Chapter 1

INTRODUCTION

A multitude of important physical and chemical processes promoted by ultrasonic cavitation can be implemented on industrial scale by utilizing high-capacity flow-through ultrasonic reactor systems. These systems can permit processing large volumes of liquids and commonly comprise an ultrasonic-frequency electrical signal generator, an electromechanical transducer which converts the electrical signals into ultrasonic vibrations, an ultrasonic horn, which amplifies and transmits the vibrations into the liquids, and a flow-through reactor chamber (flow cell) which contains the flowing liquids. A general schematic of such a system is presented in Figure 1 [1, 2]. Several conditions must be fulfilled in order to ensure effective and continuous operation of an ultrasonic reactor system:

a. technologically necessary intensity of ultrasonic cavitation must be achieved in the liquid;
b. size and homogeneity of the cavitation region formed in the liquid must be maximized (well developed cavitation region);
c. reactor chamber must direct all of the liquid through the cavitation region (no liquid bypass);
d. electromechanical transducer must be electrically save, capable of continuous operation at full power for extended periods of time, and able to provide high radiation power levels;
e. ultrasonic horn must be capable of amplifying vibration amplitudes (high gain) while maintaining maximum possible size of the resulting cavitation region (large output diameter);

f. mechanical stresses present in the electromechanical transducer and the ultrasonic horn must not approach the limiting fatigue strength values for the corresponding materials;
g. entire system as well as each of its components must not be in danger of becoming overheated during continuous operation at full power.

High-quality engineering calculations of ultrasonic reactor system components can only be properly performed if all energy parameters of the cavitation region are correctly evaluated, since this region represents the active acoustic load of the electromechanical transducer (through the ultrasonic horn) and is the target "consumer" of all produced ultrasonic energy. We will, therefore, start by providing a detailed model of acoustic cavitation, explaining the mechanism by which the ultrasonic energy is absorbed in the cavitation region. A discussion of design principles of the main ultrasonic reactor system components will follow.

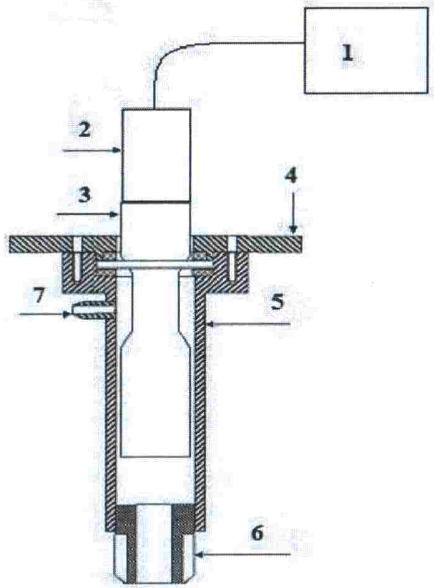

Figure 1. Schematic of an ultrasonic reactor system is presented. 1 – ultrasonic electrical generator, 2 – electromechanical transducer, 3 – ultrasonic horn (in this case, barbell horn), 4 – mounting flange, 5 – reactor chamber, 6 – working liquid inlet, 7 - working liquid outlet.

Chapter 2

SHOCK-WAVE MODEL OF ACOUSTIC CAVITATION

In order to properly design powerful ultrasonic sources for ultrasonic reactors, it is necessary to know the exact value of the intensity of acoustic energy radiated into the working liquid. This information is usually obtained experimentally because no adequate physical model of acoustic cavitation that would allow one to obtain such data through calculation exists. The development of an adequate model of acoustic cavitation, although of great importance, has in the past been severely restricted by considerable mathematical difficulties associated with the necessity of finding numerical solutions to nonlinear equations describing the cavitation region (visible region of large cavitation bubble population) [3]. The utilized direct analytical solutions of these equations in different approximations do not give practical results suitable for the design of ultrasonic equipment [4, 5].

The literature on acoustic cavitation mainly tends to involve numerical models of spatio-temporal characteristics of the cavitation region [6-8]. Large number of theoretical acoustic cavitation models has been developed along with the corresponding methods of numerical analysis of such models. Computer simulation-based investigations of acoustic cavitation have also been proposed, involving complex non-linear physicomathematical models and including many aspects of spatial movement of cavitation bubbles in an acoustic field, spatial distribution of the characteristics of these fields in a liquid, interaction between the bubbles themselves, properties of acoustical flow, etc [9-12]. Water is most frequently used for the experimental verification of such theoretical models.

No adequate explanation of the mechanism by which dissipation of the primary acoustic energy of a radiator occurs in a liquid at cavitation is, however, available from the literature. Additionally, no theoretical method permitting to calculate this energy in a manner adequate to the available experimental data currently exists. Meanwhile, the exact knowledge of the mechanisms by which heating of a liquid in the presence of cavitation-inducing acoustic waves occurs is important not only for the understanding of the related sonochemical processes, but also for the practical design calculations that would permit constructing improved high-capacity ultrasonic radiators and reactors.

2.1. Visual Observations of Acoustic Cavitation

Several authors provided common [13], high-speed [14] and stereoscopic high-speed [15] photographs of the cavitation region, obtained in the presence of relatively low-intensity acoustic fields. At these conditions, the cavitation region is located some distance away from the radiating surface and has a typical pattern similar to that of an electrical discharge.

Photographs of the cavitation region formed by powerful ultrasonic radiators have also been provided [16, 17]. The diameters of the radiating surfaces of the radiators were greater than the sound wavelengths in the given liquid at the working frequencies. In these cases, plane acoustic waves are radiated into the liquid. The photographs show that at relatively low acoustic radiation intensity, the cavitation region is also located some distance away from the radiating surface, has an irregular pattern and is composed of thread-like collection of cavitation bubbles. As the radiation intensity goes up, however, the cavitation region approaches the radiating surface and grows in size. When the intensity reaches the value of, approximately, 1.5 W/cm^2, the cavitation region “sits” on the radiating surface and its shape starts to resemble an upside-down circular cone. The so-called “cone bubble structure” begins to form. Further radiation intensity increases have little effect on the shape and position of the cone bubble structure. The photographs in the abovementioned studies show that at high radiation intensity the cone bubble structure is in contact with the radiating surface. Reference [18] provides photographs of the radiating surface of a metal radiator which was utilized for a period of time to create high-intensity cavitation in a liquid. The surface of the radiator contains clear traces of metal degradation due to cavitation.

Therefore, it can be concluded with certainty that at high radiation intensities, acoustic cavitation starts at the surface of the acoustic radiator. This location in the liquid is known, according to theory, to have the lowest value of tensile strength due to the constant presence of adsorbed gas inclusions at the metal surface [4].

However, at low radiation intensities just above the cavitation threshold, the cavitation region is always formed at a significant distance away from the radiating surface, which contradicts the abovementioned theory. Clearly, the tensile strength of the liquid at any location away from the metal surface should be higher than near it, since the concentration of the preexisting bubbles (inceptions) that "weaken" the liquid at that location should diminish with time.

2.2. JUSTIFICATION FOR THE SHOCK-WAVE APPROACH

At low radiation intensity, harmonic acoustic wave is not capable of inducing cavitation even at the weakest location in the liquid near the radiating surface. Formation of cavitation away from the radiating surface in this case can be explained by the effect of the increase of the planar acoustic wave-front steepness during its propagation through a liquid. As a result of such an increase, at some location in the liquid a discontinuity in the wave profile is formed. Since such discontinuity is physically not possible in a continuous media, a shock-wave with a steep front is formed as a result. This effect has to do with the acoustic radiation-induced nonlinearity of the compressible media properties and is very well known and documented [19].

This explanation, however, seems contradictory to the common shock-wave theory, since the attainable amplitude of vibration velocity of the radiating surface is always much lower than the speed of sound in the pure liquid and, therefore, the necessary conditions for the creation of such a discontinuity in the wave profile are not fulfilled. The explanation may, nevertheless, still be valid due to the following two considerations. It is well known that during propagation of an acoustic wave of slightly lower intensity than the cavitation threshold, an ensemble of tiny bubbles is formed in the liquid. This occurs due to the so-called "rectified diffusion" [4]. It is also well known that the speed of sound in a liquid containing gas bubbles is significantly lower than that in a pure liquid [20, 21], and, under certain conditions, it may become similar to the amplitude of vibration velocity (the radiating surface.

It may, therefore, be considered that bubbles formed in an acoustic wav due to rectified diffusion help forming a discontinuity in the profile of th acoustic wave at a location away from the radiating surface by significantl lowering the sound speed in the liquid. Further, at the location of th discontinuity in the acoustic wave, these tiny bubbles begin to undergo suc rapid nonlinear movements that they loose dynamic stability an consequentially, rapidly multiply forming the cavitation region.

The abovementioned observations and analysis formed the basis of th shock-wave model of acoustic cavitation described in this section. Th model shows how the primary energy of an acoustic radiator causin cavitation in a liquid is absorbed in the cavitation region owing to th formation of spherical shock waves inside each cavitation bubbl Calculation of the total energy absorbed in the cavitation region using th concept of a hypothetical spatial wave moving through the cavitation regio is possible with this model using the classical system of the Rankine Hugoniot equations. Additionally, the proposed model makes it possible t explain some newly discovered properties of acoustic cavitation of water th occur at extremely high oscillatory velocities of the radiating surfaces.

2.3. THEORY

Let us assume that an acoustic radiator emitting a plane-wave is used t generate cavitation in a liquid. The diameter of the radiator's output surfac is comparable with the length of the acoustic wave in the liquid at the give frequency of vibrations. The vibration frequency is much lower than th resonance frequency of the cavitation bubbles. We assume that the liqui always contains an equilibrium concentration of dissolved gas as well a some cavitation nuclei (tiny spherical bubbles filled with the gas) and consequentially, the liquid possesses no tensile strength during rarefactio caused by acoustic waves. As, for example, indicated in reference [4], wate that has not been purified of gas inclusions ruptures at a negative acousti pressure of, approximately, 1 bar. The density of the liquid with the tin cavitation nuclei is taken to be equal to the density of the pure liquid, ρ_l Surface tension of the liquid and the presence of stable (non-cavitational) ga bubbles are neglected. Thus, within the framework of the model, only the so called low-frequency transient gas cavitation is considered. We, additionally assume the liquid to be non-viscous, non-compressible and non-volatile.

Let us represent acoustic cavitation in the liquid as a sequence of the following events. When an acoustic rarefaction wave of certain amplitude passes through a volume of the liquid, an explosive growth of cavitation nuclei occurs, leading to the formation of the gas-filled cavitation bubbles. Possible parameters of such a rarefaction wave are described, for example, in [22]. A mixture of the spherical bubbles and the liquid is, therefore, formed. The gas dissolved in the volume of the liquid passes inside the free space formed by the bubbles. The density of the liquid medium, therefore, drops. At this point, the bubbles are so small, compared to the acoustic wavelength, that the liquid/bubble mixture can be considered a continuous medium. The rarefaction wave phase is followed by a compression wave phase, whose passage results in a collapse of all gas bubbles, restoring the density of the liquid to ρ_f. The reverse diffusion of the gas back into the liquid during compression is insignificant and should be ignored. This particular stage of acoustic cavitation completes the total cavitation cycle and is further considered here in great detail, since it is this stage that is mainly responsible for the sonochemical effects of acoustic cavitation.

2.3.1. Oscillations of a Single Gas Bubble

The problem of the liquid motion during compression of an empty spherical bubble in liquid was solved by Rayleigh (see reviews [4, 5]). On the basis of this solution and Ref. [19], the instantaneous pressure distribution in the liquid can be written as:

$$p = p_\infty + \rho_f \frac{\dot{U}r + 2U^2}{\xi} - \rho_f \frac{U^2}{2\xi^4} \quad (1)$$

Here, p_∞ is the pressure in the liquid at infinity, U is the velocity of the bubble boundary (wall), $\xi = R/r$, r is the current bubble radius, and R is the current radial coordinate. For the boundary of a gas-filled bubble at $\xi = 1$, the following equality must be met:

$$p_g = p_\infty + \rho_f (\dot{U}r + \frac{3}{2}U^2) \quad (2)$$

Here, p_g is the gas pressure in the bubble. This expression is the well-known Noltingk-Neppiras equation (see reviews [4, 5]).

For an empty bubble, taking $p_g = 0$ and $p_\infty = p_0$, integration of equation (2) gives Rayleigh's equations for the velocity of the bubble wall movement and the time of the bubble collapse:

$$U^2 = \frac{2p_0}{3\rho_f}\left(\frac{r_{in}^3}{r^3} - 1\right)$$

$$\tau = 0.915 r_{in}\left(\frac{\rho_f}{p_0}\right)^{0.5} \tag{3}$$

Here, p_0 is the static pressure, and r_{in} is the initial bubble radius.

From equations (1) and (2), an expression for the instantaneous distribution of the pressure in liquid during the compression of a gas-filled bubble can be obtained:

$$p = p_\infty\left(1 - \frac{1}{\xi}\right) + \frac{p_g}{\xi} + \frac{\rho_f U^2}{2}\left(\frac{1}{\xi} - \frac{1}{\xi^4}\right) \tag{4}$$

Let us single out a spherical liquid volume that includes a gas bubble. The gas bubble/surrounding liquid system has a certain acoustic compressibility, which determines the velocity of the propagation of small perturbations or the velocity of sound in this volume. Using the linearized form of the Noltingk-Neppiras equation, one can obtain an expression for the velocity of sound in such a system, as it was done, for example, in the work [21]. The velocity of sound, with the abovementioned assumptions taken into account, is determined using the following expression:

$$c = \left(\frac{p_g}{\rho_f \alpha (1 - \alpha)}\right)^{0.5} \tag{5}$$

Here, α is the volumetric gas concentration in the singled-out liquid volume that includes a gas bubble. From equation (5) it can be seen that the velocity of sound at a given gas pressure in the bubble has a minimum at $\alpha = 0.5$. For example, at $p_g = 1$ bar the minimum velocity of sound $c_{min} = 20$ m/s. It should also be noted that the velocity of sound in the range $0.4 < \alpha < 0.6$ changes little.

A gas bubble is formed during the half-period of the liquid rarefaction in the acoustic wave. Under the abovementioned assumptions, this occurs at the moment when the pressure in the liquid near the wall of a cavitation nucleus decreases to zero, i.e. the negative acoustic pressure is equal to p_0. At that point, the gas pressure in the formed bubble is also very small. Further, during the subsequent period of increase in the acoustic pressure, the bubble is compressed, and the gas pressure in it also increases. During the subsequent compression half-period, in the singled-out liquid volume near the gas bubble wall a spherical flow in the direction of the bubble center is formed, which is described by equation (4). From equation (5) it is seen that the velocity of sound for the singled-out system gas bubble/surrounding liquid depends on the gas pressure in the bubble p_g and the value of coordinate ξ, along which the boundary of the singled-out volume passes. If we start reducing the singled-out volume, while the radius of the bubble and the gas pressure in it are constant, the velocity of sound in this system will fall to a certain limit and then will grow again. This means that in the considered spherical volume near the moving wall of the bubble, there is a critical spherical region, where the sound velocity, c_{min}, is at the minimum at a given gas pressure in the bubble, p_g. The position of this region is determined from the condition $0.4 < \alpha < 0.6$. It is located close to the bubble wall in the coordinate range $1.18 < \xi < 1.35$. For the simplicity of further analysis of equation (4), it is taken that the velocity of the flow of the liquid particles in the critical region is equal to the velocity of the bubble wall movement, U.

In the model being considered, it is assumed that when the gas bubble/surrounding liquid system is compressed by the external pressure, p_∞, the velocity of the flow of the liquid particles in the critical region near the bubble wall increases to such a degree that at a certain gas pressure in the bubble, p_g, it reaches the minimum velocity of sound in the system under consideration, i.e. $U = c_{min}$.

At a ratio of the initial radius of an empty bubble to its current radius, $r_m/r = 2$, and static pressure, $p_0 = 1$ bar, the value of $U \approx 21$ m/s reached according to equation (3) is indeed close to $c_{min} = 20$ m/s.

Let us represent the pressure at infinity as a sum of the static and the acoustic (excessive) pressures, $p_\infty = p_0 + p'_\infty$ and transform equation (4) taking into account that $U = c_{min}$:

$$p = (p_0 + p'_\infty)(1 - \frac{1}{\xi}) + \frac{p_g}{\xi} + 2p_g(\frac{1}{\xi} - \frac{1}{\xi^4}). \quad (6)$$

This expression describes the extreme condition of equilibrium of the system. Equation (6) shows that during compression of the flowing liquid, in the vicinity of the gas bubble a pressure impulse is formed, which is stationary with respect to the bubble wall. The amplitude of the excess pressure in this impulse is $p - p_0 = 1.4p_g + 0.5\ \delta p'_\infty$, where $\delta p'_\infty = (p'_\infty - p_0)$. This value is reached at the coordinate $\xi \approx 2$ located upstream from the critical region. As we show below, the quantity, $\delta p'_\infty$, does not need to be considered for small oscillation velocities of acoustic radiators.

When the velocity of the bubble wall motion exceeds the minimum velocity of sound, $U > c_{min}$, the equilibrium state described by equation (6) becomes destroyed, and the pressure in the liquid at the bubble wall downstream from the critical region decreases to p_0. The velocity of the bubble wall movement also reduces because the driving pressure difference decreases. At the same moment, the excessive pressure amplitude in the impulse increases stepwise up to the value $p - p_0 = 1.4p_0 + 0.5\ \delta p'_\infty$, since the boundary condition in equation (2) is changed and the pressure near the bubble wall becomes $p_g = p_0$. This occurs because the bubble pressure signal does not penetrate upstream from the bubble wall when $U > c_{min}$.

Due to destruction of the dynamic equilibrium (retardation of a part of the flow), the pressure impulse located in the liquid upstream from the critical section disintegrates and begins to move relative to the bubble boundary in the form of a converging spherical wave. The supposed instantaneous distribution of excessive pressure in the impulse near the gas bubble wall at $U = c_{min}$ is shown in Figure 2.

Phenomena similar in essence are observed during the breakup of arbitrary pressure discontinuity in a gas, during hydraulic impact, and during the flow of gases and gas-liquid mixtures through nozzles. See, for example, the works [6, 8], as well as the studies on Laval nozzles and water hammers.

In accordance with the assumed form of pressure distribution in a converging spherical wave shown in Figure 2, the excessive pressure at the bubble wall first increases smoothly up to the value of $p - p_0 = 1.4p_g + 0.5\delta p'_\infty$ and, accordingly, the gas pressure inside the bubble increases smoothly (isothermally) as well. Then, when an abrupt excess pressure jump (up to the value of $p - p_0 = 1.4p_0 + 0.5\delta p'_\infty$) approaches the bubble wall, a spherical shock wave is formed in the gas inside the bubble. The pressure jump itself, evidently, is equal to $1.4(p_0 - p_g)$. After focusing in the center of