fig. 5 whereas the variation along the height (just above the horn and in the direction away from the horn axis in perpendicular direction) has been shown in Fig. 6. It can be seen from Fig. 5 that the cavitational activity (1×10^5 Pa) decreases as distance increases up to $x = 0.04$ m (along the length) and then increases up to the length $x = 0.07$ m (wavelength of ultrasound = 0.041 m) and then decreases and reaches the approximately same value at distances $x = 0.13$ m, $x = 0.23$ m, etc. Maximum activity (2×10^5 Pa)

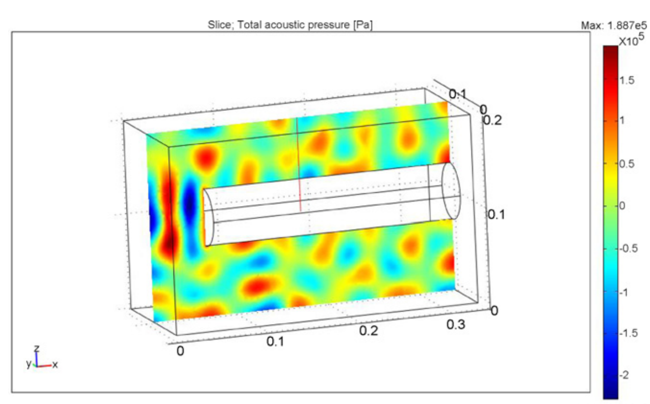

Fig. 4. The pressure field distribution in ultrasonic bath (dimensions as length (L) = 0.33 m, width (w) = 0.20 m height (h) = 0.15 m) equipped with longitudinally vibrating transducer (driving frequency $f = 36$ kHz, transducer dimensions as diameter (d) = 0.03 m, length (l) = 0.27 m).

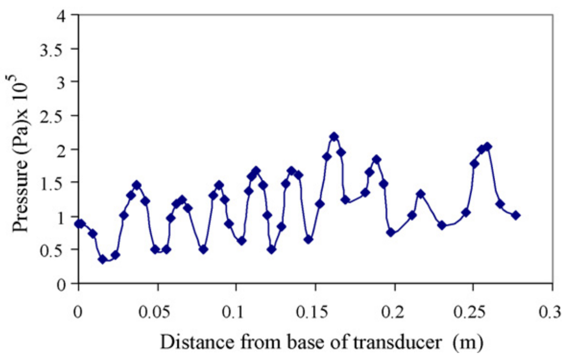

Fig. 5. Variation of pressure amplitude in the direction of transducer at height $z = 0.095$ m from the bottom of the reactor.

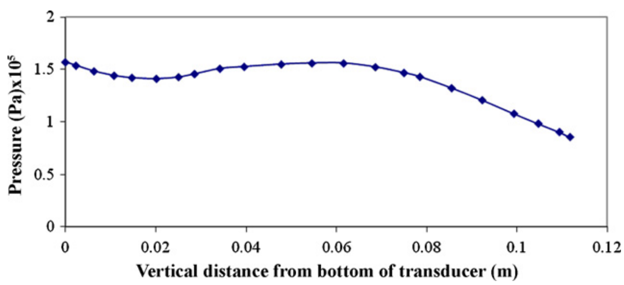

Fig. 6. Variation of pressure amplitude in the vertical direction away from bottom of the transducer .

is observed at distance 0.16 m from base. Some variation in cavitational activity is due to the interferences and formation of stationary sound wave in the bulk of liquid. The observed results are in well accordance with the experimental results obtained by Kumar et al. [16]. Fig. 6 shows the variation of cavitational activity along $z = 0.069$ m to $z = 0.108$ m (perpendicular to the direction of horn) at the center of reactor (0.165, 0.1). It can be observed that the activity is mostly constant till a height of about 0.07 m (around 1.5×10^5 Pa) beyond which a decrease with an increase in the distance has been observed.

3.3. *Comparison of the cavitational activity distribution in two reactors:*

In the case of sonochemical reactors, the cavitational activity distribution is mostly non-uniform and the extent of non-uniformity might be varying depending on the arrangement of the transducers in the reactor. Out of the two reactors considered in the present work, one is a conventional ultrasonic horn whereas the second one is an ultrasonic bath reactor with longitudinally vibrating transducer. Comparison of the cavitational activity distribution predicted using the theoretical analysis in the present work indicates that cavitational activity is concentrated only over a very small location in the ultrasonic horn (less than 10% volume), whereas in the case of ultrasonic bath it is much better with number of active zones showing some cavitational activity. It should be also noted that the maximum local cavitational activity in the case of ultrasonic horn is more as compared to the ultrasonic bath possibly due to much higher power dissipation per unit volume. It should also be noted here that this may not be able to give better results as finally the cavitational activity over the entire volume of the reactor is important in deciding the cavitational efficacy. Bhirud et al. [18] have investigated the degradation of formic acid in both the reactors and reported that ultrasonic bath gives much higher cavitational effects as compared to horn. The extent of degradation per unit power dissipation was order of magnitude higher in ultrasonic bath (0.077 mol/W) as compared to ultrasonic horn (0.0005 mol/W). These experimental results again confirm the correctness of the trends obtained in the present work using theoretical analysis.

4. Conclusions

The present study has enabled us to conclusively establish that cavitational activity can be predicted with the help of theoretical simulations. The results obtained in the present work have been validated with the earlier experimental results obtained in the same reactor and there exists a close matching between the trends. Following useful design related information can be obtained based on the simulations:

1. The ultrasonic activity is maximum in the zone very near to the irradiating surface and drastically decreases, away from the source both in axial as well as in radial direction. In the case of ultrasonic horn, the active zone of cavitational activity is much lower and concentrated only near the transducer surface.
2. Ultrasonic bath with longitudinally vibrating transducer at the bottom also shows similar variations of decreasing cavitational activity but the extent of non-uniformity is much lower as compared to the ultrasonic horn possibly attributed to higher irradiating surface.
3. The magnitude of the cavitational intensity is not the only factor in deciding the efficacy of the cavitational reactors but also the cavitational activity distribution in the reactor is equally important.

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