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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