Energy Analysis in Acoustic Cavitation

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5 authors, including:

Parag R Gogate
Institute of Chemical Technology, Mumbai

Aniruddha Pandit
Institute of Chemical Technology, Mumbai

Energy Analysis in Acoustic Cavitation

Nilesh P. Vichare, P. Senthilkumar, Vijay S. Moholkar, Parag R. Gogate, and Aniruddha B. Pandit*

Chemical Engineering Section, University Department of Chemical Technology, Matunga, Mumbai-400 019 India

In sonochemical processes, the physical and chemical effects are attributed to the phenomenon of cavitation, which is the formation, growth, and collapse of the cavities termed as activity of cavities. Energy analysis of a single cavity has been considered in order to explain, qualitatively, the effects of acoustic parameters such as intensity and frequency of ultrasonic equipment on sonochemical reactions. The experimental observations of the sonochemical reactions available in the literature are found to be consistent with the simulation results. Energy analysis of the cavity gives the possible reasons for the various sonochemical effects observed, and also the optimum equipment operating parameters can be predicted.

1. Introduction

In sonochemical transformations, the energy of the ultrasound is not utilized directly by the liquid reactants and the effect of ultrasound takes place through a process called cavitation, which is the formation, growth, and violent collapse of cavities. The efficacy of the sonochemical process depends on the cavitation process occurring in the liquid, which in turn depends on the combined effect of individual cavities, but millions of them. So the objective is to make the cavitation processes efficient, so that maximum utilization of the energy supplied to the system in the form of ultrasound (mechanical energy) can be achieved. Any effort to estimate the energy content of a collapsing cavity under various operating conditions could yield important information qualitatively on the most favorable operating conditions. A theoretical study based on the model of single cavity dynamics using Rayleigh−Plesset equation has been carried out to estimate the energy of the collapsing cavity. On the basis of this, the explanation of the results of sonochemical effects reported in the literature for various reactions has been made.

Flynn's[1] criterion suggests the adiabatic collapse phase of the cavity on the basis of the assumption of partial pressure of gas in the cavity equals the vapor pressure of the cavitating media at the operating temperature. Thermodynamic principles have been used to calculate work done during the lifetime of the cavity. The energy dissipated at the end of the cavity collapse is calculated from the "net" work done by the cavity during its lifetime. The effect of intensity and frequency of ultrasound on the quantum of energy dissipation and rate of dissipation has been studied by performing numerical simulations of the cavity dynamics.

2. Energy Analysis

2.1. Energy Analysis of the Cavity Considering Flynn's Assumption. Considering Flynn's[1] assumption, which states that the collapse phase becomes adiabatic at the point at which the partial pressure of the gas in the cavity becomes equal to the vapor pressure of the cavitating media under operating temperature, the critical radius (R_{crit}) at which this occurs has been calculated. Beyond R_{crit}, the resistance to heat and mass transfer lies inside the cavity, and heat and mass transfer between the cavity and the surroundings is neglected; i.e., the collapse has been assumed to be adiabatic. For isothermal expansion and isothermal collapse, the pressure inside the oscillating cavity is given by

$$p_i = p_v + p_{go}\left(\frac{R_o}{R}\right)^3 \tag{1}$$

where p_{go} is the initial gas pressure.

The isothermal process continues up to the critical radius (R_{crit}) beyond which the collapse is adiabatic. The R_{crit} is found from the following relation:

$$R_{crit} = R_o\left(\frac{p_{go}}{2p_v}\right)^{1/3} \tag{2}$$

The pressure at the beginning of the adiabatic collapse phase is $2p_v$. So the pressure inside the cavity at any instant during adiabatic collapse phase is

$$p_i = 2p_v\left(\frac{R_{crit}}{R}\right)^{3\gamma} \tag{3}$$

The Rayleigh−Plesset equation governing the variation in the bubble radius with time, under the given set of operating conditions, has been solved using the fourth-order Runge−Kutta method. The details of the simulation conditions and the modeling part with typical profiles of the radius history has been already described in detail (Gogate and Pandit[2]) for the case of acoustic cavitation.

The work done during the lifetime of a cavity for noncyclic process is given by

$$W = \int_{V_1}^{V_2} p_i \, dV_i \tag{4}$$

The work done during the isothermal expansion (growth of the cavity) and part of the collapse up to the

* To whom correspondence should be addressed. E-mail: abp@udct.ernet.in.

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critical radius R_{crit} is given by

$$W_{iso} = p_i V_i \ln\left(\frac{p_2}{p_1}\right) \quad (5)$$

where V_i is the volume of the cavity and p_1 and p_2 are the initial and final pressures during each simulation step, respectively. At the critical point, the pressure inside the cavity (p_i) is equal to $2p_v$ and the work done on the cavity during the adiabatic compression phase is given by

$$W_{ad} = \frac{p_i V_i}{(\gamma - 1)}\left[1 - \left(\frac{p_2}{p_1}\right)^{(\gamma-1)/(\gamma)}\right] \quad (6)$$

As the work done is a path function, the computation of the same is done by breaking up the expansion and collapse phase of the cavity into a number of time segments. The sum of the values of all the segments will give the total work exchange during expansion and collapse phases under the conditions specified earlier. As the cavity is the system under consideration, the work done during the expansion is considered to be negative and the work done by the surroundings on the cavity during compression is considered to be positive. The net work done is thus given by

$$W_{net} = W_{iso} - W_{ad} \quad (7)$$

The time scale in which the work is done on the cavity is of primary importance in order to compare and compute the efficiency of the cavities oscillating with different frequencies. So the rate of work done or power is given by

$$\text{rate of work done on the cavity, } E_c = \frac{W_{net}}{t_c} \quad (8)$$

where t_c is the lifetime of the cavity.

The content of the cavity i.e., a gas−vapor mixture has been considered to behave as an ideal gas.

Pressure inside the cavity at any instant during isothermal growth and collapse phase is thus estimated by following equation

$$p_i = (p_{go} + p_v)\left(\frac{R_o}{R}\right)^3 \quad (9)$$

considering initial equilibrium, $p_{go} + p_v = p_o + 2\sigma/R_o$.

Using Flynn's assumption, pressure inside the cavity during adiabatic collapse phase can be given by eq 3.

2.2. Energy Analysis of the Cavity Considering Isothermal Growth Followed by Complete or Partial Adiabatic Collapse. In this case expansion of the bubble is considered as isothermal and its total compression is considered as adiabatic without the consideration of Flynn's assumption.

For a vapor-saturated gas cavity, pressure inside the cavity during the isothermal expansion phase is

$$p_i = p_{go}(R_o/R)^3 + p_v \quad (10)$$

The adiabatic phase starts when $R = R_{max}$, corresponding $p_i = p_{min}$.

Pressure inside the cavity at any instant during adiabatic collapse phase thus can be obtained by modifying eq 3 in terms of the following quantities:

$$p_i = p_{min}(R_{max}/R)^{3\gamma} \quad (11)$$

Work done during isothermal growth and adiabatic collapse can be computed as discussed previously.

If the complete gaseous cavity, i.e., gas−vapor mixture is considered as an ideal gas, then the pressure inside the cavity at any instance during isothermal growth phase is

$$p_i = \left(p_o + \frac{2\sigma}{R_o}\right)\left(\frac{R_o}{R}\right)^3 \quad (12)$$

where R is the radius of the cavity at that instance and the maximum value it can reach is R_{max}.

When $R = R_{max}$, the adiabatic collapse phase starts. Pressure inside the collapsing cavity during adiabatic collapse phase at any instance can again be computed using eq 11, by the substitution of the corresponding R value at that instance.

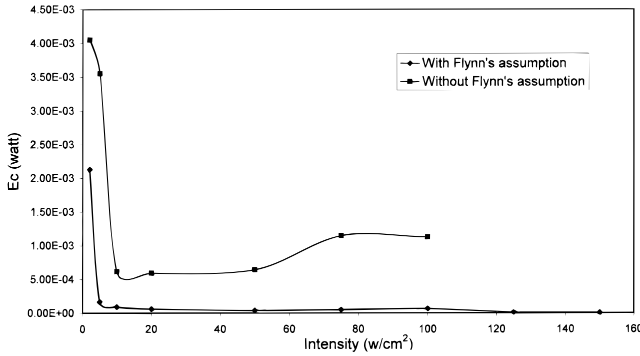

Figure 1. Effect of intensity on E_c for vapor-saturated cavity for two conditions, i.e., with and without Flynn's assumption at R_o = 10 μm and f = 100 khz.

3. Results and Discussion

During cavitation, the energy that is dissipated into the system by the passage of ultrasound is taken up by the cavities and concentrated on very small scales, but at millions of them in the liquid. The energy of the cavity continuously increases during its isothermal growth (stored up), and it is released back into the liquid as a pressure pulse and resulting shock waves with very high energy density (watts/m^3).

3.1. Effect of Ultrasound Intensity (*I*). Figure 1 shows the intensity effect on the energy dissipation rate for a vapor-saturated gas cavity for the two conditions considered, i.e., with and without Flynn's assumption. Considering Flynn's assumption for a vapor-saturated cavity, with an increase in the ultrasound intensity (I), E_c decreases. With an increase in intensity (2 to 200 W/cm^2), work stored in the cavity, W_{net} decreases (by 10−20 times), and the lifetime of the cavity increases (1.5−2 times). Hence, energy released by the cavity decreases by 100−200 times with an increase in the intensity from 2 to 200 W/cm^2. It should be noted that the extent of decrease in the E_c is also a function of initial radius of the cavity used and at higher value of initial radius R_o (of the order of few mm), decrease in E_c with an increase in the intensity (2−200 W/cm^2) is not substantial (10−20 times), with much lower absolute values of E_c.

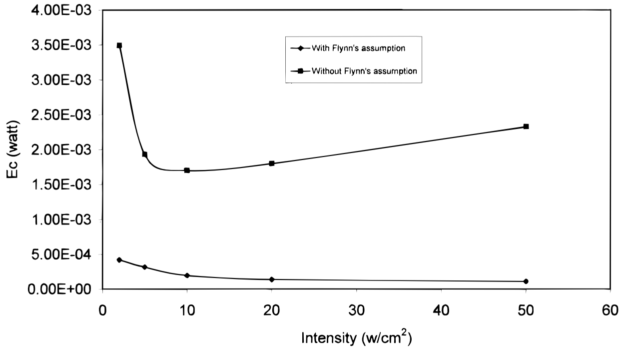

Figure 2. Effect of intensity on E_c for complete gaseous cavity for two conditions, i.e., with and without Flynn's assumption at $R_o = 10$ μm and $f = 250$ khz.

If isothermal growth followed by complete adiabatic collapse of the vapor-saturated cavity is considered then for a constant frequency and initial cavity radius (R_o), with an increase in the ultrasound intensity W_{net} decreases and energy released by the cavity, E_c decreases initially but increases further at higher intensities although marginally (10−50%, extent of increase is also a function of the initial radius). The energy released in this case is found to be partially higher (2−10 times more) as compared to the earlier case, i.e., considering Flynn's assumption.

Figure 2 shows the variation in energy dissipation rate with intensity of ultrasound for a **completely gaseous cavity**. At higher intensities, with constant R_o and frequency, values of E_c are partially lower (2−10 times) than those for the vapor-saturated cavity; but the trend of variation in the E_c with ultrasound intensity is the same as in the case of vapor-saturated cavity for both the cases, i.e., with and without Flynn's assumption. The magnitudes of energy dissipation rate in the case of complete adiabatic collapse of a cavity are higher (10−100 times) than those in the case of Flynn's assumption consideration where the collapse is isothermal until the condition of critical radius and then adiabatic.

It should be noted that the value of the critical radius is also a function of the operating conditions, and hence the extent of variation between the two pressures or the energy dissipation obtained by the two assumptions, i.e., assuming the collapse as completely adiabatic and partly adiabatic (Flynn's assumption), will be dependent on the operating conditions. The simulations are terminated in the present case when the bubble wall velocity reaches the value of 1500 m/s but in reality the velocities will be much larger when the medium is compressible. The contribution of the adiabatic phase of collapse hence will be much stronger in the case of compressible mediums, and hence, we may end up in underestimating the collapse pressures and energy intensities when partly isothermal collapse is considered and simulations are terminated at a bubble wall velocity of 1500 m/s. Gogate and Pandit[2] have studied the effect of compressibility of medium on the values of the collapse pressures generated and have shown that the collapse pressures are much larger when the compressibility of the medium has been considered i.e., the velocities are in excess of 1500 m/s.

Entezari and Kruus[3] have studied the effect of intensity on sonochemical decomposition of potassium iodide to liberate iodine using different sizes of the horn tip, which changes the intensity of irradiation. The rate of the decomposition was found to be higher at lower intensities, i.e., with the large tip. A large horn tip produces more bubbles as it can contact larger surface area of the liquid and therefore irradiates a larger volume of liquid. **So the logical conclusion is that the cavities are less effective at higher intensities.** Experimental results using pulsed ultrasound [Guiterez and Henglein[4]] shows that iodine liberation drops to zero with an increase in the intensity in a narrow range of intensities. The rate of iodine liberation at constant power input for equipments of different area of irradiation indicates that decrease in intensity (decreasing power input or by increasing ultrasound irradiating area) resulted in manifold increase in the liberation of iodine [Entezari and Kruus[3,5]]. Studies on emulsification of an oil−water system using an ultrasonic horn and bath showed better emulsion characteristics for the bath, though the power input to the horn and the bath is nearly the same [Mujumdar et al.[6]]. Sehgal and Wang[7] have also shown that the sonochemical yield continuously decreases with an increase in the intensity of irradiation.

Increasing intensity increases the amplitude of the vibration of the horn but induces a phase lag between the motion of the liquid and that of the horn, which is known as the coupling effect. So the oscillating surface does not remain in contact with the liquid all the time during its motion, and therefore, ultrasound transmission efficiency decreases. Also increase in the intensity causes the generation of a large number of bubbles, which may coalesce and escape before the collapse.

A point to be noted here is that with the lowering of the intensity, though the energy associated with the individual cavity increases, the number of cavities could also be substantially less. Thus, the net effect of this could result in an optimum intensity giving the maximum sonochemical effect. Nevertheless, it could be concluded from the above theoretical simulations that as far as the individual cavity efficiency (percent of the supplied energy released at the collapse) is concerned, lower intensities (just above the threshold intensity values) are most favorable.

3.2. Effect of the Ultrasound Frequency. It can be seen from Figure 3 that for a vapor-saturated cavity, with an increase in the frequency (f), E_c increases for two conditions, i.e., with and without consideration of Flynn's assumption. The range of frequency selected is 50−1000 kHz. For simulations with Flynn's assumption, the energy released by the cavity, E_c, at higher frequencies is almost 100−200 times more than that at lower frequencies. With an increase in the frequency, work done by the cavity during its lifetime increases (10−100 times), and the lifetime of the cavity decreases (10 times). So values of E_c are substantially higher at higher frequencies than at lower frequencies. If isothermal growth followed by adiabatic collapse of the vapor-saturated cavity is considered, then with an increase in the frequency from 50 to 1000 kHz, the value of E_c increases by 10−100 times.

For a complete gaseous cavity, trends similar to that for the case of the vapor-saturated cavity of E_c with the variation in frequency are obtained as shown in Figure 4. At higher frequencies the values of E_c are higher (10−15 times) for the vapor-saturated cavity than for the complete gaseous cavity. Similarly, for the completely gaseous cavity, considering isothermal growth followed by adiabatic collapse, energy released by the bubble at

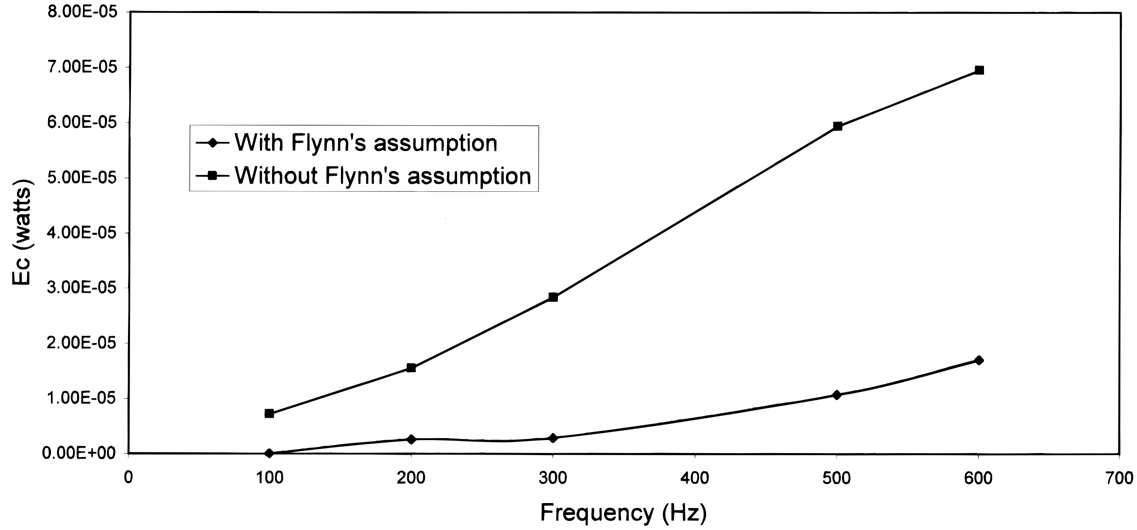

Figure 3. Effect of frequency on E_c for vapor-saturated cavity for two conditions, i.e., with and without Flynn's assumption at $R_o = 1$ μm and I = 10 W/cm^2.

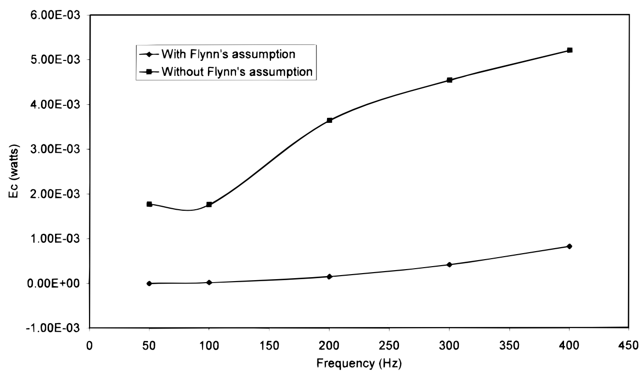

Figure 4. Effect of frequency on E_c for complete gaseous cavity for two conditions, i.e., with and without Flynn's assumption at $R_o = 5$ μm and $I = 5$ W/cm^2.

the end of collapse, E_c, increases with an increase in the frequency at constant intensity and initial cavity size. The magnitudes of E_c for gaseous and vaporous cavities are comparable at lower frequencies, but at higher frequencies (f > 500 kHz) they are almost 50 times more for the vapor-saturated cavity than for the complete gaseous cavity.

Observations reported in the literature appear to indicate that, at a fixed intensity, increasing the operating frequency increases the yield. At lower frequencies, for a given intensity, the cavity grows to a larger size and the collapse is less violent (it does not enter the adiabatic phase). So the cavities at lower frequencies are inefficient in utilizing the energy of the ultrasound.

Sonochemical degradation of aqueous carbon tetrachloride at various frequencies indicated higher efficiencies at 500 kHz than at 20 kHz [Francony and Petrier[8]]. The sonochemical oxidation of potassium iodide at 20 kHz was studied by Entezari and Krus,s[3,5] and the data were extrapolated at 900 kHz using the same conditions. Extrapolation of the results shows that the effect of frequency on the oxidation rate is almost 2 times greater at 900 kHz than at 20 kHz. Under these conditions, it is difficult to isolate the effect on sonochemical reactions due to ultrasound frequency alone. It should also be noted that the threshold intensity for cavitation, which is a strong property of the liquid could increase at higher frequencies [Mason[9]].

The simulations indicate that the energy efficiency of the cavity is higher for high frequencies and the experimental observations available in the literature also support the results of these simulations. **Hence, higher frequencies are favorable from the point of view of energy efficiency of the sonochemical processes.**

3.3. Effect of Collapse Radius. In all the above simulations, the computation is stopped when the cavity wall velocity reaches the speed of sound (1500 m/s) as the liquid phase compressibility effects, which are likely to be significant, have been neglected. However, the size of the cavity corresponding to the cavity wall velocity equal to the speed of sound is not the same in each simulation. The energy associated with the cavity at different sizes during the collapse is computed for various frequencies and intensities. The results of the analysis are shown in Figure 5. For a given intensity and frequency, the energy associated with the cavity increases as collapse radius decreases due to adiabatic nature of collapse when ratio of collapse radius to initial radius of cavity is less than 1. The values of E_c are almost same irrespective of the value of collapse radius when the ratio of collapse radius to initial radius of the cavity is more than one. When the ratio is less than one, for the same value of collapse radius, E_c value is lower (2−3 times) for high intensity. The simulations indicate a substantial decrease (100 times) in the E_c value with an increase in the intensity.

At constant collapse radius, when the ratio of collapse radius to initial radius of the cavity is less than one, E_c value increases with an increase in the frequency (2−3 times). When the ratio is greater than 1, the values of E_c are very close at various frequencies. The simulations indicate substantial increase (100−200 times) in energy dissipation rates with an increase in the frequency (50−1000 kHz). The variation in the collapse radius for different intensities and frequencies is negligible in some cases and does not affect the energy dissipation rate due to the change in intensity and frequency, or in other words, the variations in the energy dissipation rates are due to the change in the intensity and frequency only and are not due to the change in the value of the collapse radius, considered in the termination of the simulation.

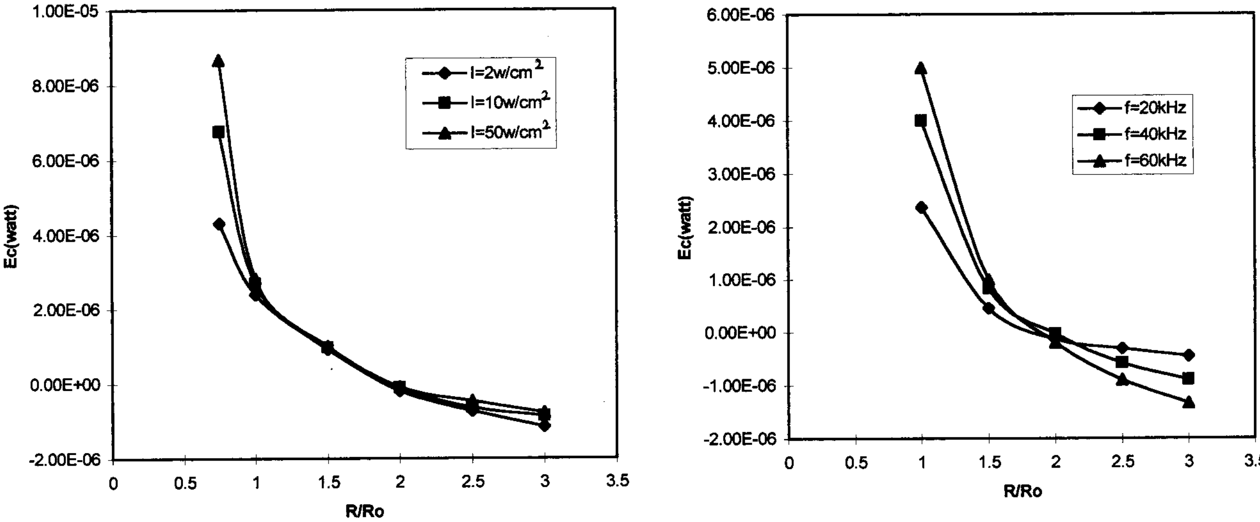

Figure 5. Effect of radius of collapse on E_c.

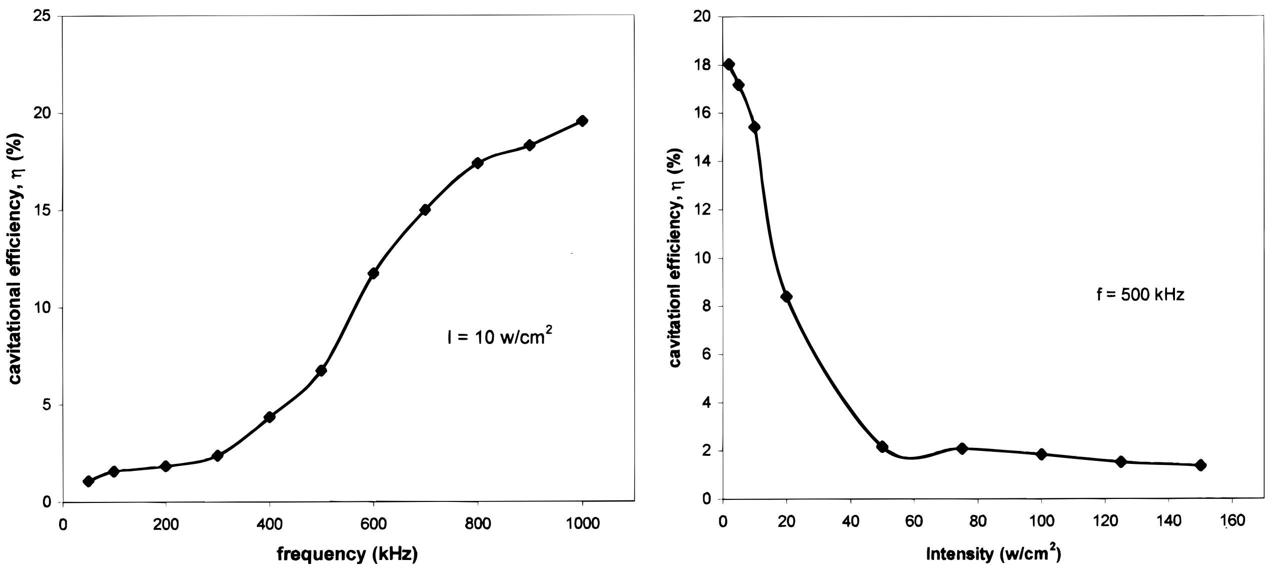

Figure 6. Cavitational efficiency vs intensity and frequency.

5. Cavitational Efficiency (η)

The fraction of the total energy supplied to the system utilized for cavitation process can be determined from the concept of cavitational efficiency.

Acoustic horn is the system under consideration. The details of the acoustic system considered are as follows:

acoustic horn (Dakshin horn):

frequency of horn, $f = 22.7$ kHz

cross-sectional area of the horn, $A_h = 4.9\ \text{cm}^2$

The amplitude of the horn oscillations, a, is estimated from the energy of a standing wave as follows (Schram[10]):

$$a = 2\pi f \sqrt{\frac{2I}{\rho_l C}} \tag{13}$$

The velocity of the horn is taken as the product of frequency and amplitude of the horn.

velocity of the horn, $v_h = 4.1$ cm/s

The power dissipated into the liquid (estimated calorimetrically) is $E_s = 24.525$ W.

Cavitational efficiency (η) can be determined as follows:

$$\eta\ (\%) = \frac{E_c}{E_s} N_c V_L \times 100 \tag{14}$$

The number of cavities generated per unit volume of liquid (N_c) was determined experimentally by Naidu et al.[11] as $2.6445 \times 10^{13}\ \text{m}^{-3}$.

The Volume of liquid displaced by the cavities, V_L, can be determined as follows:

$$V_L = A_h \times \text{distance traveled by the cavity during its lifetime} \tag{15}$$

$$\text{distance traveled by the cavity during its lifetime} = v_h t_c \tag{16}$$

The variation in cavitational efficiency with frequency and intensity is shown in Figure 6. The figure indicates higher cavitational efficiencies at lower intensities and higher frequencies, again consistent with the experimental observations of the effect of the frequency and intensity of ultrasound.

It can be seen from Figure 6 that these efficiencies range from about 1 to 20%, indicating that the actual energy utilized in the cavitational activity is about 1−20% of the mechanical energy supplied (estimated calorimetrically). Thus, the estimation of the efficiency of the cavitation reactor cannot be directly evaluated on the basis of calorimetric energy dissipation rates.

The literature[9,12] shows that even though the calorimetrically determined energy dissipation values are high for a specific equipment, the cavitational activity or cavitational yield does not show a direct relationship with this value. Thus, correlating cavitational yield with the transducer efficiencies (conversion of electrical to mechanical energies), may not be a correct method of correlating cavitational yield data, and an additional conversion factor called the efficiency of the cavitational activity (ratio of energy released by the collapsing cavity to mechanical energy dissipation) needs to be considered for a proper energy balance, correlating the cavitational yield. It also must be borne in mind that the value of the number of cavities per unit volume considered is likely to vary with the dissolved gas content of the cavitating medium, vapor pressure and the surface tension of the medium at any specific conditions. Thus, the values of the efficiencies should be taken only as indicative values, and more efforts are required to assign a correct value for the number of cavities.

6. Conclusion

Energy analysis of the cavity gives the plausible reasons for the various sonochemical effects observed, and also the optimum operating parameters for any sonochemical equipment can be predicted. The trends of energy dissipation rate with the variation in frequency in the case of isothermal expansion followed by adiabatic collapse of a cavity (without considering Flynn's assumption) are almost similar to that for the case of simulations using Flynn's assumption.

(1) Energy dissipation rate increases with an increase in the frequency and decreases with an increase in the intensity for an individual cavity, containing a vapor-saturated gas mixture, as well as for a gaseous cavity.

(2) Energy dissipation rate decreases with an increase in the intensity for Flynn's assumption consideration and for complete adiabatic collapse conditions; it decreases initially but increases at higher intensities although marginally.

(3) Rate of iodine liberation is found to increase with an increase in the frequency, and simulation results can explain the increase in the sonochemical yield due to the increase in the frequency of the ultrasound.

(4) Simulations considering isothermal growth followed by adiabatic collapse can explain intensity effects. Intensity of the ultrasound can be varied by changing the cross-sectional area of the ultrasonic horn, and sonochemical oxidation rates of iodide were reported to increase with a decrease in the intensity. Simulations based on Flynn's assumption can explain this effect.

(5) A small fraction of the energy supplied is actually utilized for cavitation (maximum up to 20%). Cavitational efficiencies are high at lower intensities and higher frequencies. The actual cavitational efficiencies (useful cavitation) are considerably lower than the mechanical efficiencies of the transducers.

Thus, for the overall success of sonochemical processes, lower intensity and higher frequency of the ultrasound is always favorable, though other system specific parameters such as threshold intensity, liquid properties, i.e., vapor pressure, and effect of nuclei (amount of dissolved gases) cannot be neglected.

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Nomenclature

a = amplitude of horn oscillations
A_h = area of the acoustic horn (m^2)
C = velocity of sound in the liquid (m/s)
E_c = energy dissipation rate at the end of bubble collapse (W)
E_s = power input to the acoustic system (W)
f = frequency of ultrasound (Hz)
I = intensity of ultrasound (W/m^2)
N_c = number of cavities per unit volume of liquid (m^{-3})
p_1, p_2 = initial and final pressures during each simulation step respectively (N/m^2)
p_{go} = initial gas pressure (N/m^2)
p_i = pressure inside the bubble (N/m^2)
p_v = vapor pressure (N/m^2)
R = radius of cavity/bubble (m)
R_{crit} = critical radius of the bubble/cavity (m)
R_o = initial radius of the bubble/cavity (m)
t_c = lifetime of the cavity (s)
V_1, V_2 = initial and final volumes of the bubble during each simulation step (m^3)
V_i = volume of the bubble/cavity (m^3)
v_h = velocity of acoustic horn (m/s)
V_L = volume of liquid displaced by the cavities (m^3)
W = work done during a process (J)
W_{ad} = work done during adiabatic phase (J)
W_{iso} = work done during isothermal phase (J)
W_{net} = net work done by the bubble during its lifetime (J)

Greeks

γ = specific heat ratio
σ = surface tension of liquid (N/m)
ρ_l = density of liquid (kg/m^3)
η = cavitational efficiency

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