

# Cavitation Reactors: Efficiency Assessment Using a Model Reaction

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*Acoustic and hydrodynamic cavitation can be used for a variety of applications ranging from biological applications such as cell disruption to chemical reactions such as oxidation of organic pollutants in aqueous effluents, including biorefractory toxic chemicals. Different equipment used for cavitation effects was compared based on a model reaction (decomposition of potassium iodide resulting into iodine liberation). A correlation was developed for the prediction of the cavitation yield in terms of the cavity collapse pressure. This correlation, when used with earlier correlations for the pressure amplitude generated during the violent collapse of cavities, will help design engineers to choose particular equipment, operating conditions, and geometric parameters to achieve a desired chemical change. The developed equation relating the macroscopic reaction rates with the collapse pressure is the first of its kind reported in the literature. Pilot-plant-scale hydrodynamic cavitation orifice plate setup is most energy-efficient, with significantly higher cavitation yields for the model reaction.*

## Introduction

Physical and chemical transformations using cavitation phenomenon is a well established concept. The obvious advantage of these processes is based on the fact that reactions can be carried out under ambient global conditions, which would otherwise require the application of rigorous conditions such as high temperature and pressure. The underlying mechanism for these spectacular effects of cavitation is the violent collapse of microbubbles or cavities, resulting in the generation of extremely high temperatures and pressures locally, however, this would take place at millions of locations in the reactor. The violent collapse of the cavities also results in the formation of reactive hydrogen atoms and hydroxyl radicals, which combine to form hydrogen peroxide, and to some extent are responsible for promoting oxidation reactions.

Generally, cavitation has been classified into four types: acoustic cavitation, hydrodynamic cavitation, optic cavitation, and particle cavitation depending on the mode of generation. However, only acoustic and hydrodynamic cavitation have

been of academic and industrial interest, due to the ease with which the required intensities of cavitation conditions can be generated. Some of the applications of cavitation are degradation of potassium iodide (Naidu et al., 1994; Suslick et al., 1997; Senthilkumar et al., 2000); hydrolysis of fatty oils (Pandit and Joshi, 1993); degradation of organic waste in the effluent (Shirgaonkar, 1997; Hua and Hoffmann, 1997; Weavers et al., 1998; Hung and Hoffmann, 1999; Kalumuck and Chahine, 1998; Sivakumar and Pandit, 2001a,b; Gogate, 2001; Pandit et al., 2001); biological applications such as cell disruption (Harrison and Pandit, 1992; Save et al., 1994, 1997; Gareth and Danver, 1996; Balasundaram and Pandit, 2001), and water disinfection (Jyoti and Pandit, 2001).

The variety of equipment used for generating acoustic and hydrodynamic cavitation is enormous and a unified criteria for the comparison of such equipment is missing in the literature. As the energy required for bringing out the chemical transformation based on the cavitation phenomena is of paramount importance, it is desirable to compare the various equipment based on this criteria. Therefore, a rational procedure must be developed for the selection of

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equipment for any particular reaction based on the specific energy consumption for bringing about the desired degree of chemical change. Attempt has been made to compare the different equipment generally used for the cavitation based transformations, such as the ultrasonic horn, ultrasonic bath, hydrodynamic cavitation setup using multiple orifices, and high speed and high-pressure homogenizer, using a unified criteria of cavitation yield based on the total energy consumption and the energy efficiency of the system. Such a comparison will definitely help the engineers and chemists to choose a particular type of equipment for the numerous uses of cavitation.

Moreover, the magnitude of pressures generated by the collapse of cavities depends on operating conditions such as the intensity and frequency of irradiation, initial size of the nuclei in the case of acoustic cavitation or inlet pressure, diameter of the orifice, and percentage free area for the flow in the case of hydrodynamic cavitation. In earlier work, the magnitude of pressure pulses generated during the cavitation process was correlated as a function of the previously mentioned parameters for the acoustic cavitation (Gogate and Pandit, 2000a) and for hydrodynamic cavitation (Gogate and Pandit, 2000b). The cavitation yield from the reactor is a function of the magnitude of the pressure pulse generated during the cavitation event, and a mathematical relationship must be established between the two in order to facilitate efficient design of cavitation reactors. Once such a relationship is established, using the earlier relationships for collapse pressures as a function of operating parameters, one can estimate the yields for the particular process under the given operating conditions without actually performing the experiments. In the present work, attempts have also been made to correlate the cavitation yields for the model reaction of decomposition of potassium iodide liberating iodine. It should also be noted that the established relationships will be valid for the particular reaction considered and, therefore, pilot-plant studies are necessary for the same purpose, for the reaction to be carried out at a larger scale.

### *Acoustic and hydrodynamic cavitation*

In acoustic cavitation, the global pressure variations in the liquid is effected using the sound waves, usually ultrasound (16 KHz–100 MHz). The chemical changes taking place due to the cavitation induced by the passage of sound waves are commonly known as sono-chemistry. The acoustic cavitation, or sonochemical processes, have been the widely studied phenomenon over the past few decades. Excellent reviews on the scope and the application of ultrasound, and processes based on the same, are available in the literature (Mason, 1986; Henglein, 1995; Lindley and Mason, 1987; Luche et al., 1989; Moholkar and Pandit, 1996; Mason, 1999; Suslick et al., 1999; Von Sonntag et al., 1999; Keil and Swamy, 1999; Shah et al., 1999). Modeling of sonochemical reactors and the bubble dynamics under a variety of conditions have also been extensively studied in the past (Yan et al., 1988; Yan and Thorpe, 1990; Kamath et al., 1993; Naidu et al., 1994; Moholkar and Pandit, 1997; Sochard et al., 1998; Moss et al., 1999; Storey and Szeri, 1999; Gogate and Pandit, 2000a). However, it should be noted that in spite of extensive research, there is hardly any chemical processing carried out on an industrial scale

owing to the lack of expertise required in such diverse fields as material science, acoustics, chemical engineering, and so on, for scaling up successful lab scale processes. Some attempts have been made to effectively scale up these reactors (Berlan and Mason, 1992; Martin and Ward, 1993).

Hydrodynamic cavitation can simply be generated by the passage of liquid through a constriction such as an orifice plate. When the liquid passes through the orifice, the kinetic energy/velocity of the liquid increases at the expense of the pressure. If the throttling is sufficient to cause the pressure around the points of *vena contracta* to fall below the threshold pressure for cavitation (usually vapor pressure of the medium at the operating temperature), millions of cavities are generated. Subsequently, as the liquid jet expands, the pressure recovers, and this results in the collapse of the cavities. During the passage of the liquid through the constriction, boundary layer separation occurs and a substantial amount of energy is lost in the form of permanent pressure drop. Engineers have generally looked with caution at cavitation in hydraulic devices due to the problems of mechanical erosion. Initial efforts to understand it were geared toward the objective of suppressing it, to avoid the erosion of exposed surfaces. In the last decade, concentrated efforts were made by a few groups around the world to harness the spectacular effects of hydrodynamic cavitation for chemical/physical transformation (Chivate and Pandit, 1993; Pandit and Joshi, 1993; Save et al., 1997; Suslick et al., 1997; Vichare et al., 2000; Kalumuck and Chahine, 1998; Senthilkumar et al., 2000; Gogate and Pandit, 2001; Jyoti and Pandit, 2001; Sivakumar and Pandit, 2001b).

### **Theoretical Framework for Acoustic and Hydrodynamic Cavitation**

The bubble behavior during the cavitation phenomena is a strong function of the operating conditions and geometric parameters, and affects the magnitude of pressure pulse generated at the end of cavitation event. These conditions of high temperatures and pressures are responsible for the desired chemical change and, therefore, the degree of these conditions depends on the magnitude of the pressure pulse generated. The bubble dynamics have been studied in detail in earlier work (Gogate and Pandit, 2000a,b) using the Rayleigh-Plesset equation and the equations given by Tomita and Shima (1986), considering the compressibility of the medium and the magnitudes of pressure pulse have been predicted as a function of different parameters. The empirical correlations obtained for the pressure pulse have been reproduced here as follows:

#### *Acoustic Cavitation*

$$P_{\text{collapse}} = 114(f)^{0.11}(I)^{-0.17}(R_o)^{-1.88}$$

where  $P_{\text{collapse}}$  is the magnitude of pressure pulse generated,  $f$  is the frequency of irradiation,  $I$  is the intensity of irradiation, and  $R_o$  is the initial size of the nuclei.

$$P_{\text{collapse}} = 7,527(A)^{-2.55} \{ (P_i)^{2.46} (R_o)^{-0.8} (d_o)^{2.37} \}$$

where  $A$  is percentage free area offered for the flow,  $P_i$  is the inlet pressure into the system, and  $d_o$  is the diameter of the orifice.

The details of these variations of the pressure pulse with the given characteristic parameters, in terms of the possible explanation and experimental support, have been described in earlier articles (Gogate and Pandit, 2000a,b).

However, it should be noted, that the preceding correlations give just an indication of the magnitude of pressure pulse generated in the cavitation reactors. The effects of cavitation may be different, depending on the type of the specific reaction to be carried out in the reactor. The quantitative information required to use the magnitude of pressure pulses in predicting the microscopic and macroscopic rates of specific reactions, is lacking in the literature. The information about the formation of intermediate species and their role in the formation of the desired product has not been elucidated at all in the literature. The only publications in this area are by Naidu et al. (1994), Sochard et al. (1998), and, more recently, by Storey and Sezri (1999). As mentioned earlier, establishing the relationship between the macroscopic rates and the magnitudes of collapse pressures, although by empirical means, with which the design engineers are comfortable is the objective of this article.

## Experimental Studies

The various equipment used in the present work is discussed, with a concise description of each. The Weissler reaction, which has been used as a model reaction quite extensively in the past (Shirgaonkar and Pandit, 1997; Suslick et al., 1997; Senthilkumar and Pandit, 1999; Senthilkumar et al., 2000; Vichare et al., 2000), has been described briefly for a better understanding of its use in quantitatively evaluating the cavitation phenomena.

### Weissler reaction

Weissler reaction is the reaction of decomposition of **KI** liberating free iodine. It should be noted at this stage that the Weissler reaction is only induced due to the cavitation and not by shear temperature and pressures (Suslick et al., 1997; Senthilkumar et al., 2000). This is due to the fact that free OH radicals are formed in the solution only under cavitating conditions. These free radicals attack **KI** liberating iodine. Iodine also reacts with some amount of remaining **KI** to form  $I_3^-$  ions which form a blue colored complex when starch is added to the solution. The extent of iodine liberated during the reaction is estimated with the help of a UV/VIS spectrophotometer by measuring its absorbance at 355 nm wavelength. The rate and amount of the iodine liberation depends on the formation and supply of OH radicals, which are dependent on the efficacy of the cavitating equipments. Therefore, decomposition of **KI** (Weissler reaction) can be effectively used for comparing various equipments used for generation of cavitation and establishing mathematical rela-

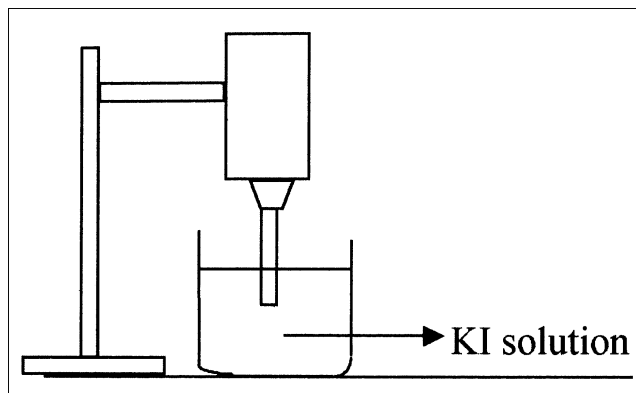


Figure 1. Ultrasonic horn.

tionships between the overall macroscopic rates and the intensity of cavitation.

If starch is added during the sonication, destruction of starch may take place giving rise to a pink color instead of the conventional blue, due to the combination of the starch molecule degradation products with iodine ions. Therefore, addition of starch to the experimental solution should be done only at the end of the experiment or just before doing the analysis. Also, care should be taken in the preparation of the starch solution. In the experimental method, starch solution was prepared by dissolving 1 g of soluble starch in 100 mL distilled water, which is then boiled for 15 min. The starch solution must be used for the analysis only after cooling to room temperatures and should be clear in appearance (this may require filtration).

### Equipment for acoustic cavitation

**Ultrasonic Horn.** An ultrasonic horn is depicted in Figure 1. For the present study two ultrasonic probe (horn) systems have been used. The first horn was procured from a local manufacturer M/s. Dakshin and has been identified as the Dakshin horn throughout the investigation. The Dakshin horn operates at a frequency of 22.7 kHz. The rated power dissipation is 240 W through a horn area of about 81 mm<sup>2</sup>. The radiating body is made of stainless steel. It has a detachable tip which can be removed for further polishing after it has been eroded due to sonication. It can be operated in continuous mode for 15 min. However, this results in heating of the horn and, hence, it should be cooled before the next operation.

The second horn was procured from Ace Glass Inc. (U.S.A.) and operates at a fixed frequency of 20 kHz with a rated output power of 600 W. The transducer is made up of lead zirconate titanate. The operation of the horn is controlled by a microprocessor. It is operated in a pulse mode. A pulse facility consists of a timer which switches the power to the probe on and off repeatedly. The off time allows the chemical system and the horn to cool between the pulses of sonication. The pulse mode can be operated at an on/off ratio of 9.9:0.1. The equipment also gives the liberty of varying the intensity in terms of the oscillation amplitude, depending on the type of horn tip (irradiating area) used (maximum 40%

allowed for a microtip). There is also the facility of tuning the horn. Tuning of the probe system is generally done to get optimum performance. Tuning is a process whereby the transducer (normally operating at 20 kHz) is brought into resonance with the vibrating tip reducing the transmission losses. This is a point where minimum electrical power is drawn during operation and delivers maximum power into the system. Tuning is performed because of shortening of the tip due to the erosion which results in a change in its resonant frequency. Tuning is generally carried out in air. The experiments with an ultrasonic horn were performed with a reaction volume of 50 mL and at the room temperature which is usually in the order of 28–30°C.

**Ultrasonic Bath.** The ultrasonic bath was procured from M/s Dakshin, and has been reported as Dakshin bath throughout the study. It has a fixed frequency of 22 kHz with a rated output power of 120 W. The internal body is made of steel below which two transducers are attached at the corners of a rectangular base. The bath has the dimensions of 15 × 15 × 15 cm. Drainage has been provided at the side of the bath. The energy to the transducer is provided by a processor which is a separate unit. The volume of the reaction medium used in the case of the ultrasonic bath was 500 mL. Representation of an ultrasonic bath is depicted in Figure 2.

**Flow Cell.** The flow cell (Figure 3) consists of a rectangular vessel with a diameter of 9.5 cm and a height of 20 cm (1.5 L capacity to hold the reacting mixture) with two sets of transducers (3 in each set) mounted on the two opposite faces. Transducers operating (independently or simultaneously) at

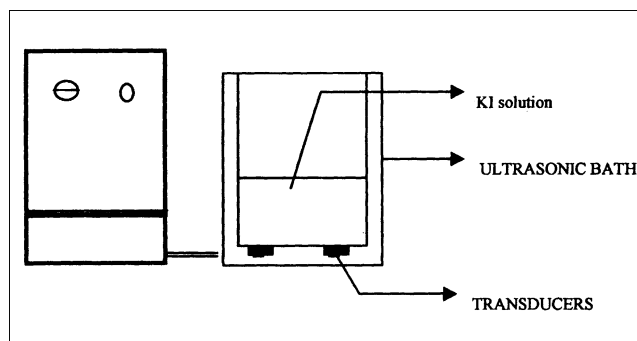


Figure 2. Ultrasonic bath.

different frequencies, that is, 25 and 40 kHz and with equal power rating of 120 W per set have been provided. The flow cell can be operated in a batch or a continuous mode.

#### Equipment for hydrodynamic cavitation

**High-Pressure Homogenizer (HPH).** The high-pressure homogenizer (APV Gualin GmbH model) is basically a high-pressure positive displacement pump with a throttling device (Figure 4). This homogenizer operates according to the principle of high-pressure relief technique. The HPH used here consists of a feed tank and two throttling valves designated as first stage and second stage. The liquid from the feed tank (capacity of 1,500 mL) is driven by a pump to the first-stage valve and pressure up to 1,000 psi can be attained by throt-

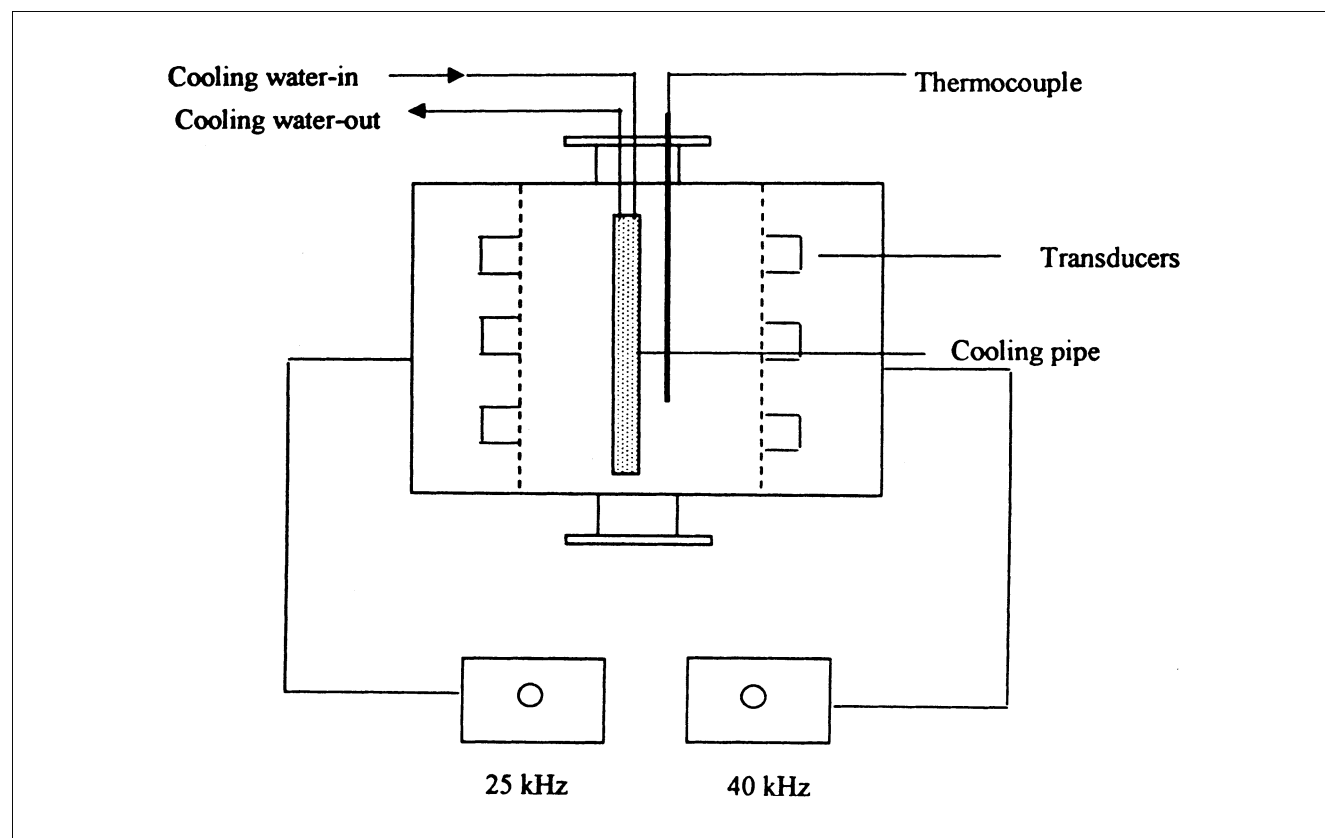


Figure 3. Dual frequency flow cell.

ting this valve. Further increase in pressure is achieved by using the second-stage valve. Upstream pressure up to 10,000 psi can be obtained in the second stage. From the second-stage valve, the liquid is recirculated back to the feed tank. The cavitating conditions are generated just after the second-stage throttling valve. When the liquid is suddenly released from second stage, evaporation takes place giving rise to cavities/bubbles. The cavitation intensity will be dependent on the magnitude of upstream pressure and also on the type of valve at the second stage. With an increase in throttling pressure, there is a rise in the temperature of the liquid. To maintain the temperature at ambient conditions, a coil immersed in the feed tank was used through which cooling water was circulated. HPH are especially suitable for the emulsification processes in the food, chemical, pharmaceutical, biochemical industries.

**High Speed Homogenizer.** The high speed homogenizer consists of an impeller and a stator both of which are made up of stainless steel. The impeller is driven by a variable voltage motor (the limit permitted for the homogenizer is 30 V or 3.5 A, resulting in a maximum rotational speed of 12,000 rpm). The impeller blades are 6 mm apart whereas stator blades are also 6 mm. The distance between the OD of the impeller blade and the ID of stator blades is 2 mm. This distance can be varied by using different impellers and stators. The combination used in this study has an impeller with 9 blades while the stator has 13 blades. A plate with holes attached to the stator top has been provided which can be used for inserting baffles, so as to avoid vortex formation and

surface aeration which decreases the intensity of cavitation. The cavitating conditions are generated after the liquid passes through the stator rotor assembly (Senthilkumar and Pandit, 1999) according to principle similar to the orifice plates setup which is described later. As one increases the rotor speed, the liquid velocities generated increase, and, beyond a certain speed defined as the critical inception speed for the cavitation, cavities are formed due to fact that local pressure falls below the vapor pressure of the medium.

**Orifice Plates (Pilot-Plant Scale).** The setup consists of a closed-loop circuit comprising of a holding tank of 50 L volume, a centrifugal pump (2,900 rpm, 5.5 kW, Calama Industries Ltd., India), and a control valve and flanges to accommodate the orifice plates shown in Figure 5. The suction side of the pump is connected to the bottom of tank. The discharge from the pump branches into two lines which helps in the control of the inlet pressure and the inlet flow rate into the main line housing the orifice with the help of valves  $V_2$  and  $V_3$ . The main line consists of a flange to accommodate the orifice plates (single or multiple holes), along with a hard glass tube next to these plates to make a visual observation. The cavitating conditions are generated just after the orifice plates in the main line. When the liquid passes through the orifice plates, due to the sudden reduction in the area offered for the flow, the velocities at the orifice increase, resulting in a decrease in the pressure. If the velocities are such that the increase in velocity is sufficient to allow the local pressure to go below the medium vapor pressure under the operating conditions, cavities are formed. Such cavities are

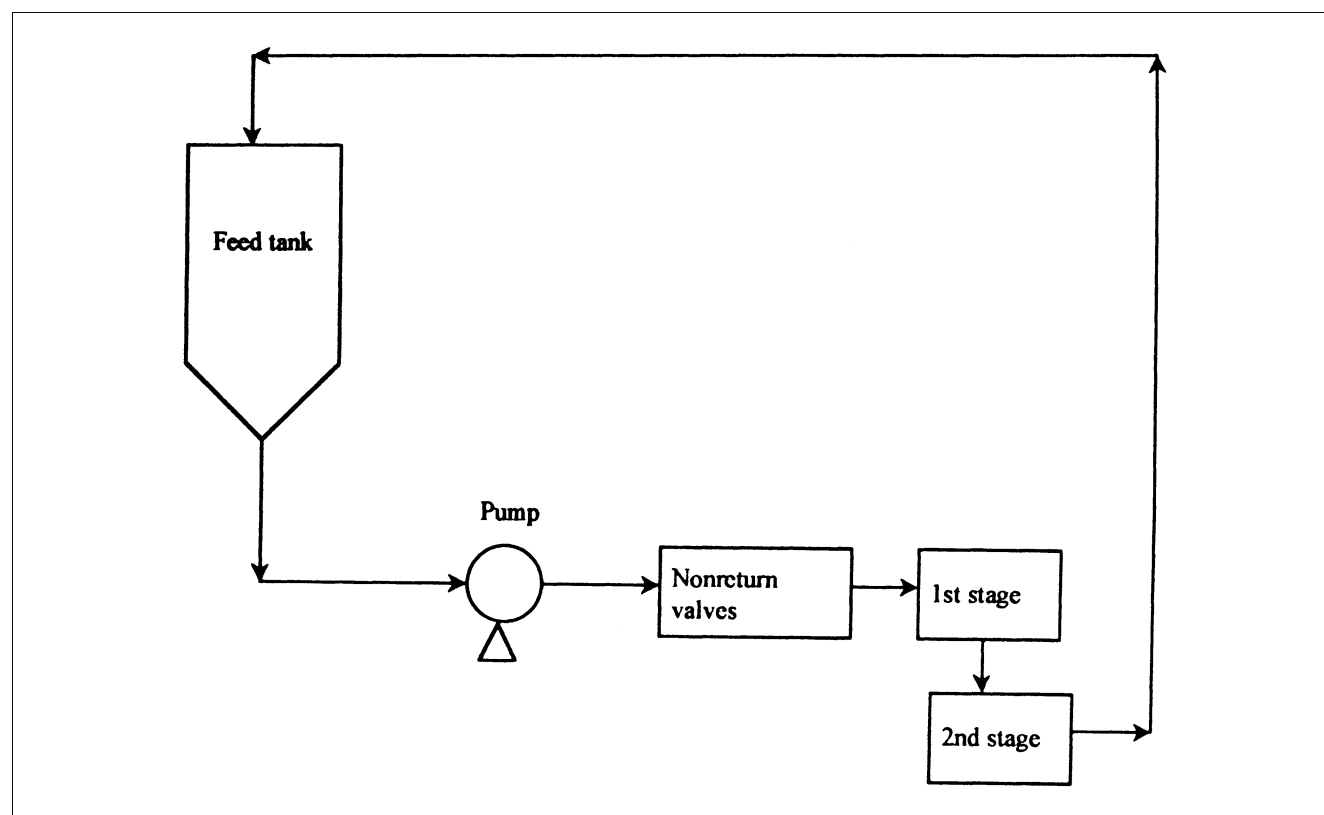
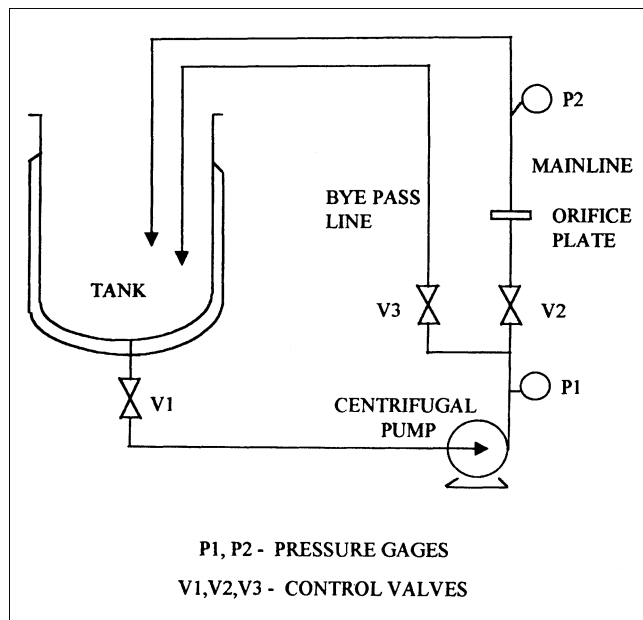


Figure 4. System for high-speed and high-pressure homogenizer.



**Figure 5. Hydrodynamic cavitation reactor setup using orifice plates (pilot-plant scale).**

formed at a number of locations in the reactor which also depends strongly on the number of holes in the plates. However, downstream of the orifice, due to the increase in the

area of cross-section, the velocities go on changing giving rise to pressure fluctuations which control the different stages of cavitation: namely, formation, growth, and collapse. The holding tank is provided with a cooling jacket to control the temperature of the circulating liquid. The inlet pressure and the fully recovered downstream pressure can be measured with the pressure gauges  $P_1$  and  $P_2$ , respectively. The holding capacity of the tank used in the experimentation is 50 L.

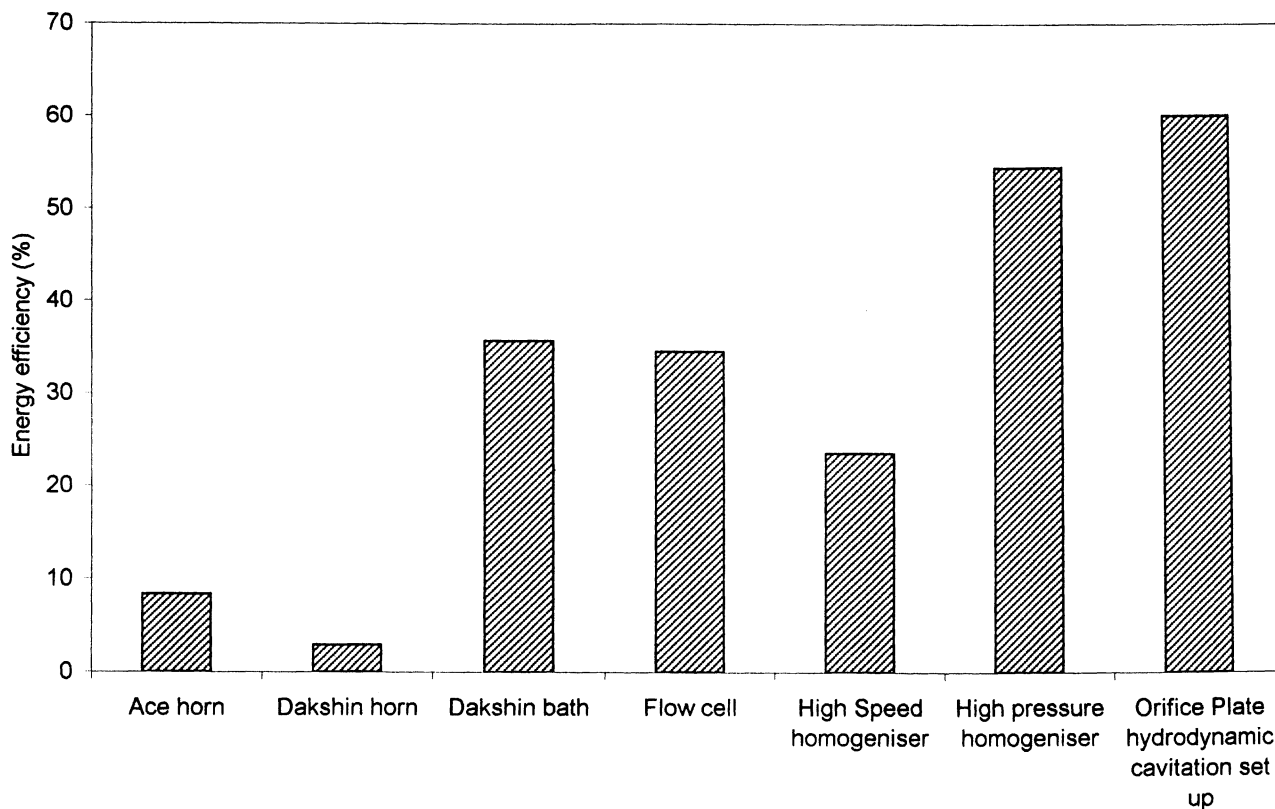
### Calculation methodology

The methodology used for the estimation of cavitation yield and the energy efficiency of the system in order to compare the various equipments is discussed.

Calorimetric methods are used to determine the energy efficiency of the equipment under study. The rise in temperature of a fixed quantity of water in an insulated container for given time was measured. Using this information, the actual energy (power) dissipated into a liquid was calculated from the following equation

$$\text{Power (W)} = m C_p (dT/dt) \quad (1)$$

where  $C_p$  is the heat capacity of the solvent ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ ),  $m$  is the mass of solvent (kg),  $dT$  is the temperature difference between the initial temperature and the final tempera-



**Figure 6. Energy efficiency of various equipment.**

ture after a specific reaction time ( $K$ ), and  $dt$  is time ( $s$ ). Energy efficiency can then be calculated as follows:

Energy efficiency = Power dissipated in the liquid/Electric power supplied to the system.

Energy efficiency gives an indication of the quantity of energy effectively dissipated in the system, a fraction of which is utilized for the generation of cavities and should be as high as possible for the particular equipment.

The second parameter which has been used for the characterization of the cavitation reactors is the cavitation yield which is given by the following correlation:

Cavitation yield

$$= \frac{\text{desired chemical change (iodine yield in g/lit)}}{\text{power density}} \quad (2)$$

where the power density is defined as the energy supplied (actual electrical energy) to the medium per unit volume of the medium. The cavitation yield for the acoustic/hydrodynamic equipment indicates the ability of the equipment in producing the desired change based on the electric energy actually used for generating cavitation.

The aim of the designers should be to maximize both the energy efficiency, as well as the cavitation yield of the reactor. This can be done on the basis of manipulation of the operating conditions and geometric parameters of the reactor, resulting in required magnitude of pressure pulses from the cavitation events to give the desired effect in terms of the observed chemical change.

## Results and Discussion

### Energy efficiency

In all the above equipment, the primary form of the energy supplied as input is the electrical energy. This energy goes through various changed forms, that is, pressure, velocity, vibrations, and so on, before it is used to generate cavitation, resulting in the desired chemical change. In most cases, the electrical energy is converted to mechanical energy (vibratory motion of the transducer in acoustic equipment and liquid flow with pressure in the hydrodynamic cavitation equipment). The mechanical energy is used to generate cavitation, and, finally, the violent collapse of generated cavities dissipate this diffused form of energy by concentrating it through a number of cavitation events and induce the chemical reactions. In each case, there is a continuous loss of energy during its transformation from one form to another, and, hence, the comparison of the energy efficiency can aid in understanding the productive energy utilization of various equipment for the desired cavitation activity. The mechanical energy generated from the electric power consumption is utilized in two useful forms, that is, it is either used for the chemical reaction or it appears as the heat energy which results in the increase in the temperature of the medium. The energy used for the actual chemical reaction is negligible; therefore, the electrical energy supplied finally appears as the heat energy, as it is a closed-loop system. Hence, the energy efficiency calculation based on the heat energy dissipated in the system is justified.

The results of the study for acoustic and hydrodynamic cavitation equipment are given in Figure 6. From the figure, it is observed that among the acoustic equipment, the ultrasonic bath is the most energy efficient (energy efficiency = 43%) due to the uniform energy dissipation over a wider area rather than the concentrated energy dissipation in both the horns (energy efficiency < 10%). The efficiency of flow cell ( $\approx 35\%$ ) was also found to be equally good again due to the fact that the energy is dissipated over a wider area. This observed increase in the energy efficiency for equipment with a higher irradiating surface (lower intensity of irradiation) can be explained on the basis of the decoupling effect. There is an optimum intensity below which there is no significant contribution of the decoupling effect, and the sound energy absorbed by the medium is proportional to  $I^{1/2}$  where  $I$  is the intensity of irradiation. If one operates nearer to the optimum intensity, there will be a significant decoupling effect and, therefore, lower amounts of energy will be transmitted as compared to operating at lower intensities of irradiation. It should also be noted at this stage, that the optimum intensity range also depends on the mode of operation. If there is continuous irradiation, usually the observed decrease in the yields is in the form of a broad range of intensity, whereas, if it is pulsed irradiation of shorter durations, there is a sharp decrease in the yield with increase in the intensity of irradiation. Moreover, if one increases the amount of irradiation time in the pulse, the decrease in yield flattens out with an increase in intensity. So, the pulsed ultrasound equipment can be operated effectively over a narrow range of the intensity. This fact has been provided experimentally with the reaction of decomposition of KI by Gutierrez and Henglein (1990). The observed effect can be explained on the basis of phenomena of acoustic streaming. At shorter pulses, the contribution due to acoustic streaming which renews the liquid continuously around the irradiating surface, such as a horn tip, is much less, that is, even before some effects of irradiation are felt by the new layer of liquid, the irradiation is stopped, thereby decreasing the overall yields from the reaction.

The energy efficiency of the acoustic equipment based on the commercial piezoelectric transducers has never been known to exceed 60% and the single largest transducer has a power input of only 2.5 kW (Sochard et al., 1997). This becomes a serious constraint during the scale-up of sonochemical processes and multiple transducers have to be employed for large-scale sonochemical systems which are capital intensive and pose a number of operational difficulties. Therefore, the better energy efficiencies arising out of larger scales, as available in the conventional processes, is not available in this type of process.

It can also be seen from the figure that the hydrodynamic cavitation equipment is relatively more energy efficient, as compared to their acoustic counterparts. HSH and HPH (typically laboratory-scale equipment with a capacity of 1.5 and 2 L, respectively) have energy efficiency levels of 43% and 54%, respectively. The orifice type of hydrodynamic cavitation reactor having a capacity of 50 L (typically a pilot-plant scale) has an observed energy efficiency of 60%. Therefore, it can be said that the energy efficiency increases with an increase in the capacity of the reactor, which is most important for efficient scaleup.

It can be said from these results that the hydrodynamic equipment gives better performance as compared to the acoustic equipment at industrial scales of operation, at least in the first step of the energy transformation cascade, that is, electrical energy to mechanical energy. Also, it should be noted that the operating scales in the case of hydrodynamic cavitation (capacity of the equipment is of the order of a few liters) are much larger than those in the acoustic cavitation (of the order of a few mL). This also indicates that the scale-up of hydrodynamic equipment will be much easier, as compared to the acoustic equipment where the scale-up ratio required will be of the order of a few hundred to a few thousand (for ultrasonic horn).

### Cavitation yield

The cavitation yield is the actual net production of desired products from the supplied electrical energy, whereas the energy efficiency is indicative of the capacity of the equipment in converting the supplied energy to the actual mechanical energy, some part of which will be used for generation of cavitation events.

The cavitation yield for various acoustic and hydrodynamic equipment is given in Table 1. It can be seen that the desired chemical change is higher for a given amount of electrical energy supplied to the system for the hydrodynamic cavitation reactors. However, it should be noted that the comparison made here is valid only for the model reaction (decomposition of potassium iodide) and the efficiencies of the various equipment may or may not be the same for the variety of cavitation based transformations and also other applications. The present work is an initial step in the correct direction for developing the methodology, and engineers should follow these guidelines when designing and choosing the cavitation reactors and their operating parameters for various applications.

It should also be noted at this stage that the acoustic cavitation generates more intense cavitation (indicated by larger magnitudes of pressure pulses generated in acoustic cavitation) which is discussed in detail later. Among the acoustic cavitation equipment, the bath is more efficient than the acoustic horns. This can be attributed to uniform energy dissipation in the case of the bath due to a larger area available, as compared to localized dissipation of energy in the case of the ultrasonic horn, which may not result in full utilization of the supplied energy which is also indicated from the values of energy efficiency (Figure 6). The detailed explanation of the decoupling effect which governs this observed effect has been discussed earlier.

Therefore, the characterization of different equipment used for generation of cavitation can be taken as a basis for a similar analysis of the reaction/transformation to be carried out on an industrial scale, in order to effectively choose a particular type of equipment. For the decomposition of KI reaction, hydrodynamic cavitation reactors are the best.

### Relationship between macroscopic rates and the collapse pressures

The cavitation yield can be expressed as a function of the quantum of the collapse pressure ( $P_{\text{collapse}}$ , in atm) that will be generated at the collapse of the cavities and can be given as follows

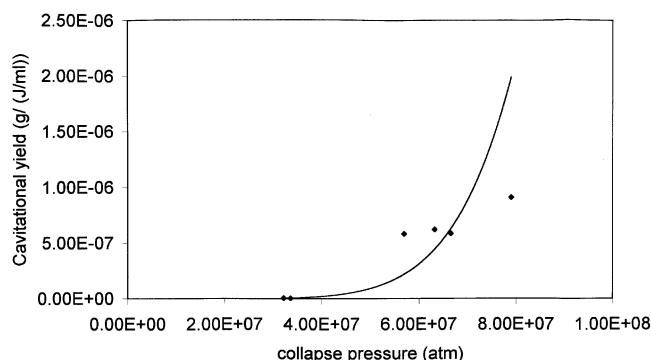
$$\text{Cavitation yield} = K(P_{\text{collapse}})^n \quad (3)$$

where  $K$  and  $n$  depend on the type of the reaction used and also the mode of generation of the cavitation, that is, hydrodynamic or acoustic. The constant  $K$  is also inclusive of the concentration of KI and the time of reaction used in the present work, since it was kept constant throughout the entire

**Table 1. Characterization of Various Equipment Used**

Equipment		Electrical Consump. (W)	Vol. (mL)	Time of Treatment (min)	Actual Energy Dissipated (W)	Energy Efficiency (%)	Iodine Liberated at the End of Reaction (g)	Cavitation Yield g/(J/mL)
Dakshin horn		240	50	10	7.31	3.04	$1.02 \times 10^{-5}$	$3.53 \times 10^{-9}$
Dakshin Bath		120	500	10	46.63	38.86	$8.4 \times 10^{-5}$	$5.83 \times 10^{-7}$
Ace Horn	10%	40	50	10	7.77	15.43	$1.00 \times 10^{-6}$	$1.39 \times 10^{-9}$
	20%	80	50	10	13.42	16.77	$7.55 \times 10^{-6}$	$5.25 \times 10^{-9}$
	30%	120	50	5	19.08	15.9	$5.90 \times 10^{-6}$	$5.48 \times 10^{-9}$
Flow Cell	25 kHz	120	1,500	15	51.66	43.05	$4.47 \times 10^{-5}$	$6.21 \times 10^{-7}$
	40 kHz	120	1,500	15	32.37	26.97	$4.22 \times 10^{-5}$	$5.85 \times 10^{-7}$
	25 + 40 kHz	240	1,500	15	71.14	33	$1.31 \times 10^{-4}$	$9.12 \times 10^{-7}$
High-pressure Homogenizer (5000 psi)		2,090	2,000	30	1,136.97	54.4	$6.637 \times 10^{-5}$	$7.38 \times 10^{-5}$
High-speed Homogenizer		105	1,500	30	45.23	43.07	$1.83 \times 10^{-4}$	$6.645 \times 10^{-7}$
Pilot-plant scale (Orifice plates)	% free area = 2.28%	5,500	50,000	60	3,277	59.58	$9.82 \times 10^{-2}$	$2.48 \times 10^{-4}$
	% free area = 9.14%	5,500	50,000	60	3,344	60.8	$7.50 \times 10^{-2}$	$1.90 \times 10^{-4}$





**Figure 7. Variation of cavitation yield per unit power density with the collapse pressure.**

experimentation (5% *KI* solution and 10 min as the reaction time for the case of acoustic cavitation. In hydrodynamic cavitation, however, the reaction time was taken as 60 min as there was much less iodine liberation during the first 10 min [of the order of  $10^{-5}$ ]).

Variation of the cavitation yield obtained for various acoustic cavitation equipment, with the collapse pressures which are calculated from the correlations given earlier, are shown in Figure 7. The constants  $K$  and  $n$  in Eq. 3 as obtained from Figure 7 are given below

$$K = 1.228 \times 10^{-59} \text{ and } n = 6.737$$

It is worthwhile at this stage to compare the obtained relationship with some of the earlier studies on decomposition of potassium iodide. Naidu et al. (1994) have studied the decomposition of potassium iodide in an ultrasonic bath having a cross-sectional area of  $0.0404 \text{ m}^2$  and a height of 15 cm. The operating parameters used in the experimentation were 25 kHz driving frequency and  $0.6188 \text{ W/cm}^2$  intensity, which are quite similar to that used in the present work. The results obtained indicate a linear variation in the iodine liberation with the initial concentration of potassium iodide and time of the reaction, which confirms our consideration of inclusion of the concentration and the reaction time in the constant  $K$ . Further studies can be aimed at developing a generalized correlation which also includes the concentration of *KI* and the time of reaction as follows

$$\text{Cavitation yield} = [K' \times \text{Conc. of KI} \times \text{time of reaction}] \times (P_{\text{collapse}})^n \quad (4)$$

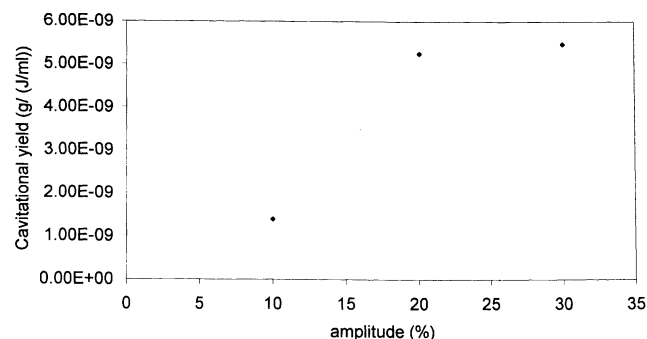
Naidu et al. (1994) obtained the iodine liberation of the order of  $4.15 \times 10^{-4} \text{ g/L}$  for a reaction time of 10 min and initial *KI* concentration of 5%. The correlation developed in the present work predicts an iodine liberation of  $1.79 \times 10^{-4} \text{ g/L}$ , which is two to three fold lower than that predicted by Naidu et al. (1994). The variation in the obtained values can be attributed to the following two reasons:

(1) The system used in the experimentation of Naidu et al. (1994) is saturated with gas. The rates of reaction are much

larger when the system is saturated with gas due to the availability of gas bubble nuclei for easy cavitation, and it decreases with the degassing effect (Senthilkumar et al., 2000). The present work considers a completely degassed system. In our earlier work (Vichare et al., 2000) it has also been shown that the degradation rates are much higher (2 to 3 fold) in the initial stages when the system is completely saturated with gas and, as de-aeration occurs, the system becomes stable giving comparatively much lower rates of reaction and linear variation in time.

(2) The development of correlation in the present work is based on global rates of reaction, that is, it considers the entire available volume for the reaction in the equipment, whereas Naidu et al. (1994) had placed the test tube containing the reacting medium at the particular location where there is maximum intensity of cavitation as detected by hydrophones. The intensities of cavitation are different at different points in the ultrasonic bath which will result in different rates of reaction. The work correlating the degradation rates with the local intensities of cavitation is presently being carried out. The preliminary studies indicate 2 to 10 fold variation in the observed decomposition rates (Gogate et al., 2001) with the location of the test tube used for experimentation, and, therefore, the difference between our studies and those of Naidu et al. (1994) can be easily explained. Moreover, the experimental conditions such as the thickness of the glass tubes used (Gogate et al., 2001) and also the presence of compounds such as  $\text{CCl}_4$  (Shirgaonkar and Pandit, 1997) also affects the rate of decomposition of *KI*. Thus, the rates of degradation of *KI* will be much larger in the case of Naidu et al. (1994) as compared to present experimentation; nevertheless, the order of magnitude appears to be similar and gives confidence in the type of correlation to be used.

Variation of the sonochemical efficiency with the amplitude for the ace horn is shown in Figure 8. As the amplitude of the ace horn increases, more energy is being supplied to the system resulting in an increase in the intensity of irradiation. An increase in the intensity leads to a decreased collapse pressure for a single cavity (Gogate and Pandit, 2000a), but at the same time the number of cavities generated also increases when the amplitude is increased from 10 to 20% resulting in an increase in the total quantum of pressure energy liberated (given as = number of cavities  $\times$  collapse pressure due to a single cavity) during the collapse of cavities. This results in a corresponding increase in the cavitation yield.



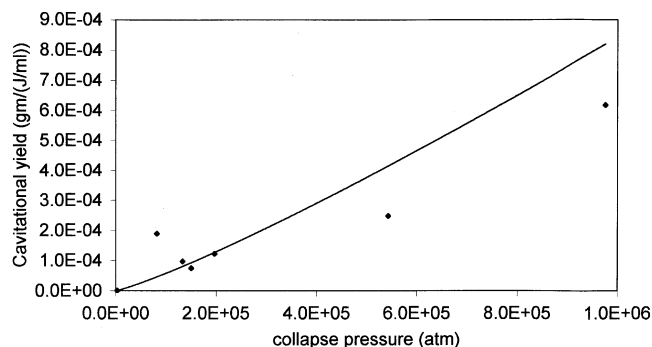
**Figure 8. Variation of cavitation yield with amplitude.**

yield. It is also observed that the increase in the cavitation yield is marginal when the amplitude is increased from 20 to 30%. Ondruschka et al. (2000) have also shown that the rate constant for sonolytic degradation of MTBE increases linearly up to an intensity of 5 W/cm<sup>2</sup> corresponding to a power input of 100 W beyond which there is only a marginal increase in the rate constant. At higher intensities of irradiation, there exists a large number of gas bubbles in the solution which scatters the sound waves to the walls of the vessel or back to the transducer. Therefore, less energy remains in the liquid in this way although the vessel is exposed to higher and higher intensities. Gutierrez and Henglein (1990) have also obtained similar results with decomposition of potassium iodide solution. Also, it may occur that, due to a very high number of cavities per unit volume or area, there is a likely coalescence of the cavities resulting in formation of a larger cavity (the collapse pressure is inversely proportional to the size of the cavity, Gogate and Pandit, 2000a,b). If the amplitude is further increased, then we may observe a decrease in the cavitation yield of the system. A detailed explanation of this fact and the experimental confirmations for presence of optimum intensity has been offered in some of the earlier works (Gutierrez and Henglein, 1990; Gogate and Pandit, 2000a; Ondruschka et al., 2000).

A similar variation of the cavitation yield with the collapse pressure for hydrodynamic cavitation equipment is shown in Figure 9. It should be noted that the theoretical modeling is much more complicated for a high-pressure homogenizer system due to the complex flow fields and turbulent pressure variations; therefore, the prediction of collapse pressures existing in the system are not possible at this stage. The correlation developed for the present work (hydrodynamic cavitation equipment orifice plate setup and HSH) is given as

$$\text{Cavitation yield} = 8.834 \times 10^{-11} (P_{\text{collapse}})^{1.1633} \quad (5)$$

The various points considered for the hydrodynamic cavitation reactors are due to the difference in the geometry of the reactor used for the orifice setup. Different plates differing in the arrangement of holes in terms of the number and diameter (Table 2) have been used in the setup, and experiments have been performed for the estimation of cavitation yield. Figure 10 shows the effect of geometry on the rate of liberation of iodine. It can be seen from the table and figure that the rate of iodine liberation is inversely proportional to the percentage free area offered for the flow, and also to the diameter of the hole used on the orifice plate. Therefore, the arrangement of the holes on the orifice plate is also a crucial factor and the manipulation of the same can be done depending on the required intensity of the cavitation. In the earlier work (Gogate and Pandit, 2000b) it has also been shown that using the theoretical simulations of the bubble dynamics equation, the magnitude of pressure pulse generated due to the collapse of a single cavity increases with a decrease in the percentage free area offered for the flow. Thus, the observed experimental results can be well explained on the basis of theoretical pressure pulse magnitude predictions. The effect of variation in the operating conditions and geometric parameters on the liberation of iodine



**Figure 9. Variation of cavitation yield with collapse pressure for hydrodynamic cavitation equipment.**

has been also reported in detail in earlier work (Vichare et al., 2000), whereas similar studies have been done for the reaction of the degradation of rhodamine B dye (Sivakumar and Pandit, 2001b) and for cell disruption (Balasundaram and Pandit, 2001) in the department.

Hydrodynamic equipment, although comparatively far more energy efficient in converting electrical to mechanical energy, generates less intense cavitation (observed from the values of collapse pressure generated in the two cases). It should be noted that the collapse pressure generated in the case of acoustic cavitation is order of magnitude higher than the hydrodynamic cavitation. Therefore, one can conclude that the equipment which is more energy efficient in the first step of the energy transformation cascade, does not necessarily generate intense cavitation for driving the chemical reactions. The only exception to this is the pilot-scale orifice setup (the collapse pressures generated are 100 times more than the HSH or HPH). This configuration is more efficient in terms of the dissipation of the energy, as well as in generating cavitation activity. In fact, the study with the decomposition of potassium iodide shows that under optimum operating conditions, the setup is far more cavitationally active than the acoustic type (almost three orders of magnitude higher yields are obtained). It should be noted at this stage that the results obtained in the present work in terms of the superiority of hydrodynamic cavitation reactors over acoustic cavitation is valid only for the model reaction used in the present work and may or may not be applicable to other reactions where there are stringent cavitation conditions. Thus, there is a need for laboratory-scale studies of the reaction under question along the lines of the methodology used in the present work for the comparison of different equipment that may be

**Table 2. Geometrical Details of the Different Plates Used in the Experimentation**

Plate	No. of Holes	Dia. of Hole (mm)	Flow area (mm <sup>2</sup> )
Plate 1	33	1	25.92
Plate 2	8	2	25.13
Plate 3	33	2	103.67
Plate 4	16	3	113.1
Plate 5	20	3	141.4
Plate 6	8	5	157.1

available to the potential user, and then an optimum selection of equipment may be possible.

It must also be stressed at this juncture that the cavitation activity in the pilot-scale plant can be varied using the different types of plates, with varying number and diameter of holes as shown in Figure 11. Collapse pressures similar to acoustic cavitation can also be generated where such severe conditions are required. In a recent work (Sivakumar and Pandit, 2001b), Rhodamine B complex destruction, which needs much more intense conditions as compared to the current model reaction, was studied in different equipment, both using acoustic and hydrodynamic means of generation. It has been found that the cavitation yield for the hydrodynamic cavitation reactor is the highest, as compared to the ultrasonic equipment. To give a quantitative idea, the cavitation yield for plate 1 is twice as high as compared to an ultrasonic bath (250 mL operating volume; for details refer to earlier work of Sivakumar and Pandit, 2001a), which is the best among the various ultrasonic equipment considered in the work. Moreover, Kalumuck and Chahine (1998) have shown that the cavitating jets give an order of magnitude higher cavitation yields for destruction of p-Nitrophenol, as compared to the ultrasonic horn used in their experimentation. In their work, the degree of intense cavitation was increased by arranging multiple orifice plates one after another in a cavitational active zone. These illustrations clearly indicate that depending upon the required intensity of cavitation suitable for a certain application, the geometry of the cavitation reactor can be altered so as to get similar conditions to acoustic cavitation, but in a more energy efficient manner.

Also, based on the data shown in Table 1, the efficacy of HSH and HPH cannot be totally underestimated. Studies

have shown that the critical pressure necessary to generate cavitation in HPH is 5,000 psi (34 MPa) (Shirgaonkar, 1997), while the critical speed to generate cavitation in the case of HSH is 9,000 rpm (Senthilkumar, 1998). Depending on the physical properties of the liquid, one can easily for higher pressures and speeds, respectively, to obtain more intense cavitation and improve the performance of this hydrodynamic equipment for cavitation yield.

Further work is also required to check the suitability of the hydrodynamic cavitation equipment for carrying out complex reactions. The present work is just an indication of the higher energy efficiency of the hydrodynamic cavitation for a specific model reaction considered.

## Conclusions

An exhaustive study to compare the efficacy of various acoustic and hydrodynamic cavitation equipments for driving a model chemical reaction have been carried out by studying *KI* oxidation reaction. From the results obtained in the study, the following conclusions can be made:

(1) Hydrodynamic cavitation reactors are more energy efficient as compared to the acoustic counterparts, and also give higher cavitation yields, at least in the case of the model reaction considered in this work. The energy efficiency increases with an increase in the scale of operation, thus making the scale-up of hydrodynamic cavitation equipment easier and more efficient.

(2) Even though equipment based on acoustic cavitation is able to generate very rigorous/intense cavitation as indicated by the magnitude of collapse pressures generated during the cavitation phenomena, it is less energy efficient and offers

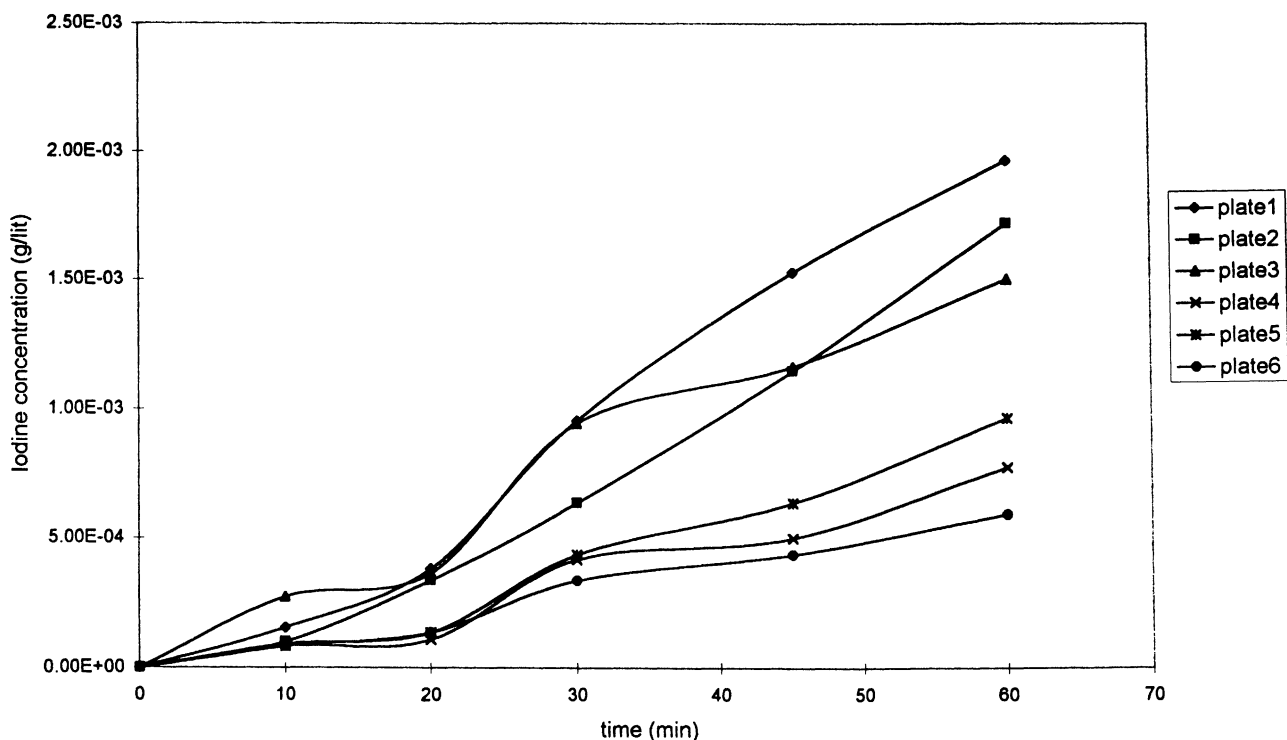
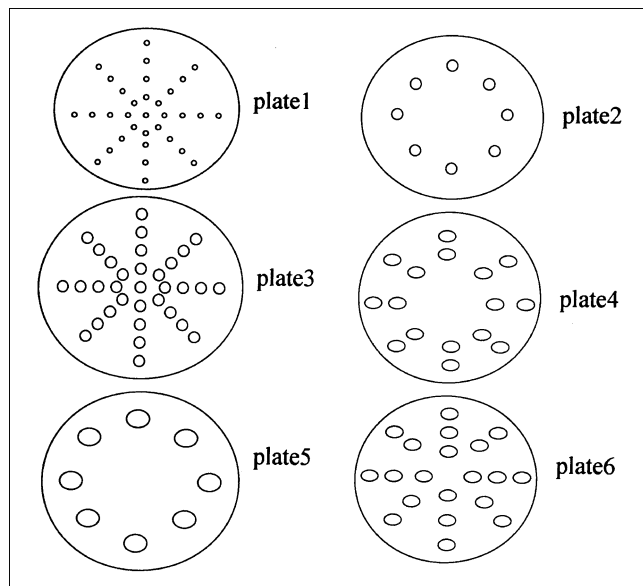


Figure 10. Variation of iodine liberation with time for different plates.



**Figure 11. Arrangement of different free areas on the plates.**

lower cavitation yields. Therefore, in order to improve the energy efficiency of acoustic cavitation reactors, future research should concentrate on the development of efficient transducers for sonication and better geometric reactor configurations.

(3) Reactors based on hydrodynamic cavitation show a considerable optimization possibility due to the presence of multiple pressure oscillation frequencies, present as turbulent and chaotic flow and resulting in multiple size resonating cavities, unlike acoustic equipment which operates at a fixed frequency. Moreover, different geometries can be used for achieving the desired degree of cavitation intensity depending on the application of the reactor.

This study clearly shows that hydrodynamic cavitation (pilot-plant scale) has the twin advantage of greater energy efficiency and ability to generate intense cavitation under optimum operating conditions for the present reaction. Moreover, the scale-up of these reactors is relatively easy.

(4) A correlation has been developed for the estimation of the cavitation yield in terms of the cavity collapse pressure for the acoustic and hydrodynamic equipment. Such a correlation, along with the correlations developed in earlier work for the prediction of collapse pressure, will help the designer to choose and optimize the operating and geometrical conditions in the reactor, in order to achieve the desired chemical change. It should be again noted that the predictions of the correlation are strongly dependent on the operating conditions used in the system such as the presence of gas like nitrogen, oxygen or certain additives such as  $\text{CCl}_4$  and also, that it varies from reaction to reaction. Such relationships between the observed macroscopic reaction rates and the collapse pressures are the first reported in the literature, and further studies should be concentrated in developing the same for a variety of applications of cavitation reactors.

It would also be worthwhile to study instantaneous reactions with acoustic cavitation on the hydrodynamic equip-

ment, and the reactions which only use the mechanical effects of ultrasound, that is, the reactions which are not truly sonochemical, could be studied in the hydrodynamic cavitation reactors.

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## Notation

- $A$  = percentage free area offered for the flow
- $C_p$  = heat capacity of the solvent,  $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
- $d_o$  = diameter of the orifice, m
- $dT$  = temperature difference, K
- $dt$  = time, s
- $f$  = frequency of irradiation, kHz
- $I$  = intensity of irradiation,  $\text{W}/\text{cm}^2$
- $K, n$  = constants used in Eq. 3
- $K'$  = constant used in Eq. 4
- $m$  = mass of solvent, kg
- $P_{\text{collapse}}$  = pressure pulse generated during the collapse of the cavities, atm
- $P_i$  = inlet pressure into the system,  $\text{N}/\text{m}^2$
- $R_o$  = initial size of the nuclei, m

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