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Sonochemical reactors: Important design and scale up considerations with a special emphasis on heterogeneous systems

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ABSTRACT

The spectacular effects observed during acoustic cavitation phenomena have been successfully employed for a number of applications on laboratory scale of operation but a well defined design and scale up methodology is lacking. The present work aims at developing a unified approach for the selection of different operating and geometric parameters for large scale sonochemical reactors with a special emphasis on heterogeneous systems. In the case of heterogeneous systems, apart from optimum selection of operating and geometric parameters, it is also important to understand the mixing and hydrodynamic characteristics due to the presence of solid/gas phases in the liquid medium. Also the quantification of attenuation of the incident sound energy has been discussed, which can be important design consideration in heterogeneous systems. Recommendations have been made for optimum selection of frequency of irradiation and power dissipation rate/irradiation intensity as well as the liquid phase physicochemical properties for the given physicochemical transformation. The discussion also highlights' the recent advances in development of sonochemical reactors focusing on reactor geometry and location of transducers in batch and continuous scale of operation.

1. Introduction

The spectacular effects of cavitation phenomena, viz. very high temperature and pressure locally, strong acoustic streaming, high shear stress near the bubble wall, microjets near the solid surface due to asymmetric collapse of bubble and turbulence, has been successfully harnessed for numerous applications such as chemical synthesis (in homogenous and heterogeneous systems), waste water treatment, biotechnology, polymer chemistry, etc. [1–4]. Mason [1,2] has reviewed the applications of cavitation phenomena in chemical and allied fields such as green chemistry, electrochemistry, nanotechnology, polymer chemistry, food industry, etc. and also highlighted the important issues restricting the successful large scale operation of sonochemical reactors. Adewuyi [3] has presented an excellent review on the applications of sonochemical reactors in the environmental sciences whereas Rohkina et al. [4] have presented a state of the art about applications of sonochemical reactors in biotechnology. In spite of possible diverse field applications, it should be noted that there are hardly any physicochemical transformations carried out on industrial scale of operation, owing to the lack of unified design and scale up strategies. The problems are more severe for heterogeneous systems as the presence of second phase in terms of external gas or solids leads to additional resistances in terms of mixing or mass transfer and also can result in significant attenuation of the incident sound energy leading to lower energy dissipation rates as compared to the design values. Currently, there are very few illustrations of reactor configurations operating at commercial scales of operation [5–8]. Son et al. [5] have reported large scale rectangular type of sonochemical reactor (dimensions 1.2 m × 0.6 m × 0.4 m) of working volume of 250 L with transducer module consisting of nine transducers located at the sidewall. The transducers have variable frequencies in the range of 35–170 kHz and fixed maximum power rating of 400 W per transducer. The performance of reactor was evaluated by quantifying the variation in power dissipation, sonochemical efficiency and damping factor for water and potassium iodide solution. Asakura et al. [6] have also reported the application of a large scale rectangular sonochemical reactor (0.508 m × 0.508 m × 0.672 m) with maximum operating capacity of 112 L. The design is based on the use of twelve transducers (six in each module of irradiating frequency as 500 kHz and maximum power rating of 620 W) located at the bottom or sidewall. The performance was quantified by analysing the variation in the cavitational activity with height by means of chemical dosimetry. Despite the large scale operation, it should be noted that efficacy of these reactors has not yet been checked for any industrially important transformations. Vinatoru [7] have reported an application of sonochemical reactor of operating capacity of 750–800 L for extraction of active ingredients from herbs using different solvents. Horst

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et al. [8] have illustrated a design of sonochemical reactor with concentrator horn of frequency 20 kHz and intensity of irradiation in the range of 14–160 W/cm^2 and its application for heterogeneous solid–fluid reactions at annual operating capacity of 4 tonne. It has been shown that the concept of a conical funnel for the transducers fits the demand for perfect radiation effectiveness and a good reaction management.

The major problems identified in the effective design of large scale reactors are local existence of cavitation events very near to the irradiating surface, wide variation of the energy dissipation rates in the bulk volume of the reactor coupled with the inability of the existing tools to accurately predict the cavitating zones in the reactor and link it with the observed chemical or physical effects of cavitation, erosion of the sonicator surfaces at the high power intensities required for industrial scale operations and lack of robust design and scale up strategy establishing the importance of different aspects such as hydrodynamics, mixing and mass transfer which control the effectiveness in the physical/chemical processing applications. The problems related to cavitational activity distribution in the sonochemical reactors and its quantification can be addressed by different mapping techniques [9,10], which can be either experimental or based on theoretical bubble dynamics simulations. The available techniques have been reviewed by Sutkar and Gogate [9] highlighting the basic aspects of each technique, their applicability and merits/demerits and the presented analysis has allowed to quantify the behaviour of cavitational activity in reactor geometry over a range of operating parameters. Gogate et al. [10] have also tried to develop correlations to link the observed cavitational effects to the operating parameters using the bubble dynamics analysis. The correlations were aimed at quantifying the cavity collapse pressure or temperature as a function of operating parameters (frequency, intensity and initial radius of the nuclei). However, applicability of these equations poses crucial limitation due to the fact that these correlations were based considering dynamics of a single cavity (in reality dynamics of single cavity is influenced by the presence of other cavities) and requires the magnitude of actual radius of bubbles generated during cavitation event, which is difficult to compute. The problem of single cavity dynamics was rectified by study of Kanthale et al. [11], who considered cavity cluster (group of cavities) and its dependence on the different operating parameters.

In the present work, an attempt has been made to provide unified selection criteria for important operating parameters for any possible physicochemical transformation by effectively analyzing the existing data available in the literature. In addition, we have discussed some difficulties in calorimetric power measurements and a modified power measurement equation has been proposed. The present work also aims at addressing the hydrodynamic behaviour of two commonly used sonochemical reactor configurations and the dependency of the mixing time, mass transfer coefficient, attenuation coefficient on the operational parameters. Also the recent advances in development of sonochemical reactor especially in terms of novel geometry, scale of operation and type of operation, viz. either batch or continuous, have been highlighted.

2. Selection of operating parameters

In the case of sonochemical reactors, two aspects of cavity dynamics, i.e. the maximum size reached by the cavity before a violent collapse (dictates the cavitational intensity) and the life of the cavity (decides the active cavitational volume), are of prime importance. The aim of the equipment designer should be to maximize both these quantities by suitably adjusting the different operating/geometric parameters.

2.1. Selection of frequency of irradiation

Selection of irradiation frequency for any physicochemical transformation predominantly depends on the desired effects viz. physical or chemical. For instance, low frequency (in the range of 10–100 kHz) operation should be employed where intense physical effects are required in applications such as biotechnology (cell disruption), textile processing, polymer degradation and extraction (solid–liquid), etc. High frequency operation (in the range of few hundred to thousand kHz) should be opted for degradation of various compounds in wastewater treatment and chemical synthesis. This selection criterion is fixed for any scale of operation and for single frequency operation. However, in literature, multi-frequency operation (combination of same or different frequency) has been reported [12–17] to enhance the overall cavitational activity and generating intensities suitable for chemical processing applications at higher energy efficiencies. Servant et al. [12] have reported that in the case of dual frequency operation, cavitation bubble volume fractions are higher as compared to that observed in single frequency sonochemical reactors. It has also been reported that the cavitation medium is intensely disturbed due to the combination of frequencies resulting in overall higher cavitational activity due to generation of more cavities and stronger bubble–bubble, bubble–sound field interaction due to primary and secondary Bjerkens forces. In another theoretical investigation, Tatake and Pandit [15] have performed the modelling of dual frequency system with an objective of understanding the dependency of collapse pressure, temperature, time of collapse and extent of growth of bubble on the different combinations of frequencies. It has been reported that multiple frequency operation results in higher intensity of cavitational collapse. There have been some experimental investigations also based on the multiple frequency operation. Sivakumar et al. [13] have reported efficient application of dual frequency sonochemical reactors for the degradation of p-nitrophenol whereas Thoma et al. [16] have investigated the sonochemical destruction of dichloromethane and o-dichlorobenzene in aqueous solution using a near-field acoustic processor. Feng et al. [17] have reported better efficacy of dual frequency operation based on an experimental investigation of oxidation of KI in a rectangular type of reactor operating with frequency combination of 28 kHz with 0.75, 0.87, 1 and 1.4 MHz.

2.2. Selection of power rating

Another important factor affecting the cavitational effects is the amount of energy entering in the bulk of liquid as well as the area of transducers used for the said transfer of energy. Bubble dynamics studies indicate that the bubble distribution in terms of size, number of bubbles, maximum life time and collapse pressure are complex function of power dissipation rate. Also, the extent of temperature rise in bulk of liquid is a function of rate of power dissipation, which ultimately leads in altering gas solubility and vapor pressure affecting the ease of generation of cavitational events as well as final collapse intensity. In development of continuous scale sonochemical reactors, it is important to have the knowledge of hydrodynamic behaviour including mixing time, which critically depends on the power density. Thus, it is important to quantify the actual power dissipated in the bulk of liquid and hence available for the generation of cavitating conditions. The exact power input also helps in optimizing operating cost for a given physicochemical transformation.

In sonochemical reactors, the amount of energy dissipated into liquid is often expressed in terms of energy efficiency, which is a ratio of the energy dissipated in the system to the supplied electrical energy. Energy dissipated in laboratory scale reactor is generally calculated by measuring the change in temperature of liquid with time though there are some additional methods based on the use of acoustic impedance (Z) as a measurement parameter. Margulis and Margulis [18] have described several such methods including measuring the acoustic amplitude of transducer by vibrometer, by using piezoelectric or magnetorestrictive sensitive elements, thermoelectric method and optical measurements (interference and semi-shady methods) which effectively measures the refraction index. It should be noted that, all these reported methods pose serious limitation due to the fact that the sonochemical reactor involves a non-homogeneous liquid medium, owing to uneven distribution of bubbles in the bulk of liquid. This clearly indicates that physicochemical properties of bulk liquid especially density (ρ) and speed of sound (C) change resulting in a decrease in the acoustic resistance or impedance ($Z = \rho C$). This limitation in the measurement methods can be overcome by the calorimetric method.

The calorimetric method is based on the first law of thermodynamics, i.e. one form of energy gets converted into other form. In sonochemical reactors, absorbed energy is usually transformed into heat, which brings about a temperature change in the liquid medium, which can be accurately measured. For more accurate results, it might be a good idea to take into account the part of heat utilized by reactor wall and transducers, though neglecting this will result in about 5% errors in the overall measurements. Another important factor which needs to be considered is the energy loss to ambient air due to convective mode of heat transfer. In laboratory scale operation with operating volume in the range of few mL, convective and conductive heat transfer hold little significance, however in a larger scale operation with higher reaction volume and higher contact surface (liquid–wall, liquid–air), these losses may be a significant fraction of the overall energy dissipation. Thus, deliberation of these factors gives more realistic estimation of the energy dissipated on large scale operation and modified equation for calorimetric power measurement takes the form as:

$$\begin{aligned}&\text{Calorimetric power dissipation}\\&\quad = (\text{Energy utilised to raise the temperature of bulk liquid})\\&\quad + (\text{Energy absorbed by reactor wall and transducers})\\&\quad + (\text{Energy lost to ambient air by convection}) \qquad (1)\end{aligned}$$

Mathematically it can be written as:

$$P_{cal} = (mC_p\,\Delta T)_{liquid} + (m_iC_{pi}\,\Delta T)_{inner\ reactor\ wall} + (hA\,\Delta T) \qquad (2)$$

where m is mass of liquid (kg), m_i is the mass of reactor/transducers (kg), C_p is specific heat of liquid at constant pressure (J/kg K), C_{pi} is the specific heat of material of reactor (J/kg K), ΔT is change in temperature (K), h is convective heat transfer coefficient (W/m^2 K) and A is the area of heat transfer (m^2).

In majority of the cases, sonochemical reactor is made up of low thermal conductivity materials specifically teflon, polyurethane, etc. where energy absorbed by wall can be neglected owing to the complexity involved in wall temperature measurement.

Once the measurement of calorimetric power has been achieved successfully, it is important to optimize the power input by considering the effect of reaction volume or height of liquid medium in the reactor. Koda et al. [19] have analyzed the variation in the cavitational activity with power dissipation by quantifying the sonochemical efficiency in various laboratory scale sonochemical reactors of operating volume 50 cm^3 and power in the range of 35–220 W with irradiation frequency in the range of 25–1200 kHz. The obtained results indicate that cavitational activity as quantified by chemical dosimetry increases linearly with energy input, though the extent of variation is dependent on the geometry of reactor as well as frequency of irradiation. Similar results have also been reported by other researchers [20–24]. Auzay et al. [24] have evaluated the performance of sonochemical reactor by comparing different chemical dosimetry techniques namely iodine, nitrite, nitrate dosimetry and investigated the effects of temperature, power density and reactor configuration. The obtained result show that, sonochemical efficiency increases with acoustic power, irrespective of the dosimetry, however with different rates indicating that the effect of acoustic power is also dependent on the end application. Further, the effect of power dissipation depends on the range of power dissipation and sometimes optimum power dissipation might exist depending on the reactor configuration and application under question [25–30]. Beyond this optimum power dissipation, it is expected that the cavitational activity would be marginally affected or in some cases it might decrease significantly. The optimum power dissipation can be attributed to the shielding effect and acoustic decoupling losses. As power dissipation rate increases, the number of bubbles formed around the transducer surface increases significantly and results in a cloud formation due to coalescence effects. These bubble clouds can absorb or scatter the incident sound waves resulting in decreased transfer of energy. Decoupling losses can also be attributed to the change in acoustic impedance which lowers the energy transfer efficiency from the ultrasound source to the medium. Henglein and Gutierrez [25] have studied the effect of power dissipation on the oxidation of KI in sonochemical reactor of operating frequency as 20 kHz. It has been observed that amount of iodine increases linearly till a power dissipation of 50 W, marginally increases with a further increase till 100 W and then decreases sharply, as shown in Fig. 1. Henglein and Gutierrez [26] have also quantified the effect of power by varying reactor volume and the operating pressure (in the range of 0.7–10 bar) in the presence of external gases (oxygen, argon and mixture of oxygen and argon). The obtained result showed that optimum power strongly depends on reacting volume and operating pressure. Contamine et al. [27] have measured power delivered in water and toluene, in a sonochemical reactor equipped with horn (diameter of 1.3 cm) of irradiation frequency of 20 kHz with maximum power of 600 W and reported that power dissipation rate not only depends on the physicochemical properties of bulk liquid but also on the surface area of irradiating element and frequency of operation. Whillock and Harvey [28] have reported the effect of frequency (in the range of 20–60 kHz), power (maximum upto 100 W) and location of horn on cavitational activity in terms of erosion of stainless steel in horn type of reactor. The obtained results showed that erosion rate (at a distance of 0.5 mm) for 40 and 60 kHz frequency increases linearly till an optimum power of 40 W and 20 W, respectively, whereas in the case of 20 kHz non linear variation was observed. Also, the existence of optimum power for various reaction systems has been reported by Nanzai et al. [30]. The effect of power on the formation of hydrogen peroxide and chlorine ions has been investigated in a rectangular type of reactor (42 cm × 32 cm × 21 cm) with transducer (diameter of 6.5 cm) of irradiating frequency as 200 kHz and maximum power rating of

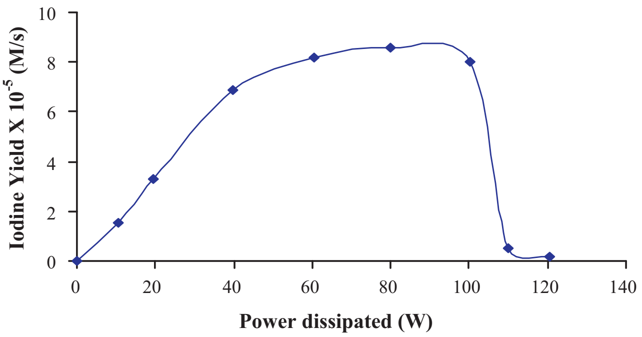

Fig. 1. Effect of power dissipation on iodine yield [25].

200 W. Different cylindrical glass reactors (having diameter of 2, 5, 9 and 12 cm) immersed in a bulk of liquid at 9 mm from transducer, have been used in the work. The results showed that there exists optimum power (16 W) for both H_2O_2 and chloride formation and confirmed that this optimum value is also function of the diameter of reactor. Ratoarinoro et al. [31] have performed the power measurements in a heterogeneous reaction system of preparation of polycyclic molecules, in which KOH and trimethylbenzylammonium chloride (TMBA) were suspended in the reaction medium. The obtained results indicate that the yield of reaction increases with power dissipation and remains constant after optimum power dissipation levels.

Thus, it can be clearly established that the optimum magnitude of power dissipation is a function of frequency of irradiation, reactor geometry, physicochemical properties of bulk liquid, location and surface area of irradiating element and the desired application. Also the presence of additional phases such as solids or two immiscible liquids would play a role in deciding the optimum levels. The presence of solid particles also have a negative effect on the overall cavitational activity owing to higher attenuation of sound wave (reflection, refraction and absorption of sound is higher) [32,33]. Thus, the magnitude of the optimum power dissipation as well as the trends (yield remaining constant or decreasing sharply) beyond the optimum power dissipation would also depend on the type and concentration of solid particles. Extreme care has to be taken in investigating the power dissipation in immiscible reaction systems like in the case of production of biodiesel or emulsification operations, etc. in which the amount of energy dissipated in the system is sum of energy dissipated in the individual phases [34]. Also, in the case of exothermic and endothermic reactions scheme, the corresponding amount of heat evolved or absorbed needs to be accounted in terms of deciding the actual power dissipation levels.

Once the optimum power to be dissipated in the reactor is established for a given set of operating parameters, it is important to quantify power density (W/m^3) which gives tentative idea about the energy requirement per unit volume for given physicochemical transformations. Table 1 [6,19,20,24,34–37] depicts some of the representative work illustrating the type of equipment used with operating parameters, reactor volume, chemical yield for a given type of chemical dosimetry and operating levels of power density. From the table, it can be established that the power density requirement for different transformations is in the range of 0.2×10^3–1600×10^3 W/m^3. Another important question arises that whether the required power should be dissipated from a single or number of segregate power sources, i.e. transducers and what should be the optimum dimensions of these energy transferring agents.

In large scale operation (operating volume in the range of few to few hundred litres), it is extremely difficult to dissipate the entire power in the given reactor volume using single transducer with good efficacy due to the limitations in the construction materials (vibrating piezoelectric crystal). In addition to this, active zones with required minimum cavitational activity in the case of sonochemical reactors are concentred very near (2–3 cm away) the irradiating surface. Thus, in order to attain uniform distribution of the cavitational activity, definitely more than one irradiating surface will be required and the arrangement of these transducers at various locations needs to be optimized. The exact number will depend on the operating volume, dimensions of each transducer and power requirement for the given application. Thus, the efficient design of any sonochemical reactor depends on two key parameters viz. total irradiating surface area and actual power dissipation levels. The total area of transducers can be distributed using multiple transducers which also give location of transducers as additional design parameter. It is always a good choice to dissipate same power using larger irradiation surface (as possible) so that the active cavitational volume is increased and also the collapse intensity is higher due to lower operating intensity of irradiation. A typical range of optimum intensity of irradiation (power dissipated per unit area of irradiating surface, W/cm^2) is 5–20 W/cm^2 which is also dependent on the actual reactor system and the end application.

2.3. Liquid phase physicochemical properties

The important liquid phase physicochemical properties which affect the cavitation phenomena and hence the extent of cavitational effects for the given application include vapor pressure, viscosity and surface tension.

Usually lower vapor pressure of the liquid is preferred, attributed to the fact that with an increase in the vapor pressure of the liquid, the vapor content of the cavity increases thereby lowering the energy released during the collapse. Thus the net cavitational effects will be lower for liquids with higher vapor pressure.

For cavitation to occur in a liquid, it has to overcome the natural cohesive forces present in the liquid. Any increase in these forces will tend to increase the threshold pressure and hence the energy required to generate cavitation. In highly viscous liquids, severe attenuation of the sound intensity occurs and the active cavitating zone gets reduced substantially. Thus it is preferred to use liquids with lower viscosity.

Liquids, with the highest values of surface tension such as water, generally result in higher cavitational intensity. Theoretical studies have indicated that the aqueous liquids and organic liquids such as glycerol show higher cavitating effect as compared to acetone, ethanol and formamide. Presence of surfactants in the system as additives can further enhance the cavitational activity.

2.4. Geometrical design of the reactor

The reactor design in terms of ratio of the diameter of the immersion transducer to reactor diameter, liquid height and position of the transducers plays a important role in deciding the cavitational activity distribution and hence the efficacy of sonochemical reactors for the specific application. Based on a critical analysis of the existing literature [9], following important design related information can be recommended:

- With an increase in the diameter of immersion transducer relative to the reactor diameter, the cavitational activity increases till an optimum ratio, usually dependent on the application. The ratio mainly affects the level of turbulent dissipation of energy and the intensity of the acoustic streaming and hence would be more crucial in the applications where physical effects are more important.
- The extent of immersion of the transducer in an ultrasonic horn or the extent of liquid height, which affects the extent of reflection of the incident sound waves from the liquid surface as well as the reactors walls, also shows an optimum value.
- The position of the transducers in the reactors based on multiple transducer arrangement (with possibly multiple frequency operation) should be done in such a way that maximum and uniform cavitational activity is obtained. Theoretical analysis of cavitational activity distribution aids in arriving at an optimum location of the transducers. Typically flow cell type of arrangements are more suitable for large scale operation as this definitely give flexibility in terms of the continuous operation and also gives an option of arranging the transducers on the wall of the reactor on opposite faces so that standing wave patterns can be generated. Typically hexagonal or rectangular cross-sections have been reported to yield excellent distribution of the cavitational activity and are recommended for the large scale operation. The

Table 1
Some of the representative work highlighting the operating power density for different applications with the corresponding yields.

Sr. No	Reference	Type of sonochemical reactor with operating parameters	Chemical yield (mol/J)	Mapping method/ Reaction type	Power density (*P*/*V*) (W/m^3)
1	Auzay et al. [24]	Experiments were performed in two different cylindrical type sonochemical reactors (denoted as A and B). The operating capacity of reactor A was 0.4 L (diameter of 6.5 cm and height of 18.4 cm) and was fitted with a piezoelectric transducer (P1-89) of frequency as 365 ± 1 kHz and maximum power rating in the range of 15–60 W, where as the operating capacity of reactor B was 2 L (diameter of 10.2 cm and height of 37 cm) of frequency 367 ± 1 kHz and similar power rating as that of reactor A. Actual power dissipated in reactor calculated by calorimetric study for reactor A and B was 19.0 ± 2.6 W and 27.5 ± 2.2 W, respectively.	$8.8 \pm 1.4 \times 10^{-10}$ $0.71 \pm 0.09 \times 10^{-10}$ $14.0 \pm 0.5 \times 10^{-10}$ $19.9 \pm 6.1 \times 10^{-10}$ $3.8 \pm 0.4 \times 10^{-10}$ $0.54 \pm 0.05 \times 10^{-10}$ $2.1 \pm 0.5 \times 10^{-10}$ $9.2 \pm 0.7 \times 10^{-10}$	Iodine dosimetry Nitrite dosimetry Nitrate dosimetry H + dosimetry Iodine dosimetry Nitrite dosimetry Nitrate dosimetry H + dosimetry	Reactor A $(41–54) \times 10^3$ Reactor B $12.65–14.85 \times 10^3$
2.	Cintas et al. [34]	Rectangular continuous scale reactor with three circular transducers (of diameter of 5 cm) located at bottom of reactor with irradiating frequency as 21.5 kHz and maximum power rating as 1520 W (900 W form transducers + 100 W form pump + 20 W form electronics + 500 W form thermostat). Maximum operating volume reactor was 0.5 L, in which flow rate was varied in the range 20–100 mL/min (optimal flow rate as 55 mL/min) and residence time was varied in the range of 5–20 min.	–	For biodiesel production	280×10^3
3.	Asakura et al. [6]	Rectangular type of sonochemical reactor (of length 50.8 cm, width 50.8 cm and height 67.2 cm) equipped with modular unit (dimension as 23.6 cm in length, 16.1 cm in width and 7.3 cm in height) located at bottom and/or side of the reactor. This unit consists of total of six transducers of diameter 5 cm each with frequency of irradiation as 500 kHz and maximum power rating of 620 W. Experiments were performed by varying the reaction volume in the range of 77–131 dm^3.	6.3×10^{-10}	Iodine dosimetry	$4.73–8.73 \times 10^3$
4.	Nikitenko et al. [35]	The experiments were performed in two type of sonochemical reactor (designated as A and B), both reactor were fitted with transducer of irradiating frequency of 20 kHz. The actual power dissipation and intensity was varied in the range of 45–80 W, 2.1–4.5 W/cm^2 for reactor A and 30–65 W, 31.4–62.7 W/cm^2, respectively, for reactor B.	7.9×10^{-10} 8×10^{-10} 8.3×10^{-10} 7.5×10^{-10} 7.3×10^{-10} 7.4×10^{-10}	Hydrogen peroxide (Reactor A) Hydrogen peroxide (Reactor B)	800×10^3 1100×10^3 1600×10^3 600×10^3 900×10^3 1300×10^3
5.	Bhirud et al. [36]	Rectangular type of reactor (length 15 cm, width 33 cm and height as 20 cm) of operating capacity 7 L, equipped with a longitudinal vibrating horn (of length 30 cm and diameter as 30 mm) of irradiating frequency 36 kHz, intensity as 0.53 W/cm^2 attached to the side wall at the centre of reactor. Maximum power rating was 150 W and actual reactor was operated at 85% input power that is 127.5 W.	2.13×10^{-5}	Formic acid degradation	18.21×10^3
6.	Koda et al. [19]	Experiments were performed in seven different types of reactor by keeping the operating volume constant as 50 dm^3 and varying frequency of irradiation in the range of 19.5–1200 kHz and power dissipated in the range of 35–220 W. For the sake of convenience, the reactors were designated based on frequency and power, such as Reactor 1 (20 kHz, 40–200 W) indicating that reactor 1 was operated at frequency of 20 kHz with power dissipation in the range of 40 and 200 W, similarly the other types of reactor used were: Reactor 2 (40 kHz, 35 W), Reactor 3 (45 kHz, 110–220 W), Reactor 4 (96 kHz, 50–150 W), Reactor 5 (96 kHz, 50–100 W), Reactor 6 (130 kHz, 50–200 W), Reactor 7 (200 kHz, 140–200 W), Reactor 8 (400 kHz, 120 W), Reactor 9 (500 kHz, 10–40 W), Reactor 10 (1200 kHz, 120 W)	$2.3 \pm 0.1 \times 10^{-10}$ $0.60 \pm 0.02 \times 10^{-10}$ $2.8 \pm 0.1 \times 10^{-10}$ $0.60 \pm 0.02 \times 10^{-10}$ $3.7 \pm 0.1 \times 10^{-10}$ $0.67 \pm 0.06 \times 10^{-10}$ $4.5 \pm 0.2 \times 10^{-10}$ $16.8 \pm 1.0 \times 10^{-10}$ $4.1 \pm 0.2 \times 10^{-10}$ $22.6 \pm 1.6 \times 10^{-10}$ $5.6 \pm 0.4 \times 10^{-10}$ $15.2 \pm 0.9 \times 10^{-10}$ $8.3 \pm 0.6 \times 10^{-10}$ $19.3 \pm 1.2 \times 10^{-10}$ $7.8 \pm 0.2 \times 10^{-10}$ $20.3 \pm 1.2 \times 10^{-10}$ $7.1 \pm 0.2 \times 10^{-10}$ $2.6 \pm 0.1 \times 10^{-10}$ $0.64 \pm 0.3 \times 10^{-10}$	Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry Fricke dosimetry KI dosimetry	$0.8–4 \times 10^3$ (Reactor 1) 0.7×10^3 (Reactor 2) $2.2–4.4 \times 10^3$ (Reactor 3) $1–3 \times 10^3$ (Reactor 4) $1–2 \times 10^3$ (Reactor 5) $1–4 \times 10^3$ (Reactor 6) $2.8–4 \times 10^3$ (Reactor 7) 2.4×10^3 Reactor 8 $0.2–0.8 \times 10^3$ (Reactor 9) 2.4×10^3 (Reactor 10)
7.	Gogate et al. [37]	Experiments were carried out in four types of reactor namely Reactor 1: ultrasonic horn of operating volume of 50 ml (irradiating surface area 4.91 cm^2, frequency as 22.7 kHz and actual power dissipation of 24 W), Reactor 2: ultrasonic bath of dimension 15 × 15 × 15 cm with volume of 1500 ml (frequency as 22 kHz and actual power dissipation rate of 48 W), Reactor 3: dual frequency rectangular flow reactor with three transducers mounted on the opposite faces with operating volume of 1.5 L (frequency as 25 and 40 kHz with power dissipation as 88.8 W), Reactor 4: triple frequency hexagonal flow cell reactor of operating volume of 7 L (frequency 20, 30, 50 kHz and maximum power dissipation as 702 W)	6.9444×10^{-8} 1.6667×10^{-6} 2.3611×10^{-6} 3.3333×10^{-6}	Formic acid degradation	480×10^3 (Reactor 1) 137×10^3 (Reactor 2) 59.2×10^3 (Reactor 3) 100×10^3 (Reactor 4)
8.	Entezari et al. [20]	Experiments were performed in horn (of frequency 20 kHz) type of reactor having area of irradiation as 1.1, 2.46 and 8.04 cm^2 and power dissipation as 38, 37 and 36 W, respectively, with an operating volume of 100 cm^3	0.197×10^{-10} 0.533×10^{-10} 0.985×10^{-10}	KI dosimetry	380×10^3 370×10^3 360×10^3

formation of standing wave patterns also increases the effective cavitational intensity in the reactor system and can aid in increasing the processing rates of given applications. The number and location of the transducers also affect the hydrodynamic behaviour and the mixing characteristics in the reactor which can be of prime importance especially for the physical processing applications. While maintaining the geometric similarity for the design of large scale sonochemical reactors, it is also important to maintain similar conditions of hydrodynamics and mixing characteristics in the reactor so as to achieve the desired objectives.

3. Hydrodynamic behaviour

The knowledge of flow behaviour is also one of the key parameters in obtaining an effective scale up of the reactor. This is due to the fact that the generation of acoustic streaming can accomplish effective improvement in the rate of the transport processes specifically the heat and mass transfer in the case of multiphase reactions. Acoustic streaming currents also provide good mixing, possibly change the morphology of the biological cells, remove loosely adhering surface layers, etc. which may be factors of critical importance depending on the specific application under question. In addition, the understanding of the hydrodynamic behaviour can be useful in determining the mean residence time of the reacting species which can aid in designing continuous reactor systems.

Propagation of the sound wave in the liquid results in the movement of fluid molecules. This movement occurs in the direction of sound wave propagation, away from the transducer as a result of the pressure gradient and has been described as acoustic streaming. The flow behaviour depends on the physicochemical properties of liquid as well as on the operating parameters like power dissipation, frequency of irradiation and the geometric parameters such as dimension of transducers and reactor geometry. The dependency of the local velocity and flow patterns in the given system can be successfully quantified by various methods namely laser tomography, electrochemical method, particle image velocimetry (PIV) and laser doppler anemometer (LDA) techniques. Table 2 [21,38–44] depicts some of the representative work in the quantification of acoustic streaming giving details about reactor type with the scale of operation and techniques used for quantification of the hydrodynamic behaviour. We now discuss some of the important hydrodynamic aspects in the case of conventional reactor systems, i.e. ultrasonic horn and ultrasonic bath.

3.1. Ultrasonic horn reactor

Promising work in quantification of acoustic streaming for ultrasonic horn has been reported by Kumar et al. [42]. It has been observed that there is a spatial variation in the mean (in axial, radial and tangential direction) and RMS fluctuating velocity, which also depends on the operating power density. The results showed that maximum axial velocity (1.6 m/s for 15 kW/m^3 of power density) was obtained very near to the irradiating surface and decreases away from the transducer along the axial direction with minimum at the bottom of the reactor. A sample vector plot obtained in the work has been shown in Fig. 2. In addition to this, a correlation has also been proposed to calculate the axial velocity, which is valid for intensity range of 226–527 kW/m^2, as follows:

$$V_{axial} = 2.63 \times 10^{-9} (I)^{1.41} (Z)^{-0.37} \left(\frac{(R-r)}{R} \right)^{9.4} \tag{3}$$

where V_{axial} is velocity in axial direction (m/s), I is the intensity of irradiation (W/m^2), Z is axial distance (m), R is radius of reactor (m) and r is a radial distance (m).

The velocity in the radial direction also decreases with an increase in the radial distance (away from the transducer towards the wall) and also with an increase in the axial distance. It should be also noted that the radial component of velocity has much lower magnitude (10% of the mean axial velocity). So, it should be believed that horn type reactor behaves like a jet loop reactor, where axial velocities are dominating.

Similar variation in the mean velocity with distance from the transducer has been reported by Tsochatzidis et al. [45] (maximum velocity of 1.6 m/s was obtained very near to the irradiating surface) in a rectangular glass type of reactor (0.1 m × 0.15 m × 0.23 m) with irradiating frequency of 20 kHz and actual power dissipation of 179 W. The radial velocity components have been quantified and a mathematical equation has been established to quantify the proportions of the radial and axial velocity components in the system:

$$V_{radial} = V_{axial\,\max}\, e^{(-0.693(y/b)^2)} \tag{4}$$

where V_{radial} is velocity in radial direction (m/s), $V_{axial\,\max}$ is maximum velocity in axial direction (m/s), y is axial distance (m) and b is radial distance (m).

The results obtained by using Eq. (4) show that, higher velocity was obtained near to the transducer and lower at the wall. However, the equation also predicts that the variation remains unchanged along the axial distances, which is not entirely correct. This is clear due to fact that Eq. (4) fails to incorporate attenuation of sound wave, which has a significant effect. So, it is more recommended to use the correlation proposed by Kumar et al. [42] to obtain a quantitative idea about the acoustic streaming patterns in the horn type reactor.

3.2. Ultrasonic bath reactor

The variation in the liquid circulation velocity generated due to acoustic streaming in the bath type sonochemical reactor has also been studied [38,39,43,44,46]. Generally it is believed that, streaming generates liquid currents in direction away from the transducer towards the free liquid surface and undergoes reflections at the reactor wall or the liquid surface generating a mixed recirculation

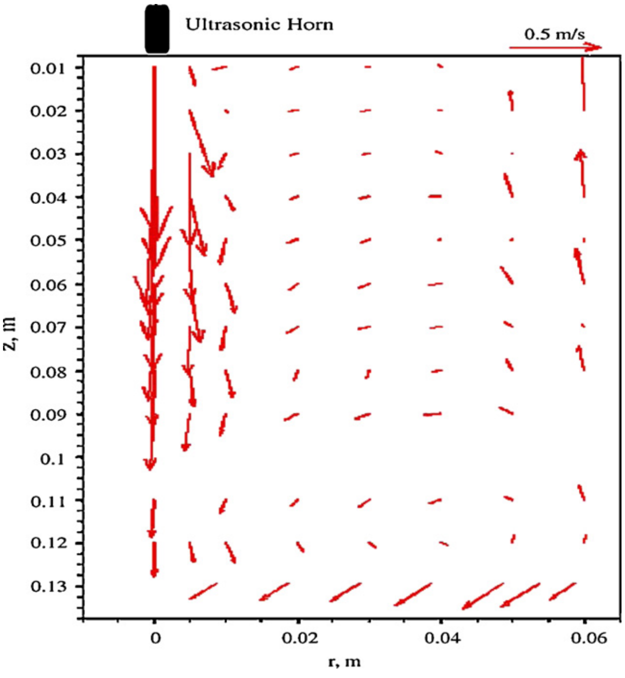

Fig. 2. Two dimensional ($r-z$) vector plot for the mean velocity at 35 kW/m^3 [42].

Table 2
Some of the representative work in understanding the hydrodynamic behaviour of sonochemical reactors.

Sr. No	Details about reactor and operating parameters	Method used	Magnitude of velocity and important findings	Reference
1	PVC cylindrical reactor (diameter 0.10 m, height 0.10 m) equipped with stainless steel plate fixed at the bottom of reactor (diameter 0.15 m, thickness of 5.84×10^{-3} m) with piezoelectric transducer of irradiating frequency as 500 kHz (diameter 0.04 m) with maximum power rating of 100 W.	Hydrodynamic behaviour has been analyzed by determining Residence time distribution (RTD) in the bulk of liquid by means of tracer (NaCl solution) and obtained results were compared by numerically solving equations proposed by Eckart and CSTR model.	The obtained RTD response was exponential, which indicates that sonochemical reactor behaves like ideal reactor system and acoustic streaming velocity was in the rage of 0.01–0.03 m/s for power dissipation of 40 W and 0.05–0.10 m/s for higher power of 100 W.	Gondrexon et al. [38]
2	Sonochemical reactor (maximum operating volume as 3.5 L) equipped with two types of longitudinal vibrating horn (cylindrical and stepped) of irradiating frequency 20 kHz and maximum power rating of 1000 W.	The variation in acoustic streaming velocity was studied using the PIV and photographic technique.	High velocity region was observed very near to irradiating surface along the direction of horn with magnitudes in the range of 0.3–0.35 m/s.	Dahlem et al. [21]
3	Sonochemical reactor (filled with water up to 10 cm height) with piezoelectric transducer of irradiating frequency of 500 kHz with power rating in the range of 25–150 W.	Acoustic streaming velocity in the reactor was measured by laser tomography techniques and numerical investigations have been performed by using Nyborg model.	It has been reported that: 1. Streaming velocity increases with power dissipation, from 0.0066 m/s at 25 W to 0.023 m/s at 100 W and 0.033 m/s at 150 W. 2. As volume of liquid increases the velocity decreases linearly with a maximum of 0.0266 m/s obtained at liquid height of 0.05 m at a power dissipation of 100 W.	Chouvellon et al. [39]
4	Simulations were performed in cylindrical type of reactor (diameter 6 cm and liquid height of 3 cm) with transducer of irradiating frequency of 500 kHz, located at the bottom of reactor.	Streaming velocity was determined using CFD simulations and obtained results were compared with experimental investigation by PIV techniques.	The obtained results showed that higher velocity zone exits at the centre of reactor with recirculation patterns, with velocity variations in the range of about 1 cm/s	Laborde et al. [40]
5	Experiments were carried in rectangular type of reactor with obstacle (approximately 4 cm in diameter) located 3.5 cm from transducer of emitting surface area as 5 cm^2 and irradiating frequency of 3 MHz with intensity as 2.2 W/cm^2.	Acoustic streaming velocity was determined by PIV technique.	Effect of intensity of irradiation on acoustic streaming velocity has been investigated and a linear variation in velocity was observed as per the correlation: V = 0.0992 I, where V is velocity in cm/s and I is intensity in W/cm^2. The maximum velocity of 0.27 m/s was obtained at 2.1 W/cm^2	Frenkel et al. [41]
6	Experiments were performed in horn (diameter 0.013 m) type of reactor of frequency of irradiation as 20 kHz, by dipping the horn tip 2 cm from the free liquid surface with maximum power density up to 35 kW/m^3.	Generation of flow patterns in reactor were studied using Laser Doppler anemometer (LDA) technique and using k–ε model in the CFD simulation. In addition, mixing time was measured using conductivity probes	Variation of mean velocity in axial, radial, tangential direction and fluctuating rms velocity with power density has been quantified. Maximum mean axial velocity was obtained at a distance of 0.01 m away from horn at the centre of reactor as 1.6, 1.3 and 0.57 m/s, respectively, for 35, 25 and 15 kW/m^3 dissipated power density, whereas mean radial velocity has been found to decrease with an increase in axial distances but it increases with an increase in power dissipation rate. Similar variation in rms fluctuating velocity was obtained but with higher magnitude.	Kumar et al. [42]
7	Experiments were performed in rectangular parallelepiped reactor (operating volume 4 L) made from quartz glass of internal dimensions as 200 mm × 200 mm × 650 mm, and fitted with transducer (diameter 50 mm) located at centre of reactor of irradiating frequency 490 kHz with power rating in the range of 5–50 W.	Velocity in reactor was quantified by Laser Doppler Velocimeter (LDV) technique.	The effect of power dissipation on acoustic velocity has been studied with and without mechanical string. Mixed flow patterns (in axial and radial direction) can be achieved by employing mechanical agitation. The local velocity is a strong function of pressure gradient in the bulk of liquid and it has been also observed that magnitude of velocity increases with an increase in the power dissipation, Maximum velocity of 1.8 cm/s was observed at 30 W.	Kojima et al. [44]

Table 2 (*Continued*)

Sr. No	Details about reactor and operating parameters	Method used	Magnitude of velocity and important findings	Reference
8	Experiments were performed in glass cylindrical reactor (190 mm height and 60 mm diameter) with operating volume of 300 ± 7 mL and fitted with transducer (of diameter 6, 12, 25 cm) of irradiating frequency of 20 and 40 kHz with maximum power variation in the range of 45–240 W. Furthermore, glass electrode of diameter 8 mm was used as a external obstacle.	The velocity in axial and tangential direction was quantified with and without presence of obstacle (electrode) by PIV and laser tomography techniques.	The effect of frequency of irradiation and intensity of irradiation on the local velocities have been analyzed in the presence of external electrode: 1. Velocity (both axial and radial) increases with increase in power dissipation for lower frequency, however for higher frequency it goes through maxima. The maximum velocity without presence of electrode was 1.13 m/s for transducer of diameter 12 cm and irradiating frequency 20 kHz with power rating 90 W; whereas for transducer of diameter 25 cm of irradiating frequency 40 kHz with power rating 120 W, maximum velocity was 0.392 m/s. The maximum velocity in the presence of electrode at distance of 30 mm and 10 mm from the bottom of reactor was 1.35 m/s and 1.49 m/s, respectively, for transducer diameter of 12 cm, frequency of 40 kHz and power rating of 90 W. 2. As intensity of irradiation increases the velocity in axial and radial direction increases up to approximately 15 W/cm^2 and then remains constant for 20 kHz frequency.	Mandroyan et al. [43]

patterns. Kojima et al. [44] have quantified the variation in velocity with power dissipation in the range of 10–50 W and reported that, at low power dissipation levels (10 W) velocity fountain was observed from the transducer surface resulting in a random flow in the liquid. As the power dissipation increases, the mean velocity of the flow very near to transducer was directed upward with higher magnitude and was reflected in downward direction near the wall as shown in Fig. 3. Similar effect of intensity of irradiation has also been reported by Frenkel et al. [41], who have shown that the maximum acoustic streaming velocity increases linearly with the intensity of ultrasound. Chouvellon [39] have reported the effect of power dissipation in the range of 50–100 W and have reported a similar increase in the intensity of acoustic streaming.

Mandroyan et al. [43] have investigated the hydrodynamic behaviour in the presence of additives and observed that the presence of external objects (above the irradiating surface) results in the generation of velocity component in tangential direction. Quantitatively speaking, for a frequency of 20 kHz (horn diameter of 25 mm and power input of 45 W) the velocity in axial and radial direction was 14.4 and 4 cm/s, respectively, where as velocity component in axial, radial and tangential directions were 17.4, 5.4 and 7.8 cm/s, respectively, in the presence of external object (at similar operating conditions). Thus, it can be said that the dominant axial flow pattern in the sonochemical reactor can be altered in the presence of additives.

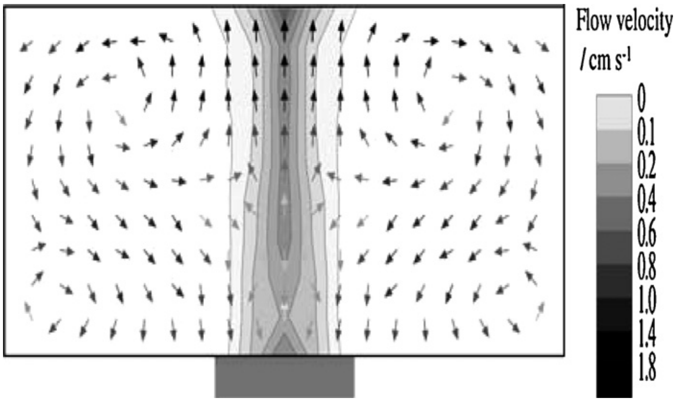

Fig. 3. Distributions of the flow velocity in a sonochemical reactor of dimension (length = 20 cm, height = 10 cm width = 20 cm) with transducer diameter = 50 mm, of irradiating frequency as 490 kHz and power of 50 W. [44].

4. Mixing time characteristics

In the design of semi-batch or continuous reactor, the knowledge of mixing time plays an important role in deciding the stream flow rates and dimensions of the reactor. There are a number of techniques used for the quantification of mixing time in multiphase reactors such as mechanically agitated contactors, bubble column, etc. however only few techniques has been used for sonochemical reactors namely measurement of pH, conductivity, dissolved oxygen, particle image velocimetry and theoretical investigations like computational fluid dynamics (CFD). In this section, we will discuss the variation in the mixing time in conventional sonochemical reactor and dependency of the mixing time on different operating parameters such as frequency, power and reactor geometry.

The earliest work related to mixing characteristics was by Monnier et al. [47,48], who have reported quantification of the mixing time (micro) in batch as well as continuous sonochemical reactors by measuring the segregation index (for perfect mixing segregation index is equal to zero) by using a model reaction of iodide and iodate, coupled with a neutralization reaction, well known as Dushman reaction. Also the effect of operating parameters such as frequency (in the range of 20–955 kHz with maximum power rating of 67W), intensity, power and liquid viscosity on micromixing has been investigated. The obtained results indicate that more uniform mixing was obtained at lower frequency due to stronger contribution of the physical effects of cavitation phenomena and the mixing time decreases with an increase in the intensity of irradiation as well as net power dissipation.

Gondrexon et al. [38] investigated the mixing characteristics using a NaCl tracer pulse being injected into the inlet stream entering the ultrasonic cell. The residence time distribution was identified by monitoring the conductivity at the outlet. The experimental reactor consists of PVC cylindrical body (diameter 0.10 m, height 0.10 m) equipped with a stainless steel plate fixed to the bottom (diameter 0.15 m, thickness half a wavelength 5.84×10^{-3} m). The high-frequency ultrasound is produced by a 500 kHz piezoelectric transducer (diameter 0.04 m, titanate lead-zirconate) with a variable electrical output ranging from 0 to 100 W. It has been reported that over the tested range of ultrasonic powers and reactor flow rates, the reactor behaved as a continuous stirred-tank reactor (CSTR) attributed to strong convective currents occurring within the reactor.

Vichare et al. [49] have investigated the mixing time variation in horn type of reactor (irradiating frequency as 22.7 kHz with actual power dissipation 24.5 W) by quantifying the average liquid circulation velocity based on the fact that minimum five circulations are required for good mixing. The obtained results were compared with the experimental investigations involving the measurement of conductivity of NaCl solution added into the system as a tracer. A correlation has been developed for the prediction of mixing time considering the dimensions of the reactor, horn tip and location of transducer as follows:

$$\theta_{mix} = 7 \times 10^{6} d^{-0.235} \left[\frac{Z^{3/2} T^{3} (T + 2Z)^{-2} d_h^{-4}}{\nu_h^{2} g^{1/2} \mu^{-2} \rho_l^{2}} \right] \tag{5}$$

where θ_{mix} is mixing time (s), d is distance between horn tip and bottom of reactor, Z is height of liquid in reactor (m), T is reactor diameter (m), d_h is tip diameter of horn (m), ν_h is velocity of horn (m/s) (amplitude of oscillation in m × frequency in Hz), g is acceleration due gravity (m/s^2), μ is viscosity of liquid (N s/m^2) and ρ_l is bulk density of liquid (kg/m^3).

Kumar et al. [42] have also investigated the variation in mixing time by varying the D/T ratio (where D is diameter of horn = 0.013 m and T is the reactor diameter) in the range of 0.089–0.15 for different operating volumes in the range of 0.5–2.5 L at constant power density (35 kW/m^3) using two techniques viz. experimental (by measuring conductivity of sodium chloride tracer) and CFD simulation (by considering standard $k - \varepsilon$ model). The obtained results indicate that the mean velocity in axial, radial and tangential direction increases with a decrease in D/T ratio. Also, higher mixing time was obtained for reactor diameter of 0.11 m as 62 s and lower for diameter of 0.14 m (smaller D/T ratio 0.089) as 12 s at same power density level of 35 kW/m^3. This can be attributed to the fact that average turbulent kinetic energy (TKE) is higher for the smaller vessel. This study clearly implies that sonochemical reactor requires optimum turbulence level to yield beneficial mixing characteristics. Deshpande et al. [50] have developed a unified correlation, based on MBR statistics, to quantify the mixing time in various types of reactors. The correlation successfully takes into account geometry of reactor and is based on the quantification of the velocity by PIV techniques and large eddy simulation (LES) and the obtained data have been analyzed using multiresolution wavelet transforms (WT). Kumar et al. [42] have also investigated the effect of power density on mixing time and reported that as power density increases from 15 to 35 kW/m^3 mixing time decreases from 131 to 17 s.

It has been generally observed that the mixing time investigations are based on the use of externally added tracers such as salts and also sometimes detection hardware is installed in the system [38,47,49]. The introduction of the injection and detection hardware into the system can add a substantial volume which distorts the RTD. Ideally, the actual input to the system should be measured or at least quantified in some manner so that deviations from the ideal impulse or step inputs can be accommodated if necessary through appropriate analysis of the tracer concentration histories. Yusaf and Buttsworth [51] have reported a direct visualization technique for the understanding of the mixing process, which has been applied also within the high energy region close to the tip of the ultrasonic probe. The analysis proceeds by determining an approximate turbulent diffusivity in a batch reactor arrangement for different values of ultrasonic energy input. It has been observed that for the input electrical power levels between 70 and 120 W and a processing volume of 30 mL, the effective turbulent diffusivity varied from about 0.2×10^{-3} to 0.7×10^{-3} m^2/s. It has also been reported that the mixing rate within the present batch arrangement (30 mL) is a function of the applied ultrasonic power, but the macroscopic mixing is substantially complete within one second for absorbed ultrasonic power levels greater than 40 W (thermal dissipation).

It is also worth mentioning here that all the reported analysis for mixing time has been for a single frequency operation with single transducer used in the reactor. Similar analysis needs to be performed for the multiple frequency and multiple transducer based reactors which are likely to be used in the large scale operations. The liquid circulation patterns is expected to be significantly different in the case of multiple transducer system as compared to a single transducer based conventional sonochemical reactors.

5. Mass transfer characteristics

Generally speaking, the mass transfer characteristics can be quantified in terms of the overall gas–liquid mass transfer coefficient by measuring the dissolved concentration of oxygen in the liquid at regular time intervals. The equation for the prediction of mass transfer coefficient can be given as follows:

$$K_L a V (C^{*}_{o_2} - C_{o_2}) = V \frac{\mathrm{d}(C_{o_2})}{\mathrm{d}t} \tag{6}$$

where $K_L a$ is the volumetric mass transfer coefficient (1/s), V is the volume of the liquid in the reactor (m^3), $C^{*}_{o_2}$ is the saturation concentration of dissolved oxygen (mmol/m^3) and C_{o_2} is the concentration of dissolved oxygen in the reactor at any time t (mmol/m^3).

Actual mass transfer coefficient can be determined by integrating Eq. (6) and plotting concentration against time data (which is a straight line passing through origin with slope as $K_L a$) and is given by the following equation:

$$\log \left[\frac{C^{*}_{o_2}}{C^{*}_{o_2} - C_{o_2}} \right] = K_L a t \tag{7}$$

Here it should be noted that the applicability of the method highly depends on the operating temperature, which can constantly vary in the liquid bulk due to the energy dissipation from the incident sound waves. Thus, proper precaution must be taken during the experimental runs to ensure the correctness of the measurements.

In one of the very earliest works on mass transfer characteristics, Cadwell and Fogler [52] measured the rate of CO_2 absorption by glycerol in a batch reactor with and without ultrasonic irradiation. It has been observed that the absorption rate is enhanced due to the presence of ultrasound, which results in a surface renewal effect associated with the vortex structures observed in batch configurations with 20 and 800 kHz irradiation. This work is a important basic work to show that the mass transfer coefficient is indeed enhanced due to the acoustic streaming effect [53] and the associated turbulence.

Kumar et al. [54] have investigated the quantification of overall volumetric mass-transfer coefficient ($K_L a$) in a conventional horn reactor with irradiation frequency as 20 kHz, maximum power rating of 65 W and ultrasonic bath reactor (of operating capacity of 3.3 L and transducers arranged in triangular pitch) with irradiation frequency as 20 kHz and rated power of 120 W. Based on this study, a mathematical correlation has been proposed for the air–water system:

$$\begin{aligned} K_La &= 0.029\left(\frac{P}{V}\right)^{0.17}(V_g)^{0.37} \quad \text{for ultrasonic horn} \\ K_La &= 0.0039\left(\frac{P}{V}\right)^{0.4}(V_g)^{0.6} \quad \text{for ultrasonic bath} \end{aligned} \tag{8}$$

where (P/V) is the power dissipated per unit volume (W/m^3) and V_g is superficial gas velocity (m/s).

Laugier et al. [55] have investigated the mass transfer characteristics (gas–liquid as a function of pressure) in a closed sonochemical reactor of irradiating frequency as 20 kHz and power dissipation of 16.7 W by employing mechanical agitation using Rushton turbine with maximum agitation speed as 3000 rpm. The mass transfer coefficient has been quantified by measuring the solubility of nitrogen gas, which has been selected due to low solubility (mass transfer resistance is located only in the liquid film). The obtained results showed that the mass transfer coefficient increases substantially due to the presence of ultrasound (0.18 s^{-1}) as compared to that obtained in the absence of ultrasound (0.08 s^{-1}). Also the mass transfer coefficient significantly depends on the operating temperature and pressure in the reactor.

Gondrexon et al. [56] have investigated the effects of several parameters, such as the volume of solution and acoustic power, on the degassing and re-gassing phenomena when the upper liquid surface is exposed to the atmosphere. In addition, a reflector has been used to identify the role of the acoustic fountain and cavitation bubbles. It has been observed that the dissolved O_2 concentration reaches a steady state soon after sonication begins. The 'equilibrium' dissolved oxygen, seems to result from the superposition of two simultaneous mechanisms: a degassing action that results from cavitation bubbles, and an absorption phenomenon that involves the acoustic fountain effect. The 'equilibrium' dissolved concentration of oxygen was observed to be independent of the ultrasound operating parameters and it appears to depend only on the true thermodynamics meaning the operating temperature and pressure of the system. The rate of absorption was found to depend on the ultrasonic power dissipation as well as the volume of the reactor. It has been observed that the rate of absorption is higher at higher power dissipation and lower overall volume of the reactor contents.

It should be again noted here that similar to mixing time measurements additional work is required for quantifying the mass transfer characteristics in the case of multiple frequency and multiple transducer operation on a relatively large scale of operation.

6. Attenuation of sound waves in liquid bulk

Propagation of sound wave results in an increase in the local temperature due to the fact that kinetic energy associated with the wave is directly converted into thermal energy due to absorption and reflection meaning that the net kinetic energy will also decrease. This decrease in the energy will be dependent on the liquid physicochemical properties. Furthermore, scattering of the incident wave due to the presence of suspended solid particles and reactor wall also contribute to the loss of intensity [57,58].

Eckart [57] has quantified the attenuation factor of sound wave in liquid owing to shear and bulk viscosity as follows:

$$\alpha_f = \frac{\omega^2 \mu b^*}{2\rho C^3} \tag{9}$$

where α is attenuation factor (1/m), ω is angular frequency of irradiation, μ is viscosity of bulk of liquid (kg m/s^2), b^* is constant ($b^* = (4/3) \times 10^7$ for water containing cavitating bubbles), ρ is density of liquid (kg/m^3) and C is speed of sound in liquid (m/s). It should be noted here that the applicability of this equation is limited only for the low frequency operation.

Majumdar et al. [59] have reported that attenuation coefficient is a function of physicochemical properties of liquid and the frequency of irradiation and is given by the following equation:

$$\alpha_f = \frac{8\pi^2 \mu f^2}{3\rho C^3} \tag{10}$$

where α_f is attenuation factor (1/m), μ is viscosity of bulk of liquid (kg m/s^2), f is frequency of irradiation (Hz), ρ is density of liquid (kg/m^3) and C is speed of sound in liquid (m/s).

Kirchoff [60] have reported the dependency of sound wave on the thermal conduction in the liquid medium as follows:

$$\alpha_{th} = \frac{2\pi^2 K(\gamma - 1)f^2}{\rho C_p C^3} \tag{11}$$

where α_{th} is attenuation factor (1/m), K is the thermal conductivity (w/m K), γ is the adiabatic gas constant, f is frequency of irradiation (Hz), ρ is density of liquid (kg/m^3), C is speed of sound in liquid (m/s) and C_p is the specific heat (J/kg K).

Thus, it is important to consider all the contributing effects to the overall attenuation process in terms of the liquid physicochemical properties as well as the operating parameters and the presence of additives such as solid particles or gases in the system. The total attenuation factor should be obtained by algebraic sum of all the individual attenuations contributing in the reactor. At this juncture, it should be noted that accuracy of these equations highly depends on the correctness in the prediction of physiochemical properties of the liquid. During cavitational events (formation and growth of bubbles) homogenous liquid medium behaves as a non-homogenous, i.e. single phase liquid continuum is converted into two phases. Also if initially two phases (containing suspending solids or immiscible liquids) are present, it gets converted into a three phase system. This behaviour possibly alters the viscosity, density and the speed of sound and hence can affect the final attenuation considerations. In an earlier work, Sutkar and Gogate [61] have made an attempt to quantify the variation in the attenuation factor due to presence of electrolyte solution (aqueous NaCl solution) in a high frequency sonochemical reactor. However, this approach fails to take into account the effect of local fraction of volume occupied by gas/vapor. Commander and Prosperetti [62] have successfully developed the model to determine the variation in local pressure in a bubbly liquid by calculating the bubble fraction as follows:

$$\beta = \frac{(4/3)\pi R^3 N}{V} \tag{12}$$

where β is fraction of volume occupied by gas, R is radius of bubble (m), N is number of bubbles and V is volume of liquid (m^3). The obtained value of β from equation (12) can be used to calculate the density of mixture as follows:

$$\rho_m = (1 - \beta)\rho_l + \beta\rho_g \tag{13}$$

where ρ_m is density of liquid mixture (kg/m^3) ρ_l is density of liquid (kg/m^3) and ρ_g is density of gas (kg/m^3). After successful determination of the density of liquid mixture, speed of sound can be calculated by utilizing the elastic modulus of liquid as follows:

$$\text{Speed of sound} = \left(\frac{\text{Elastic modulus}}{\text{Density of liquid medium}}\right) \tag{14}$$

Thus, it is recommended to use these equations which consider the effect of formation of bubbles on the physiochemical properties for getting a realistic idea about the attenuation of incident sound waves.

7. Recent advances in the development of sonochemical reactor

There are a number of illustrations in literature where the effects generated due to cavitation have been successfully utilized for physicochemical transformations but are constrained to simple horn and bath type of reactor configurations. A major problem for construction of large scale reactors is unavailability of efficient transducers as well as configuration which can give uniform cavitational activity.

The problem with availability of transducers is more from material science background and hence will not be discussed in details here. Still the work of Gallego-Juárez et al. [63] is worth mentioning here which comprehensively gives the difficulties for effective utilization of piezoelectric transducers for higher scale of operation viz. lower power rating, limited dimensions and alteration in the performance due to variation in operating voltage. A basic structure of new family of high power ultrasonic (HPU) transducers, with higher power capacity, efficiency, radiating surface area and more sophisticated control system viz. stepped-plate transducer, grooved-plate transducer, stepped-grooved-plate transducer, flat-plate transducer with reflectors have also been reported. In addition, efficacy of novel design has been fruitfully applied to some industrial applications in food industry (defoaming and drying), environmental processes (air cleaning and sludge filtration) and manufacturing processes (textile, coating and pigment dispersion in paint). The utility needs to be further evaluated for chemical processing applications.

In the following section, a few novel reactor configurations based on new geometry (location of transducers with batch and/or continuous scale of operation) or modifications in the conventional approach with an objective of achieving uniform cavitational activity distribution have been discussed. These reactors have a possibility of successful application at larger scale of operation.

In order to achieve more uniform distribution of cavitational activity, Mason [1] has reported development of new piezoelectric transducer array using 1–3 composite (as shown in Fig. 4) with smaller area of irradiation. In this type of array, piezoelectric transducer pillars are embedded in pliable material which provides an additional facility to perfectly fit on any curved surface of reactor.

Cravotto et al. [64] have reported a modification in the conventional horn type reactor at irradiating frequency of near 20 kHz with maximum power rating of 1000 W, in order to enhance the substitution index (percent of alkylated glucosamine). In this system, the increase in the transducer temperature due to continuous use has been controlled below 40 °C by providing external cooling. The control of the temperature allows for continuous usage of the reactor, without any alterations in natural vibrating frequency, which has been a major problem in the operation of these reactors. The non uniformities in the distribution of cavitational activity have been eliminated by providing mechanical rotations of cylindrical reactor either clockwise or anticlockwise and linear motion (up and down) in vertical direction. The modified reactor configuration has been shown in Fig. 5. The reactor was rotated at a speed of 30 rpm in the radial direction whereas linear motion speed in vertical direction was set to 1.2 mm/s (maximum movement in the vertical direction has been restricted to 10 mm). The obtained results indicate that 86% substitution index (SI) was observed for formaldehyde in modified design in 3 h as compared to 60% in the conventional horn type reactor in 5 h. Similar augmentation was observed for other compounds like CH_3-CHO with significantly reduced reaction times. It should be noted here that sufficient exposure time with reasonable uniform distribution of cavitation events in liquid bulk results in enhanced chemical yield. In the improved design of Cravatto et al. [64] more efforts have to be applied on optimizing the dimensions of the reactor, operating parameters such as reactor volume and linear motion of the reactor. However, one may think, instead of moving the reactor from one position to other, similar effects can be obtained by introducing mechanical agitation with required speed of rotation. However, this possibility has also a possible detrimental effect as agitation strongly interferes with the passage of incident sound waves. Romdhane et al. [33] have measured the cavitational activity in axial plane in a cylindrical type of reactor and reported that there was a twofold decrease in the activity in the presence of agitation due to higher attenuation of sound wave from agitator surface and some hindrance in the generation and subsequent growth of cavitating bubbles.

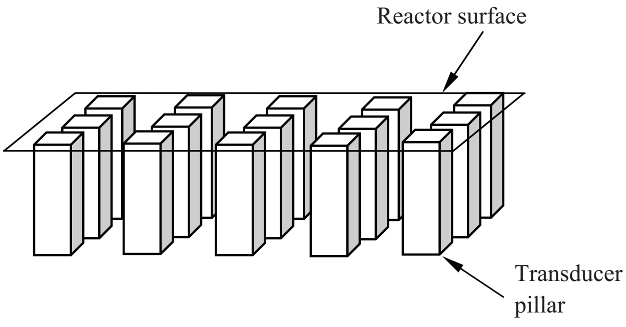

Fig. 4. Array of 1–3 composite transducers [1].

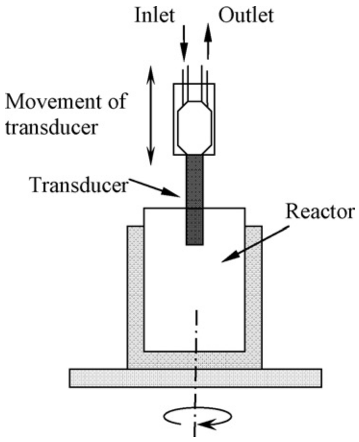

Fig. 5. Modified sonochemical reactor with mechanical means [64].

Another new design with proven efficacy for the large scale operation is the use of a single large longitudinally vibrating horn at the bottom of the sonochemical reactor. Bhirud et al. [36] have evaluated the energy efficiency based on calorimetric measurements in a sonochemical reactor equipped with longitudinally vibrating horn (diameter of 3 cm and length of 24 cm) of irradiating frequency 36 kHz and maximum power as 150 W. The internal dimensions of reactor were: length 0.15 m, height 0.20 m and width 0.33 m with maximum operating volume of 8 L and the schematic representation of the configuration is shown in Fig. 6. The obtained results showed that the maximum energy efficiency was obtained at operating capacity of 7 L (65.9%), which is much higher than the conventional ultrasonic horns. In addition, the cavitational yield (amount of formic acid degraded per unit supplied energy) was significantly higher (Table 3) as compared to the other conventional reactors [37]. Kumar et al. [65] have also quantified the distribution of cavitational activity in terms of the local pressure amplitude (using hydrophone) and the cavitational activity (using cavitation

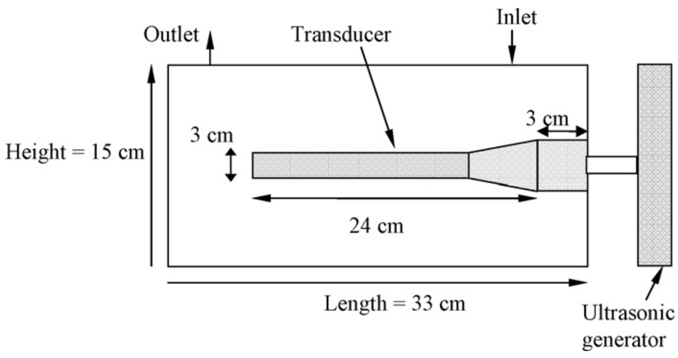

Fig. 6. Sonochemical reactor with longitudinal vibrating horn [36].

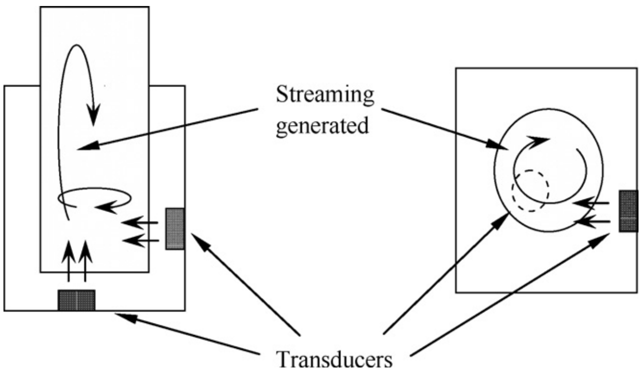

Fig. 8. Streaming generated in axial and radial direction [68].

Table 3
Comparison of cavitational yield in various types of sonochemical reactors [36,37].

S. No.	Type of reactor	Cavitational yield (mol/W)
1	Ultrasonic horn	0.0005
2	Ultrasonic bath	0.01
3	Dual frequency flow cell	0.011
4	Triple frequency flow cell	0.018
5	Longitudinally vibrating reactor	0.077

activity indicator) in axial as well as radial direction in the same reactor. The obtained results showed that the extent of cavitational activity was nearly uniform in both the direction with a variation in the range of 10–20% as compared to about 80–400% variation in the case of conventional designs.

Seymour et al. [66] have reported two reactor configurations which effectively utilized the reflection and focusing of sound wave in enhancing the performance of the reactor. Proper reflection patterns were obtained in the circular geometry by allowing sound wave to enter from sideway whereas proper focusing was obtained in cylindrical reactor by opting aluminium cap at the top of reactor as shown in Fig. 7. The performance of these reactors was evaluated by carrying KI oxidation studies (for concentration of 3.5%, w/v, of aqueous KI). It has been observed that rate of iodine liberation in reactor 1 and 2 was 2.5 μmol/W h and 4.8 μmol/W h, respectively, as compared to 0.07 μmol/W h (at 20 kHz) and 2.2 μmol/W h (at 900 kHz), respectively, as obtained in the other work [67].

In order to attain good mixing characteristics without employing any external agitation, Suria et al. [68] have effectively modified the design of sonochemical reactor by altering the locations of two transducers (of irradiating frequency as 1 MHz and 750 kHz and diameter of 20 and 50 mm, respectively). In the cylindrical configuration of the reactor, one of the transducers was positioned at the bottom located eccentrically, while the other was located on the periphery as depicted in Fig. 8. Due to this arrangement, acoustic streaming is generated in vertical and radial directions. The distance between the transducers and reactor can also be optimized to yield a significant reduction in the mixing time as compared to the conventional approach. The compatibility of this arrangement to large scale operation needs to be further optimized by having an optimum distance between the transducers, amplitude of sound wave, number of transducers and operating parameters like frequency and intensity of irradiation.

Use of multiple transducers allows achieving uniform cavitational activity as well as gives the flexibility to have multiple frequency irradiations, which can lead to higher cavitational intensity at similar levels of power dissipation. Gogate et al. [14] have reported the use of a triple frequency hexagonal flow cell as depicted in Fig. 9 with a total capacity of 7.5 L. The configuration contains total of eighteen traducers (three on each face of the hexagon) and seven different operating combinations of operating frequencies can be achieved. The performance of reactor was investigated using KI dosimetry and another industrially important reaction of degradation of a reactive dye, Rhodamine B. It has been reported that the cavitational yields and the energy efficiencies are much better as compared to the conventional reactor configurations [69]. Also detailed study in terms of distribution of cavitational activity (local pressure measurements using hydrophone and cavitational intensity measurements using cavitation activity indicator) in axial as well as radial direction [65] revealed that uniform distribution of cavitational activity in the bulk of liquid is obtained with only 10–30% variation as compared to 80–400% in the conventional horn type reactor configuration. Bal-

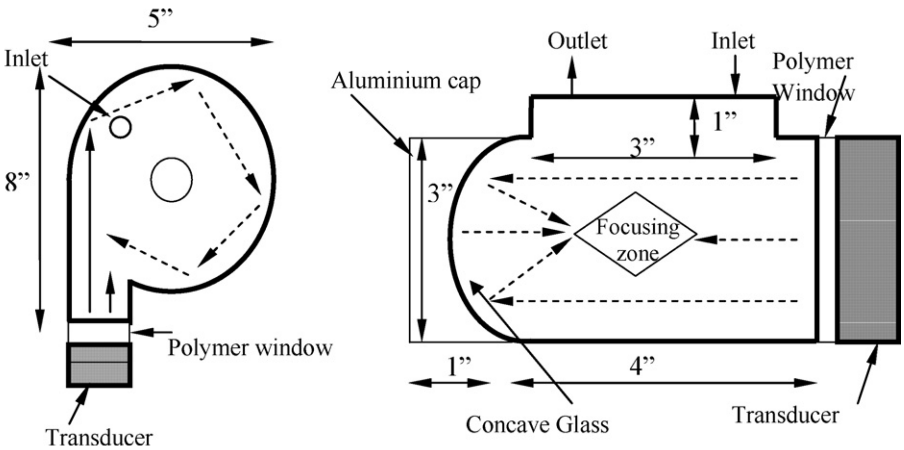

Fig. 7. Novel sonochemical reactors [66].

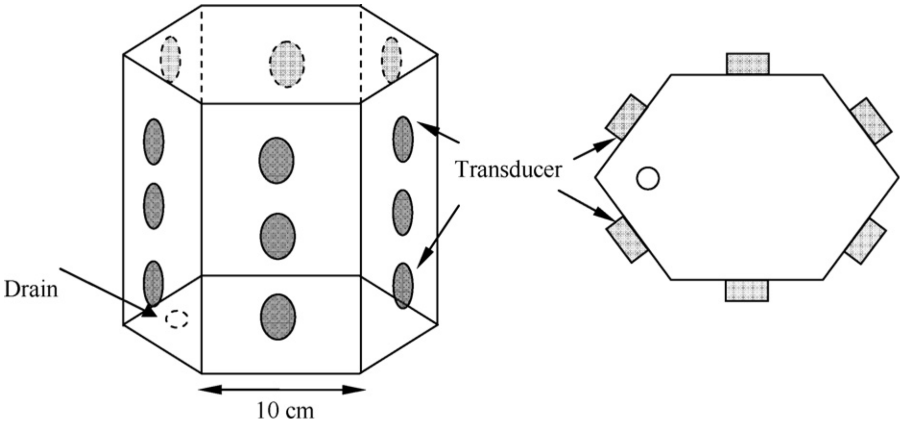

Fig. 9. Hexagonal triple frequency flow reactor [14].

asubrahmanyam and Pandit [70] have also reported similar results by measuring Aluminium foil erosion and obtained results for operations with single and triple frequencies have been shown in Fig. 10. It can be clearly seen from the erosion patterns that the cavitational activity is more uniform as well as intense in the case of triple frequency operation as compared to single frequency operation.

One of the concerns about reflection (for planar surface reflection is less) of sound in hexagonal flow reactor can be eliminated by adapting cylindrical geometry. Hodnett et al. [29] have reported the use of a cylindrical reactor (of diameter 312 mm and height 330 mm of operating capacity 25 L) with total of thirty piezoelectric transducers on outer periphery placed in ten rows with three in each row. The irradiating frequency is 25 kHz with a maximum power dissipation of 600 W. A schematic representation of the reactor has been given in Fig. 11. The advantage of this reactor is that variable power dissipation and variable frequency operation is indeed feasible.

Faid et al. [71,72] have quantified the cavitational activity in a novel sonochemical reactor (commercially known as Sonitube) of irradiating frequency 20 kHz and maximum power rating of 2000 W. A schematic representation of the reactor has been made in Fig. 12. The important feature of this type is that it can be used in both continuous (by passing fluid through resonators) and batch (by submerging emitter surface in container) modes of operation. The obtained results in batch mode of operation showed that uniform distribution of cavitational activity is obtained in the radial direction.

All the reactor configurations as discussed earlier were mostly with batch operation or even if continuous operation is possible the reported investigations were with batch operation. In the case of large scale operation, the feasibility of using a batch reactor is limited. We now discuss some of the configurations which have been applied in the continuous mode of operation.

Entezari et al. [73] have investigated sonochemical degradation of phenol in a Sonitube reactor operating at frequency of irradiation of 35 kHz. The reactor is a cylindrical stainless steel tube, with a 2 cm internal diameter and 26 cm length operated in the continuous flow mode. It has been reported that the effectiveness of the tube type reactor as compared to the other conventional reactors have been attributed to the uniform acoustic field, higher surface area of the sonicator and the geometry of the reactor. Concerning the acoustic field and geometry, Faid et al. [72] have studied the axial and radial profiles of mass transfer coefficients for a horn, a cup horn, and a tube emitter at 20 kHz. It has been reported that in the case of horn and cup horn, the maximum intensity (maximum transfer coefficient) is reached on the axis of the ultrasonic beam and decreases steeply on each side. In contrast, in the case of the tube, the value of transfer coefficient measured close to the tube wall was only slightly lower than along the axis, indicating a rather good radial homogeneity.

Gondrexon et al. [74] have investigated degradation of aqueous solution of pentachlorophenol (PCP) in a three stage continuous flow sonochemical reactor of irradiating frequency 500 kHz with maximum power dissipation as 100 W. In this reactor system, cylindrical geometry was subdivided into three units, each interconnected by polyvinylchloride pipe which functions similar to a downcomer in the conventional distillation column. The objective

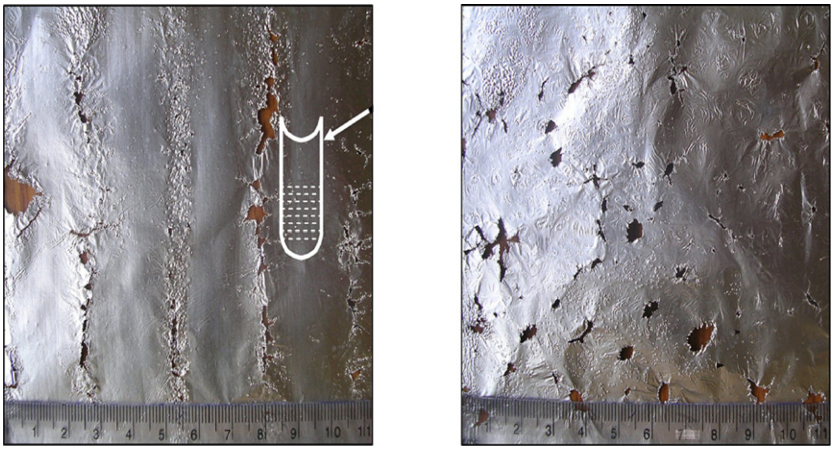

Fig. 10. Aluminium foil erosion patterns for single and triple frequency operation [70].

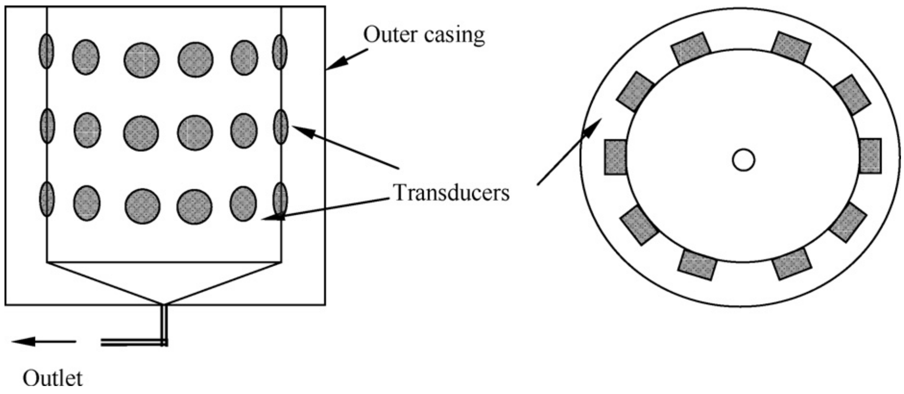

Fig. 11. Novel cylindrical reactor with multiple transducers [29].

of the connection is to give the desired direction to fluid and each unit was provided with a transducer of diameter 4 cm as shown in Fig. 13. The effect of variation in flow rate, operating volume and power dissipation on conversion of PCP in each unit has been quantified. The obtained result showed that, as the flow rate of streams increases at constant power dissipation rate, conversion decreases. The highest conversion was observed at an operating volume of 1×10^{-4} m^3, flow rate of 1×10^{-7} m^3/s and power dissipation of 80 W in unit 1 as 0.525 (in unit 2 and 3, conversion was 0.42 and 0.47, respectively) and lowest was 0.075 in unit 1 with a same power but at higher flow rate (10×10^{-7} m^3/s) and volume (3×10^{-4} m^3).

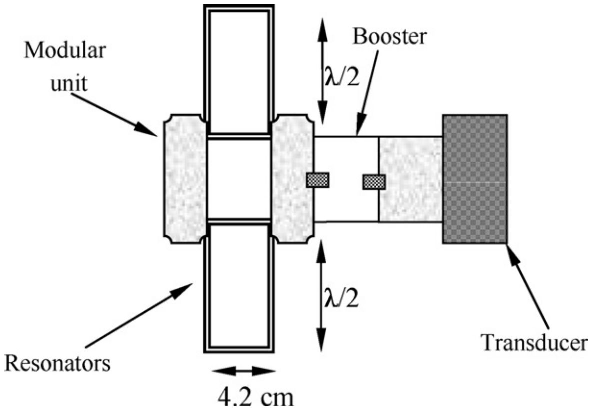

Fig. 12. Schematic representation of Sonitube [71].

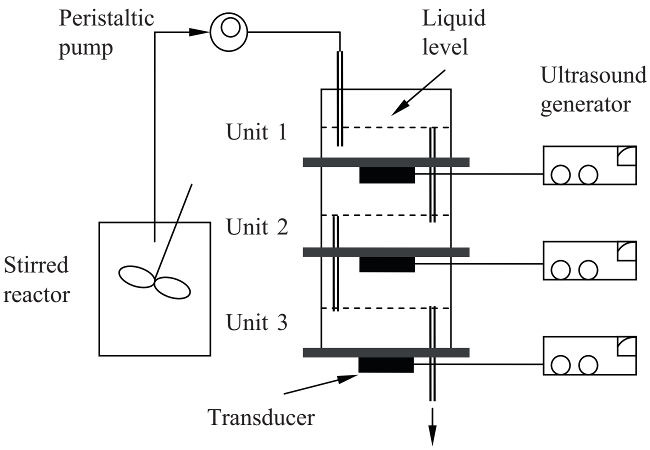

Fig. 13. A novel continuous scale sonochemical reactor with three stages [74].

Liu et al. [75] have investigated the degradation of dimethoate in ultrasonically assisted airlift loop reactor (UALR) in combination with ozone as oxidizing agent. The reaction system mainly consists of air loop reactor in which irradiating element (of frequency 40 kHz with maximum power rating of 250 W) was placed at the bottom of the reactor, as shown in Figure 14. Quantitative comparison of the degradation of dimethoate obtained using combined operation and using ozone and ultrasound separately, showed that, 90% degradation was observed in UALR as compared to 21% and 14.5% for only ozone and ultrasound, respectively. The optimum operating parameters as used in the work were reaction time of 4 h, 0.41 m^3/h flow rate of ozone, 4.64 W/cm^2 intensity of irradiation and pH of 10. This clearly indicates that effective utilization of the synergistic oxidative effects of cavitation and ozone and dispersion effects induced due to the configuration as air loop reactor, results in enhancing the performance of reactor. Similar type of efficacy was also reported in preparation of ε-caprolacton from cyclohexanone [76].

Nickel and Neis [77] have reported the use of a pilot scale continuous reactor for disintegration of biosolids for improving the biodegradation process. The sonoreactor is a vessel with 12 piezoceramic flat transducers fixed at each of the four sidewalls. The reactor volume is 1.3 L and the total rated power dissipation is 3.6 kW. Ultrasonic frequency is fixed at 31 kHz and acoustic intensities can vary within the range of 5–18 W/cm^2. Sonication is done in a pulsed mode (250 ms/s) while the biosolids were pumped in an upward flow through the square channel (45 mm × 45 mm) of the reactor. Based on the laboratory scale findings, a full scale design

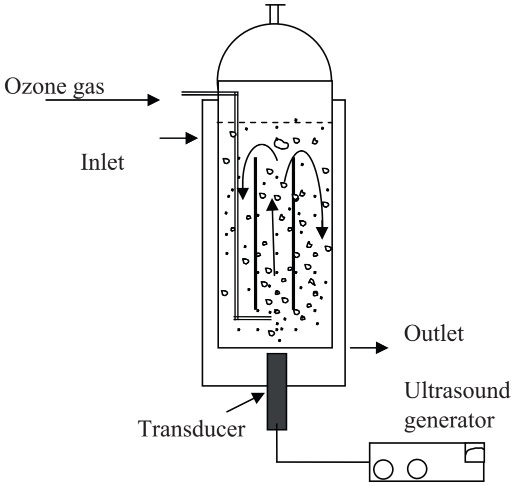

Fig. 14. Ultrasound assisted air loop reactor [75].

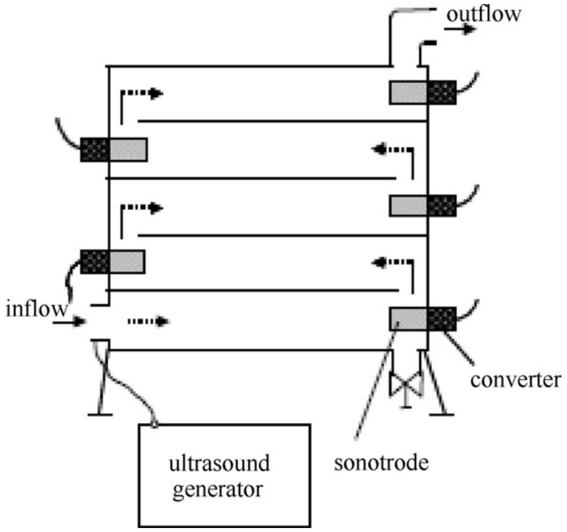

Fig. 15. Full scale sonochemical reactor for sludge treatment [77].

for sludge treatment applications has been developed. It has been reported that the design of full scale reactor should have capacity for treating large volumetric sludge flows, capacity to accept varying sludge properties (solids concentration, viscosity) and it is essential to remove the coarse and shaggy impurities prior to sonication to avoid reactor blocking. A full scale reactor configuration used successfully for biogas intensification applications has been shown in Fig. 15. The total capacity of the reactor is 29 L and is equipped with five 20 kHz sonotrodes. Each sonotrode is supplied by a 2 kW generator. The intensity can be adjusted in a range from 25 to 50 W/cm^2. The sludge is pumped upflow through the reactor channels, preventing accumulation of gas bubbles produced by degassing of the sludge water phase.

Very recently, Dion [78] has described a new continuous reactor design based on the high power converging acoustic waves in a tube to produce a relatively large volume confined acoustic cavitation zone in flowing liquid reagents under pressure. It has been reported that the new cylindrical sonoreactor design does not contaminate the processed liquids with erosion products since the cavitation zone is maintained away from the wall of the tube. The processing capacity of the largest models may be up to several tons per hour, depending on the required cavitation energy per unit volume to produce the desired process enhancement, using an electric power input of about 50 kW.

8. Qualitative considerations for reactor choice, scaleup and optimization

It is important to give some firm recommendations for selection of different design and scale up parameters as well as to check whether it is really required to use a sonochemical reactor as the cost of operation may not be always favorable. When deciding on the type of the reactor required for a particular chemical or physical transformation, the first question that needs to be addressed is whether the cavitation enhancement is a result of an improved mechanical process (due to enhanced mixing). In such case, cavitation pretreatment of a slurry may be all that is required before the system is subjected to conventional type transformation scheme and the scale up of the pretreatment vessel would be a relatively simpler task. If, however, the effect is truly based on the cavitation chemistry, then cavitation must be provided during the transformation itself either operated in continuous manner or in suitable pulsed operation. The scientific database from the laboratory study typically provides the most effective parametric window and the cause–effect-relationships between the operating parameters and the observed cavitational effects. This also involves generating design correlations for the prediction of the cavitational yield as a function of different operating parameters. These design equations will help in selection of the operating parameters to achieve desired level of transformations. The first step is to try to understand the mechanisms of interaction from the observed phenomena so that the desired cavitation field can be created on a larger scale to promote similar interactions. The important scale up consideration is then to establish the optimum conditions for the transformation in terms of the operating/design variables that influence cavitation. In this analysis, the nature of the transformation will also have an impact on the suitability of the given cavitational reactor.

Both chemical and physical properties of the reaction medium will dictate the required level of cavitation power. High viscosity media with low vapor pressure will require higher energy to generate cavitation. The presence of entrained or evolved gases will facilitate cavitation, as will the presence or generation of solid particles.

An efficient coupling of the acoustic energy to the material that will provide a transmission path for the ultrasonic energy is also very important. This is usually a major step and requires a thorough understanding of the nature of wave propagation (either in high frequency or multiple frequency multiple transducer application) and radiation from pipe, plate or channel shaped sonochemical reactors. Though a detailed discussion related to these issues is beyond the scope of the present work, the important factors that need to be analyzed have been illustrated below:

- Mechanisms of cavitation and their interaction with the reactor material.
- Wave propagation in structures.
- Acoustic coupling and mode of excitation.
- Transducers and power generator technology.
- Integration of ultrasonics into the process system.

Mason and Cordemans de Meulenaer [79] have also given the following 10 recommendations/steps in the optimization of an ultrasonic process.

(1) Make cavitation easier by the addition of solids or gas bubbles to act as nuclei.
(2) Try entraining different gases or mixture of gases.
(3) Try different solvents for different temperature ranges and cavitation energies.
(4) Optimize the power required for the reaction.
(5) When using a solid–liquid system, do not charge all the components in the reactor at once.
(6) If possible, try to homogenize two-phase systems as much as possible.
(7) Try different shapes (diameters and volumes) for the reactor.
(8) It can be better (but not always) to avoid standing wave conditions by performing sonochemical reactions under high power conditions with mechanical stirring.
(9) Where possible, try to transform a batch system into a continuous one.
(10) Choose conditions, which allow comparisons between different sonochemical reactions.

Overall economic analysis is also an essential requirement to compare the costs associated with the operation against the possible benefits that would be encountered from the selection of sonochemical reactors. Usually it has been observed that the processing costs of sonochemical reactors are on a higher side as compared to conventional techniques or processes with similar mechanism [1,80,81]. Mahamuni and Adewuyi [81] have recently analyzed the costs of different advanced oxidation processes for treatment of phenol and it has been shown that cost of using ultrasound alone is much more cost intensive operation as compared to photocatalytic oxidation for equivalent degradation of phenol from the effluent. It thus becomes very important that the obtained benefits outweigh the possible enhanced costs, at least for some applications where other cost effective alternatives exist, before the selection of sonochemical reactors can be recommended. A standing example where use of sonochemical reactors is recommended is enhanced crystallization efficiency for pharmaceutical derivatives where the final particle size distribution can be tailored based on the optimum use of the sonochemical reactors in terms of power dissipation and the time and stage of the ultrasonic treatment [82]. A possible combination of the sonochemical reactors with other processes [83–85] such as photocatalytic oxidation, microwaves, etc. can also result in enhancing the productivity of the processes and reducing the energy requirements for equivalent production capacities.

9. Conclusions

Maintaining uniform distribution of cavitational activity is one of the most important design aspects in effective scale up of the sonochemical reactors and can be optimized by making a proper choice of the operating (frequency of irradiation, intensity of irradiation and operating power dissipation per unit volume) and geometric parameters (reactor configuration and number and location of the transducers). Liquid phase physicochemical properties also play important role in controlling the number of cavitational events as well as the resultant cavitational intensity. In addition, the other important design aspects, which also need to be maximized for better efficacy, include hydrodynamics, mixing time and mass transfer characteristics. A logical analysis of the existing literature in terms of effect of different parameters on these design aspects have lead us to following important design related information:

1. The frequency of irradiation should be selected depending on the desired effects for the specific application, viz. higher frequency or combination of lower frequencies is recommended for application dominated by chemical effects and lower frequency is recommended for applications where the physical effects are controlling.
2. Optimum power dissipation levels (W/m^3) needs to be maintained for the given physicochemical transformation, an idea of which can be obtained from the laboratory scale studies or similar studies in the literature. Development of design correlations is required which may be generalized considering the different controlling factors for the said application. It would not be possible to dissipate same power through a single transducer at larger scales of operation and hence it is advisable to use number of transducers with as large irradiation surface as possible to ensure lower operating intensity. Using larger irradiation surface also gives an enhanced cavitationally active volume.
3. It is important to select liquids with low vapor pressure, low viscosity and higher surface tension so as to maximize the cavitational events. Presence of impurities or additional phases either in the form of solids or gases, under optimized conditions, can also influence the cavity generation process and overall cavitational effects in a favorable manner.
4. Flow behaviour in the sonochemical reactor plays a crucial role in selecting the dimensions of reactor and location of transducers. An optimized reactor configuration should give the required mixing effects especially for heterogeneous systems and adjustment of operating parameters mainly the power dissipation levels is required to ensure the presence of requisite hydrodynamic behaviour and mixing characteristics.
5. Similar to mixing time, suitable levels of power dissipation and lower frequencies of irradiation are recommended to ensure required levels of mass transfer rates are obtained in the larger scales of operation. Additional work is required for quantifying the mass transfer as well as the mixing characteristics for multiple frequency multiple transducer operation as such data is lacking in the existing literature.
6. It is important to consider the attenuation effects while estimating the requirement of power dissipation levels in the large scale reactor and the guidelines offered in the work would be useful in estimation of the attenuation coefficient for the specific application under question.
7. Based on the analysis of different advancements in the large scale operation, it can said that development of continuous reactors with tubular or hexagonal geometry is the key to effective large scale operation and use of multiple transducers with a possibility of multiple frequency operation is recommended to get required cavitational effects also minimizing the required energy consumption.

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