9 The principles of cavitation

T.G. LEIGHTON

9.1 Introduction: the characterization of cavitation

Acoustic cavitation has in the past been defined in various ways. The area of concern of this book is the use of ultrasound in food processing, so that most relevant applications of cavitation will involve ultrasonic fields of high acoustic pressure amplitude. The bubble is driven into some form of response by an externally applied acoustic pressure, and it is useful to consider cavitation to be the 'activation' of a gas inclusion in a liquid by an acoustic field. This does not therefore include the emission of sound from a gas bubble on entrainment in a liquid which, however, shall be discussed briefly as it is of interest to a range of applications, including industrial injectors, and because it illustrates the existence of the bubble resonance.

In liquids of density ρ, with ambient pressure p_0, a gas bubble of radius R_0 has a well-defined resonance frequency f_0 equal to:

$$f_0 = \frac{\omega_0}{2\pi} = \frac{1}{2\pi R_0}\sqrt{\frac{3\kappa p_0}{\rho}} \tag{9.1}$$

where κ, the polytropic index, varies between unity and γ, the ratio of the specific heats of the gas at constant pressure and constant volume. (The expression is valid for air bubbles in water, if the radius R_0 is greater than around 10 μm. When the ambient pressure is 1 atmosphere, the expression reduces to $f_0R_0 \approx 3$ Hz m. For more general expressions, see Leighton 1994a.) Such bubbles pulsate as lightly damped oscillators, emitting sound. This can be observed when a gas bubble is injected into water (Minnaert, 1933; Leighton and Walton, 1987). However there is a range of other entrainment modes, as can be illustrated from the scenarios involving liquid impact: waterfalls (Leighton *et al.*, 1995a); rainfall over water (Pumphrey and Crum, 1990); or breaking ocean waves (Medwin and Beaky, 1989). Upon entrainment, the bubbles undergo shape and volume changes (Leighton *et al.*, 1991). Except in certain circumstances (Longuet-Higgins, 1989), the shape changes are not efficient radiators of sound (Strasberg, 1956). It is the volumetric pulsations which generate the characteristic 'signature' typical of an entrained bubble, i.e. an exponentially decaying sinusoid, the frequency of which indicates the bubble size (Minnaert, 1933; Strasberg, 1956). A few milliseconds after injection these passive emissions have decayed to below the level of the noise. However the bubble may still be driven into oscillation by an externally applied sound field, and it is such behaviour which is the main concern of this chapter.

There is a wide range of phenomena which arise as a result of the pulsations or surface perturbations of the gas–liquid interface of the gas inclusion (Leighton, 1994a). Driven by a sound field, the bubble can undergo relatively stable, low energy oscillations (called 'stable' or 'non-inertial' cavitation), which can give rise to acoustical effects (including resonance and non-linear acoustic scattering). Optical and electrical effects may also be observed (Leighton, 1994b). The bubble may also undergo the higher-energy phenomenon of 'inertial' (formerly known as 'transient') cavitation. There exists a threshold, based on parameters relating to the sound field and the bubble, above which inertial cavitation will occur. It is relatively simple with a low-amplitude acoustic field to cause only non-inertial cavitation to occur in a given bubble population. However, if the aim is to generate inertial cavitation through the use of a higher acoustic pressure amplitude, then in an acoustic field the threshold conditions are usually exceeded only for some of the bubbles present. [As the acoustic frequency increases, the tendency is to reduce the proportion which undergo inertial cavitation (see Section 9.5.1).] The result is that both inertial and non-inertial cavitation will occur throughout the cavitation field, and will influence one another (Leighton, 1995a). Non-inertial cavitation can cause a range of acoustic, chemical, biological and erosive effects (Leighton, 1995b). Theoretical descriptions of cavitation have been available since the start of the century. It was the presence of erosive pits in propellers which inspired Lord Rayleigh (1917) to develop his pioneering analysis for the collapse of an empty spherical cavity under a static pressure. Coupling this energetic collapse phase with the explosive growth phase of a bubble (Blake, 1949), Noltingk and Neppiras (1950) characterized a particular type of cavitation, in which appropriately small bubbles in sufficiently strong sound fields undergo growth to many times their original size, and then a subsequent rapid collapse. It has since been verified that such events concentrate the acoustic energy, generating high gas temperatures (Neppiras and Noltingk, 1951), shock waves in both the gas (Vaughan and Leeman, 1986) and the surrounding liquid (Rayleigh, 1917), and free radicals (Suslick *et al.*, 1986). The latter may be involved in sonochemical reactions (Suslick *et al.*, 1986), and may generate sonoluminescence (Leighton, 1994a). Flynn (1964) distinguished the so-called 'transient' cavitation with which such manifestations are associated, from the less energetic 'stable' cavitation, where the bubble pulsates about an equilibrium radius over many acoustic cycles. Perhaps not surprisingly, therefore, this field which grew from the observation of erosive damage in propellers developed into a problem coupling fluid dynamics with acoustics, and indeed acoustic techniques feature amongst the most common ways in which cavitation is initiated and measured (Leighton, 1995b).

Though in common use the distinction between *transient* and *stable* (or *inertial* and *non-inertial*) cavitation is inexact, many formulations have concentrated on attempting to distinguish the threshold values of acoustic frequency, pressure amplitude, and initial bubble radius corresponding to the transition between the types of cavitation (Neppiras, 1980; Apfel, 1981; Flynn and Church, 1988; Holland and Apfel, 1989; Leighton, 1994a). However, once the threshold has been exceeded, the extent to which cavitational effects are generated (the 'activity' of the cavitating field) is a key parameter (see Section 9.6).

With rare exceptions (Gaitan and Crum, 1990), whilst theory has tended to concentrate on the dynamics of single bubbles, in practice when inertial cavitation causes a given effect, it is generally *populations* of bubbles that are involved (Leighton, 1995a). Whilst the exploitation of cavitation in many common applications (such as cell killing, ultrasonic cleaning, cavitation erosion and sonochemistry) relies on inertial cavitation (Leighton, 1995b), the application of these effects depends upon the characteristics of the *bubble population* and its interaction with the sound field. These two key factors may be influenced by other forms of cavitation, affecting nucleation and the relatively low-energy phenomena of bubble migration, dissolution and exsolution (Leighton, 1995a).

An experimental characterization of cavitation which is to be of use to common applications must therefore account for the following problems. First, whilst most applications involve bubble populations, the models and theories overwhelmingly rely on single-bubble descriptors, descriptors which are themselves only approximate. Second, when inertial cavitation occurs in a sound field and produces a range of effects (chemical, erosive, biological or acoustic), non-inertial cavitation may also occur, potentially not only influencing the inertial cavitation and its consequences, but also producing effects of its own.

9.2 Types of cavitation

Conceptualizations of bubble dynamics begin with models of isolated spherical bubbles. These models in turn become modified to allow the incorporation of experimentally observed phenomena. In this way have arisen the well-known classifications of transient and stable cavitation (Neppiras, 1980; Apfel, 1981; Leighton, 1994a). An impression of the range of bubble behaviours is shown in Figure 9.1. Neither of the phenomena shown in the first two rows (slow dissolution or growth of a bubble) will give

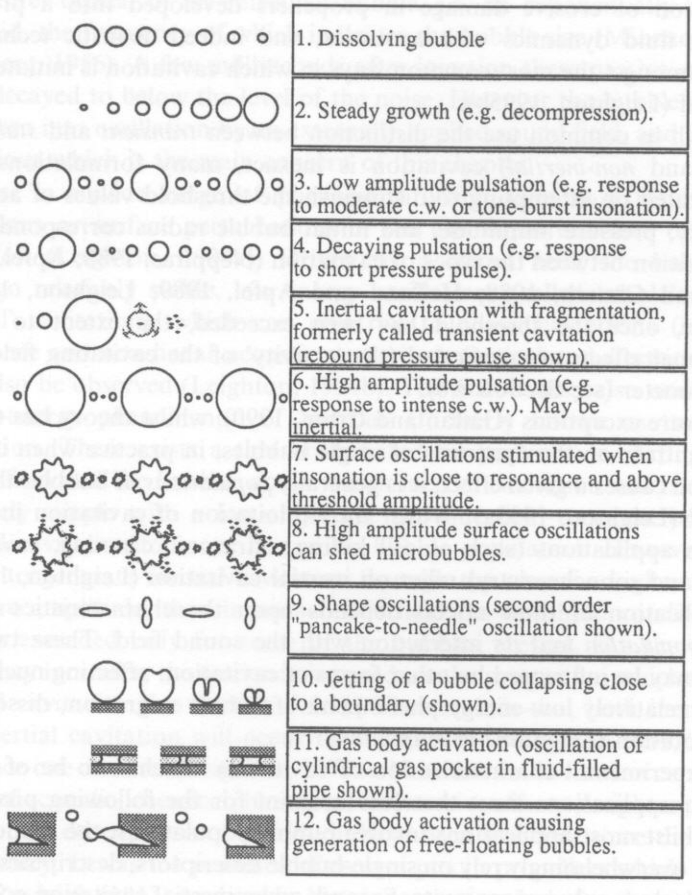

Figure 9.1 A schematic illustration of a range of bubble behaviour. Rows 3–12 are effects through which the bubble can be expected to be acoustically active. The expansion ratios drawn for these bubbles are exaggerated to more clearly illustrate the pulsations.

rise to acoustic emission. However, pulsation will give rise to such emission, and as such can be detected acoustically at a distance. As described in the previous section, the spherical bubble is, to a first approximation, an oscillator with a single degree of freedom, with a resonance frequency f_0 which is determined by the ratio of the stiffness to the inertia. The stiffness is determined by the compressibility of the gas content within the bubble, whilst the inertia is invested principally in the surrounding liquid, which must move if the bubble wall does (Leighton, 1994a).

As an oscillator, the bubble can be driven into pulsation by an incident sound field. If it is driven such that the amplitude of oscillation of the bubble wall is low (for example, the incident sound field has a low acoustic pressure amplitude, or the bubble has a size which differs greatly from that required for resonance with the driving frequency, bearing in mind that we are at present considering only models of bubbles which remain spherical at all times), then the bubble will scatter the sound field. If an ultrasonic beam is incident on the bubble, acoustic energy will be converted to heat through viscous and thermal damping mechanisms associated with the bubble motion (Devin, 1959; Eller, 1970) and scattered out of the beam (to first order as a spherical re-radiation) by the bubble. Row 3 of Figure 9.1 shows just such an oscillation. Despite damping, the illustrated bubble pulsations are shown not to decrease in amplitude, suggesting a continuous wave or tone-burst insonation, rather than a short pulse (which is the case illustrated in row 4). If pulsations of this type are of sufficient amplitude, this form of cavitation can strongly influence the effects of power ultrasound. First, such bubbles can directly generate physical and biological effects. These are usually associated with acoustic radiation forces. For example, cell disruption may be brought about because the bubbles can travel rapidly through the liquid under the influence of acoustic radiation forces (Section 9.4), generating hydrodynamic stresses which can, for example, produce haemolysis (Miller and Williams, 1989). Second, such bubbles can strongly influence both the population of nuclei available to seed inertial cavitation, and the acoustic pressure amplitudes to which those nuclei are subjected (see Section 9.5 and Leighton, 1995a).

It is well known that when ultrasound of sufficient intensity passes through a liquid containing bubbles which initially have radii that lie within a critical size range (Section 9.5), such gas inclusions may nucleate inertial cavitation (Neppiras, 1980; Apfel, 1981; Walton and Reynolds, 1984; Apfel and Holland, 1991). Inertial cavitation is typified by the sudden expansion and then rapid collapse of the bubble (row 5 of Figure 9.1). During the rapid collapse, the gas within the bubble is compressed. It may be heated to high temperatures (Walton and Reynolds, 1984; Suslick *et al.*, 1986; Kamath *et al.*, 1993), and gas shocks may propagate (Wu and Roberts, 1993). Electrical discharge and other effects may be associated with this phenomenon (Margulis, 1985; Lepoint *et al.*, 1993). Mechanisms for the generation of sonoluminescence based on all these, and yet more (Crum and Cordry, 1993; Eberlein, 1996), processes have been proposed.

Certainly the production of free radicals and electronically excited species has been associated with the extreme conditions that occur within the bubble. These can cause chemical and biological effects. However, the cavitation can also affect the medium through a range of associated mechanical processes [and indeed careful studies can distinguish, for example, the relative contributions of chemical and mechanical mechanisms to bioeffect (Miller and Thomas, 1993a, b; Miller *et al.*, 1995)].

One of the most notable such mechanisms occurs immediately after the collapse. The bubble rebounds, emitting a pressure pulse (shown figuratively in Figure 9.1, row 5 by arrows) into the liquid (Rayleigh, 1917; Noltingk and

Neppiras, 1950; Neppiras and Noltingk, 1951; Plesset and Prosperetti, 1977; Leighton, 1994a). This may cause mechanical damage. If clouds of bubbles are present, they may collapse co-operatively, enhancing this effect (Vyas and Preece, 1976). After the emission of such a pressure pulse, the bubble may fragment or repeat the growth/collapse cycle a number of times. For many years it was thought that the type of bubble activity which gave rise to such phenomena as sonoluminescence would involve fragmentation of the bubble after one (as shown in Figure 9.1, row 5) or a few cycles, reinforcing the labels of 'transient' cavitation and, for less energetic bubble oscillations, 'stable' cavitation. Following the discovery that in specialized conditions the bubble can pulsate for thousands of cycles (in a manner similar to row 6 of Figure 9.1), emitting a sonoluminescent flash at each collapse (Gaitan and Crum, 1990; Barber *et al.*, 1992), this terminology clearly needed replacing. Since models of the bubble collapse had shown that, to achieve the energy concentration expected to generate what had been known as 'inertial cavitation' the inertia of the liquid had to play a dominant role during the collapse, the terms 'inertial' and 'non-inertial' replaced 'transient' and 'stable'.

The generic oscillation shown in row 6 of Figure 9.1 is a high-amplitude pulsation of a spherical bubble (Figure 9.2). Depending on the amplitude, such oscillation may be inertial (as described above for specialized conditions) or non-inertial, but of high amplitude. If non-inertial, there are a number of interpretations of how this situation, which differs in wall oscillation amplitude from that shown in row 3, arises. It might, for example, occur if rows 3 and 6 show the same bubble, but in row 6 the acoustic pressure amplitude of the driving field is greater. Through such high-amplitude pulsations a second mechanism for mechanical damage may occur, resulting from the hydrodynamic shear stresses close to the bubble wall. The high amplitude pulsation indicates good coupling of the bubble to the sound field, and the bubble can move rapidly under acoustic radiation forces (see Section 9.4), traversing the medium as described in the previous section. Radiation forces are particularly strong on resonant bubbles, and indeed an increased amplitude of wall oscillation might be expected if the bubble is closer to resonance size. This gives a second source for the difference between rows 3 and 6: the bubbles in these rows now might be in the same sound field, but with the bubbles in row 6 being closer to resonance. However, as the bubble is more closely driven to resonance, other effects begin to occur. Most notable of these are surface waves (Figure 9.3b), which visually cause a 'shimmer' to appear on the surface of the bubble (Kornfeld and Suvorov, 1944; Neppiras, 1980; Phelps and Leighton, 1997), and acoustically may be detected through combination-frequency techniques (Phelps and Leighton, 1996). Such surface waves are illustrated in row 7 of Figure 9.1. They can be associated with an erratic 'dancing' translational motion (Crum and Eller, 1969), and at high amplitude

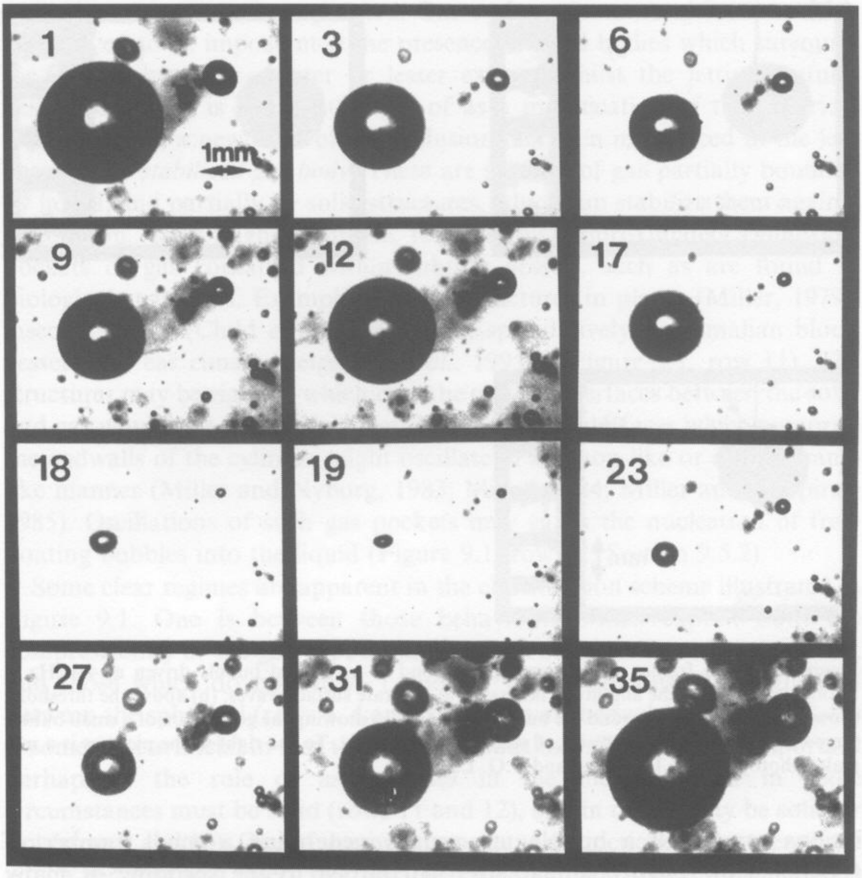

Figure 9.2 The oscillations of air bubbles in glycerol. The bubbles are driven by an inertially generated 100 Hz pressure field, of amplitude 3900 Pa, with 600 Pa static pressure. A selection of frames is shown from a sequence of 35 consecutive frames, filmed at 2000 frames per second. They illustrate the periodic unit in the motion of the largest bubble. The bubble contracts from maximum size in frame 1 to a minimum in frame 6, before expanding again to a second maximum (frame 12), then collapsing to a second minimum (frame 19), and finally expanding to reach, in frame 35, the same size as it had in frame 1. The second collapse is far more rapid than the first. After Leighton *et al.* (1990a). Reprinted by permission from the *European Journal of Physics*, vol. 11, pp. 352–358; Copyright 1990 IOP Publishing Ltd.

microbubbles might break off from the tips of the surface waves (Figure 9.1, row 8; Figure 9.3c; Leighton, 1995a). Surface waves can be associated with local circulation currents in the liquid, called microstreaming, which is a third mechanism by which an erosive effect can arise (Ahmad *et al.*, 1987).

Other departures from spherical symmetry include shape oscillations (Figure 9.1, row 9), found particularly in larger bubbles, where the tendency of surface tension to promote sphericity, is weaker (Strasberg, 1956; Longuet-Higgins, 1992a; Leighton, 1994a). If extreme, such shape oscil-

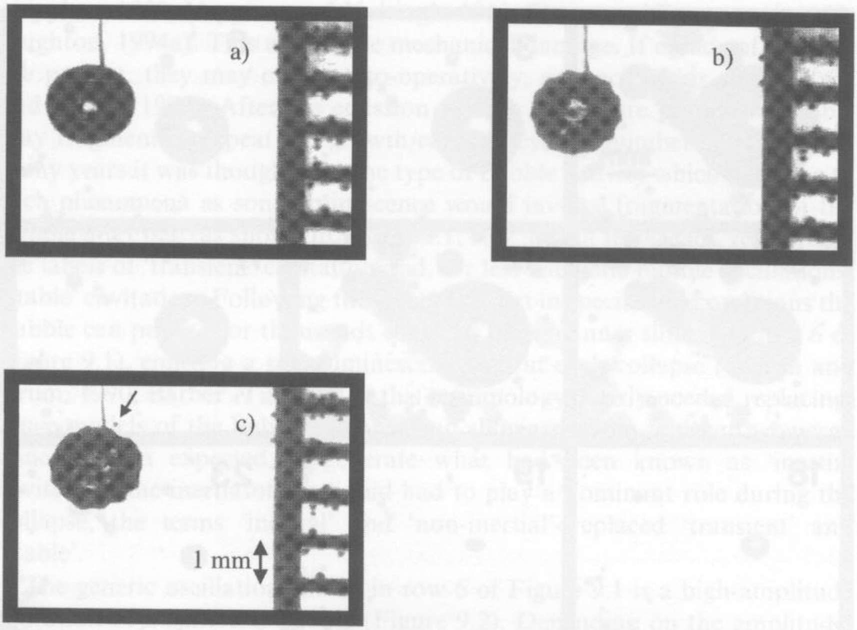

Figure 9.3 Video frames at 30 frames per second of a tethered bubble driven at 4.4 kHz at resonance (a) below the amplitude necessary to generate surface waves; (b) above the threshold, showing surface waves around the bubble wall; and (c) showing the generation of a small bubble (arrowed) pinched off as a result of surface wave activity. To the right in the pictures is a mm scale (photograph: A.D. Phelps and T.G. Leighton).

lations can break a bubble up, usually generating a small number of fragments of roughly similar size, in contrast to the shedding of many microbubbles from a larger parent bubble, shown in Figure 9.1, row 8 (Longuet-Higgins, 1992b; Leighton, 1995a; Leighton *et al.*, 1995a). Shape oscillations are encouraged by anisotropies in the local environment. Common causes for these are the presence of other bubbles, particles or walls. If such are present during bubble growth, then during the subsequent collapse the bubble may involute, one wall passing through the bubble to form a high-speed liquid jet (Plesset and Chapman, 1971; Lauterborn and Bolle, 1975; Prosperetti, 1984). This is shown in row 10 of Figure 9.1. Though the jetting of a bubble may be quite complicated, with, for example, the formation of counterjets, the net effect is that the bubble will usually fragment (Neppiras, 1980). Jetting is a fourth mechanism by which cavitation can cause mechanical damage, usually manifesting as the formation of pits in a solid body, where the liquid impact occurs.

As can be seen, the model of an isolated spherical bubble in an infinite fluid is proving inadequate to describe the range of behaviours seen: departures from spherical symmetry in both the bubble wall and the environment must be incorporated. One such environmental feature which has proved to be important is the presence of solid bodies which surround the gas pocket to a greater or lesser extent. Whilst the jetting feature, described above, is usually thought of as a modification of the spherical bubble model, a new class of gas inclusion has been introduced in the last decade, the *stabilized gas body*. These are pockets of gas partially bounded by liquid, and partially by solid structures, which can stabilize them against dissolution. They might comprise, for example, approximately cylindrical pockets of gas contained within tubular vessels, such as are found in biological structures. Examples include structures in plants (Miller, 1979), insect tracheae (Child *et al.*, 1981), and, speculatively, mammalian blood vessels and ear canals (Leighton *et al.*, 1995b) (Figure 9.1, row 11). The structures may be rigid, in which case the curved interfaces between the solid and gas would not move. However the gas/liquid interfaces which comprise the endwalls of the cylinder might oscillate in a piston-like or a membrane-like manner (Miller and Nyborg, 1983; Miller, 1984; Miller and Neppiras, 1985). Oscillations of such gas pockets may cause the nucleation of free-floating bubbles into the liquid (Figure 9.1, row 12; Section 9.5.2).

Some clear regimes are apparent in the classification scheme illustrated in Figure 9.1. One is between those behaviours which exploit both the compressibility of the gas/vapour, and the inertia of the liquid (rows 3–12), and those which do not (rows 1 and 2). Whilst obvious, the distinction between the spherical (rows 3, 4 and 6) and the aspherical (rows 5 and 7–12) is somewhat artificial in that the forms are idealized. Rather more important perhaps is the role of asymmetries in the media, which in some circumstances must be solid (rows 11 and 12), and in others may be solid or not (rows 9 and 10). One of the most important distinctions is between those which for maximal effect rely on the resonance, and those which rely on inertial cavitation. The importance of this distinction can be illustrated through reference to the common paradigm that a bubble will undertake a steady increase in equilibrium size through a process called rectified diffusion (Section 9.3), to eventually reach the radius which is resonant with the sound field. At this point it will cause maximum mechanical, sonochemical or sonoluminescence effect. This scheme has been used in some theories of pulse enhancement (Section 9.6), or cyclic cavitation (Neppiras, 1980). Whilst sometimes valid, this scenario is often used inappropriately. Consider, for example, a situation where ultrasonically induced cavitation erosion is observed (Figure 9.4). It might arise through microstreaming or bubble-mediated radiation forces (Section 9.4), both of which are maximal when the bubble reaches resonance size (rows 6 and 7). However damage by jetting is more dependent on the local geometry than the resonance (row 10); and damage through rebound pressure is dependent instead on the threshold for inertial cavitation. The source of the erosion will depend on the mechanism and the prevailing conditions. The case is

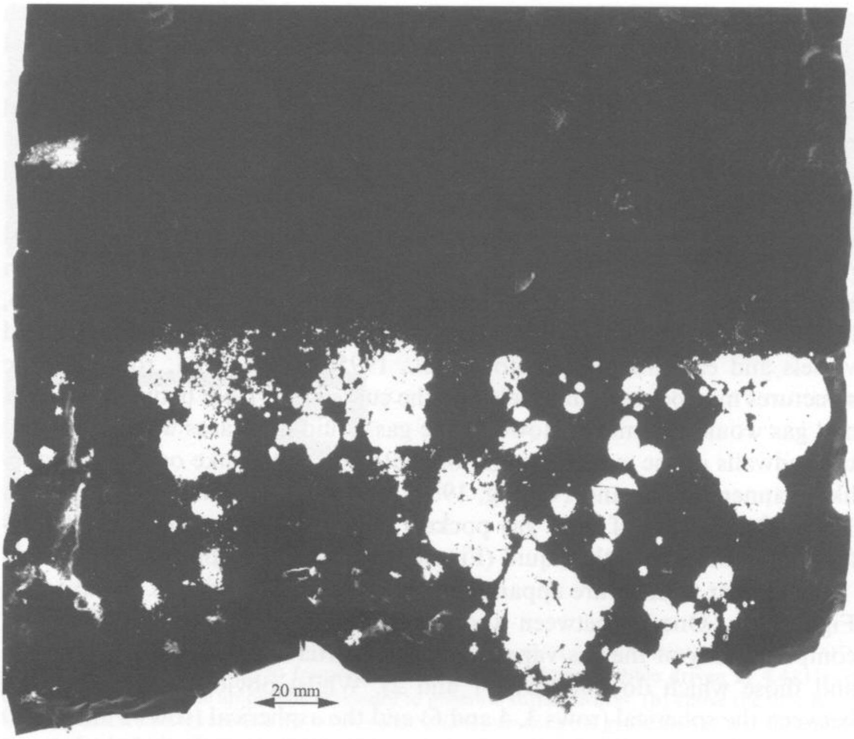

Figure 9.4 A back-lighted sample of aluminized mylar sheet, which had been subjected for around 1 minute to a cylindrical sound field in aerated water which, at the axial focus, had an acoustic pressure amplitude of 0.24 MPa. In the dark regions the aluminium has remained attached to the sheet. However in other regions it has been eroded away, such that the light shines through the transparent mylar in large, roughly circular regions (after Leighton 1994a). These can be seen to occur in the half of mylar which was submerged. The half which was not submerged is predominantly dark, the only surface removal there being a result of the mountings.

clearer for the generation of sonoluminescence and sonochemistry, which are dependent on the threshold for inertial cavitation. Therefore if the bubble is to grow by rectified diffusion to maximize a sonochemical or luminescent effect, it is likely to be the radius threshold for inertial cavitation which is critical, and not the resonance. Whether such schemes of bubble growth to critical sizes are likely when detailed bubble dynamics are incorporated into the theory is dependent on a number of parameters, not least the initial bubble size, the acoustic frequency, and the acoustic pressure amplitude (Church, 1988a). In the following sections the importance of these three parameters, and the characteristics of rectified diffusion and radiation forces on bubbles, will be expounded. Lastly, the issue of the measure of 'activity' of cavitation in an ultrasound field, and its suppression and enhancement, will be addressed.

9.3 Rectified diffusion

In many circumstances the size distribution of bubbles in a population is a key parameter in determining the acoustic effects. As introduced in the previous section, there is a critical size range in which a free-floating bubble must lie in order to undergo inertial cavitation. This will be examined further in Section 9.5.1. Similarly, the following section will detail the importance of the bubble size to the effect of acoustic radiation forces, and in turn how this can affect the cavitational activity in a population (Section 9.6). The interdependence and feedback between all these features is explored in Leighton (1995a).

The previous section also introduced the ability of a bubble to increase its equilibrium radius in an appropriate sound field through the process of *rectified* diffusion, which manifests as a net flux of dissolved gas out of solution and into the bubble. This flux is therefore counter to the direction that would be towards equilibrium were the sound field not present, since a gas bubble in a liquid will tend to dissolve owing to the excess internal gas pressure required to balance the pressure $2\sigma/R_0$ due to surface tension σ (Epstein and Plesset, 1950). Thus, in the absence of a sound field or any stabilizing mechanism, bubbles gradually dissolve (Gupta and Kumar, 1983). An appropriate stabilizing mechanism might be presented by the crevices shown in rows 11 and 12 of Figure 9.1: at equilibrium, the bubble wall is concave as seen from the liquid (row 11), as opposed to the convex meniscus of a spherical bubble, so that the pressure due to surface tension tends to stabilize the bubble against dissolution. Other mechanisms are discussed briefly in Section 9.5.1, and detailed in Chapter 2 of Leighton (1994).

In the presence of a sound field, the situation is quite different. During non-inertial cavitation, since evaporation and condensation take place so much more rapidly than the bubble dynamics, it is commonly assumed that, to first order, the vapour pressure within the bubble remains constant at the equilibrium value. However this is not so for the gas content of the bubble, a gas which will also be dissolved in the liquid. Harvey *et al.* (1944), studying the formation of bubbles in animals, suggested a mechanism by which bubbles undergoing stable cavitation in a sound field can experience a steady increase in their equilibrium radius, R_0. This inwardly directed *rectified diffusion* comes about through the active pumping of gas, initially dissolved in the liquid, into the bubble, using the energy of the sound field. There are two contributory elements to a full description of the processes, an 'area effect' and a 'shell effect'.

The area effect arises through the correlation between the direction of mass flux and the area of the bubble wall. While the bubble radius is less than equilibrium, the gas inside is at a greater pressure than the equilibrium value, and thus diffuses out into the liquid. Conversely, when the bubble radius is significantly greater than R_0, the internal gas pressure is less than the equilibrium value, and so gas diffuses from the liquid into the bubble interior. The net flow rate of the gas however, is not equal during the compressed and expanded phases of the bubble motion, because the area of the bubble wall (the transfer interface) is greater in the latter case than in the former. This process is illustrated over one acoustic cycle in Figure 9.5. Therefore, over a period of time, there will be a net influx of gas to the bubble interior.

The shell effect occurs because the diffusion rate of a gas in a liquid is proportional to the concentration gradient of the dissolved gas. As the bubble pulsates, a spherical shell of liquid surrounding the bubble will change volume, and so the concentration gradient will change. Consider the bubble shown at equilibrium in Figure 9.6(a). Two of the shells surrounding the bubble are illustrated schematically. When the bubble is expanded, each liquid shell contracts (Figure 9.6b). The concentration of dissolved gas in the liquid adjacent to the bubble wall is less than the equilibrium value (Henry's law), but the shell is thinner than when the bubble is at equilibrium radius, so that the gradient across the shell is higher. Therefore the rate of diffusion of gas towards and into the bubble is high. When the bubble is contracted, the liquid shells surrounding the bubble are expanded (Figure 9.6c). Although the concentration of gas near the bubble wall is higher than when the bubble is expanded (Henry's law), the increased thickness of the shell means that the concentration gradient is not as great as when the bubble is expanded. The two factors (gas concentration at the bubble wall, and shell thickness) work together when the bubble is expanded, but against one another when the bubble is contracted: On expansion there is a large concentration gradient driving gas a short distance, and in the second case a smaller gradient drives the gas a longer distance. The former effect is dominant.

The result is that both the bubble wall surface area and the dissolved gas diffusion rate are asymmetrical with respect to expansion and contractions:

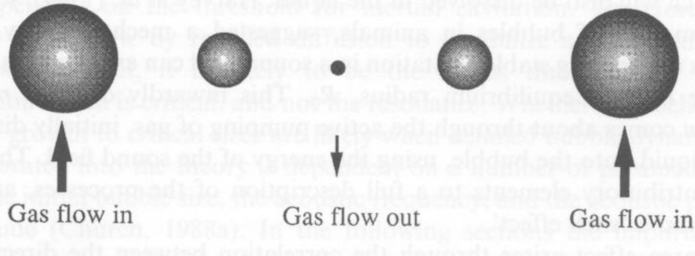

Figure 9.5 The 'area' effect. One oscillatory cycle of the bubble pulsation is illustrated, time increasing from left to right. The pressure within a pulsating bubble, and the surface area of the transfer (the bubble wall), both oscillate about an equilibrium value, causing a mass flux imbalance (after Leighton 1994a).

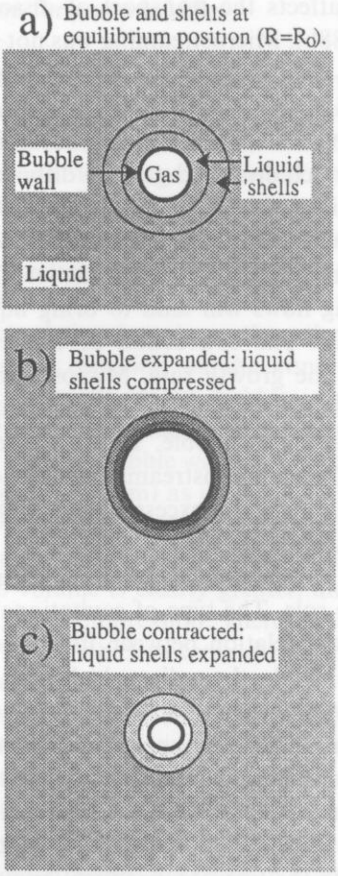

Figure 9.6 The 'shell' effect: the bubble and two of the liquid shells surrounding it are shown. (a) The bubble is at equilibrium size. (b) The bubble expands, and the liquid shells contract. (c) The bubble contracts, and the liquid shells dilate (after Leighton, 1994a). Reprinted by permission *The Acoustic Bubble;* Copyright 1994, Academic Press Ltd.

the 'area' and 'shell' effects reinforce one another. The combined effect means that during non-linear cavitation in acoustic fields which are sufficiently intense, the equilibrium radius about which the bubble pulsates will tend to increase (Eller and Flynn, 1965; Eller, 1972; Crum, 1984; Church, 1988a, b). Rectified diffusion of heat may also occur, whereby the bubble pulsation causes the establishment of thermal differentials against the gradients which would exist in the absence of a sound field (Wang, 1974).

The threshold acoustic pressure for growth is dependent on the bubble size and the acoustic frequency. Local minima in the acoustic pressure threshold are seen at harmonics and subharmonics of the resonance (Church, 1988a). The growth rate once the threshold has been exceeded is strongly influenced by the presence of surface-active agents, and by microstreaming, which affects the transport of dissolved gas beyond the bubble wall (Church, 1988b). Microstreaming was not incorporated into the above simple discussion, but its effects are qualitatively simple. As a bubble grows by rectified diffusion, the dissolved gas is taken from the liquid near the bubble. If there is no flow, then the rate at which the deficit is met depends on the rate at which dissolved gas can diffuse from regions farther out from the bubble. Since this is in general a slow process, the liquid outside the bubble wall will become depleted of dissolved gas. The resulting change in concentration gradient reduces the rate of further growth. However, microstreaming flows will tend to bring liquid from farther out close to the bubble wall. The convection of dissolved gas reduces the depletion, and increases the growth rate. Microstreaming will continually refresh the liquid at the bubble wall, giving it a dissolved gas concentration close to that found far from the bubble. The converse process is of course valid: if a bubble is dissolving, microstreaming will tend to remove from the region outside the bubble wall the excess dissolved gas concentration, so increasing the rate of dissolution.

Within the dynamics of a bubble population, rectified diffusion of gas may have a considerable role. The type of cavitational behaviour a bubble undertakes depends on the relation between its size and the other critical sizes (Figure 9.1). These include: the radius which is resonant with the sound field (governing, for example, radiation force effects, surface wave activity and microbubble shedding, etc.); the upper and lower limits of the radius range for the nucleation of inertial cavitation; and the threshold for rectified diffusion. Dissolution and fragmentation provide mechanisms by which bubble size reductions in the population can occur. Coalescence and rectified diffusion provide the ways to produce larger bubbles. How such changes affect the bubble size distribution with respect to the critical sizes mentioned above determines the bubble activity seen. For example, if a bubble fragments following an inertial collapse, and the resulting small bubbles are smaller than the size required to nucleate inertial cavitation, but larger than that required to grow by rectified diffusion, such growth can make inertial cavitation a self-nucleating process. Church (1988a) outlines some of these possible scenarios, and assesses their likelihood.

The two phenomena mentioned above, coalescence and bubble fragmentation, are dependent on inter-bubble spacing. For coalescence, the reasons are obvious. In fragmentation, the presence of a neighbouring bubble can excite the shape oscillations shown in row 9 of Figure 9.1 which, if sufficiently pronounced, can lead to bubble fragmentation (Leighton 1995a; Leighton *et al.*, 1995a). In an intense, continuous-wave or tone-burst acoustic field, among the most potent factors determining the inter-bubble spacing are the acoustic radiation forces: these are discussed in the following section.

9.4 Acoustic radiation forces on bubbles

In an accelerating liquid a gas bubble, being less dense than the surrounding fluid, will accelerate in the same direction as the surrounding liquid, but to a greater degree. That degree is governed by the magnitude of the difference in density between gas and liquid or, equivalently (assuming the bubble contains a fixed mass of gas), the instantaneous bubble volume. As an acoustic field passes through a liquid, the liquid particle acceleration oscillates, reversing direction periodically such that it is aligned with the direction of propagation for half the acoustic cycle, and contrary to it for the remainder. The bubble, being less dense than the liquid, will follow suit, accelerating in the same direction as the continually alternating liquid particle acceleration. However, if the bubble is pulsating with the same periodicity as the driving field (which a linear oscillator will do), then the phase relation is such that the bubble will be in the expansion phase (i.e. with its radius greater than equilibrium) as it travels in one direction, and in the compression phase as it travels in the other direction (Leighton *et al.*, 1990b). The bubble accelerates more during the expansion phase than the compression, since its volume is clearly greater, and its density less, during expansion than during the compression. Therefore such a bubble in a sound field will experience forces which reverse direction two times per acoustic cycle. However the net effect will be that it travels in the direction taken by the liquid acceleration when the bubble is in the expansion half-cycle. If the bubble is assumed to be an oscillator with a single degree of freedom, bubbles of less than resonance size will pulsate in antiphase to those of greater than resonance size. Therefore if one type of bubble is accelerated in one direction by the sound field, the other type will be forced in the opposite direction.

This behaviour is most readily observed in a standing wave field, where the radiation forces are commonly called ‘primary Bjerknes forces’. They cause bubbles of less than resonance size to travel up pressure gradients to collect at acoustic pressure antinodes, whilst bubbles of larger than resonance size migrate down the gradients to the nodes.

Similar comments apply in focused acoustic fields, where bubbles of less than resonance size migrate up pressure gradients towards the focus. To produce a bubble which is larger than resonance, it must either pre-exist (and continue to persist during insonation despite buoyancy and possible fragmentation); form through coalescence; or grow to a size larger than resonance by rectified diffusion. Acting against the latter scenario is the fact that it is on reaching resonance size that the bubble is most likely to lose gas through microbubble emission from surface waves (Figure 9.1, row 8). Also, if the bubble does pass through resonance intact, then rectified diffusion becomes far less efficient once a bubble exceeds resonance size, the pressure threshold increasing and the growth rate decreasing. There is also the general trend that the larger the bubble, the easier it is to excite shape oscillations (Figure 9.1, row 9) which can lead to fragmentation. Therefore in most fields, it is the aggregation of the smaller bubbles at the antinodes or the focus which is more commonly observed (Figure 9.7).

Formulations also exist for the radiation force on a bubble in travelling-wave conditions, where the force is greatest on resonant bubbles; and for the force on a particle suspended in a liquid close to a bubble (Coakley and Nyborg, 1978), an example of the latter being the aggregation of platelets in blood (Miller *et al.*, 1979). The so-called 'secondary' or 'mutual' Bjerknes force is exerted between two pulsating bubbles (Bjerknes, 1906, 1909, 1930; Prandtl, 1954; Batchelor, 1967; Leighton 1994a). There is a general rule that bubbles which are both less than, or both greater than, resonance size attract; but that if one bubble is greater than, and the other less than, resonance size, they repel. However, this is a simplification, and the results may be more complicated if the bubble population density is high or the incident sound field is strong. Such attractions can influence the size of individual bubbles by causing coalescence or, through the surface waves induced on one bubble by the proximity of its neighbour, fragmentation. If the bubble is oscillating close to a boundary (e.g. solid or free surface), migrations may be influenced through a secondary Bjerknes interaction between the bubble and its 'acoustic image', the wall acting as a form of

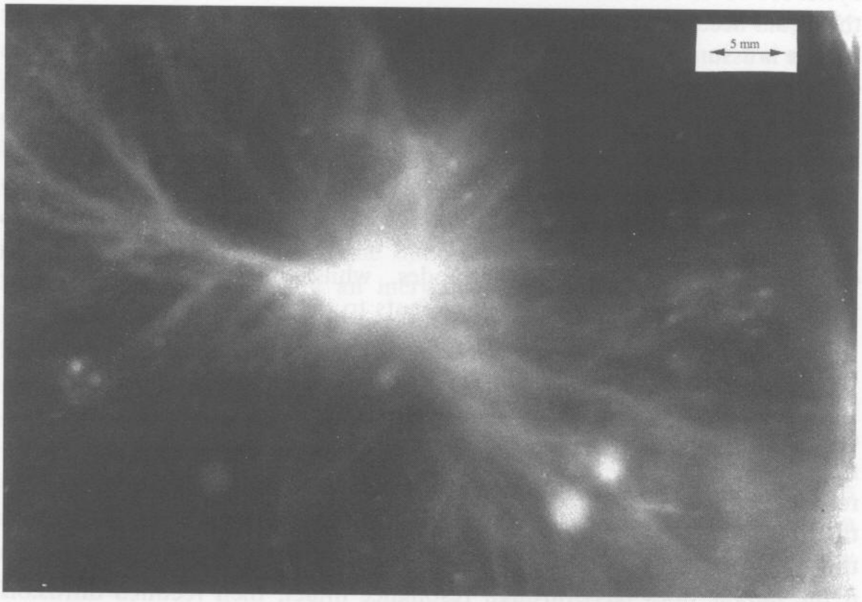

Figure 9.7 A photograph of aerated water cavitating in a cylindrically focused 10 kHz sound field, viewed along the line of the axial focus, where the acoustic pressure amplitude is 0.24 MPa. Exposure time 1/30th second. Streamers are clearly visible, comprising bubbles moving rapidly towards the focus, driven by radiation forces.

acoustic 'mirror' (Leighton, 1994a). Crevices in solids may attract bubbles, the subsequent expansion of which can extend the crevice (Brunton and Camus, 1970; Leighton, 1994a).

9.5 The thresholds between inertial and non-inertial cavitation, and the importance of stable gas bodies

Inertial cavitation is a threshold phenomenon, and whether or not it occurs depends critically on a number of parameters. The acoustic frequency is one, and this can be simply controlled [although the frequency content of an acoustic wave may change during propagation as a consequence of nonlinear effects (associated with, for example, the propagation of finite amplitude waveforms or parametric effects involving bubbles)]. The acoustic pressure amplitude of the sound field *at the bubble* is another critical threshold parameter. While the operator can readily determine the vibration of the transducer faceplate, and the acoustic field at the position of a monitor hydrophone, it is not so simple to control the field amplitude at any individual bubble. The third key parameter, the initial bubble size, is often the least controlled.

The bubble size and acoustic pressure amplitude are able to influence one another. Examples of ways in which the field can affect the size distribution were discussed in the previous sections (rectified diffusion; coalescence and fragmentation, possibly influenced by radiation forces). Other mechanisms for this, and for the effect of the population on the field, are key to the suppression or enhancement of cavitational activity.

This section therefore details the cavitation threshold, which determines the number of inertial cavitation events. Section 9.6 then describes how the 'activity' of the cavitation is a function of both the number of collapses and the ability of each to generate the effect in question; and how the suppression or enhancement of cavitational activity can be governed by the influence of the factors discussed in the preceding sections on the collapse dynamics outlined in this section.

9.5.1 The nucleation of inertial cavitation

The parameter which is often subject to least control is the initial size of the bubble. If it is too small, then surface tension forces prevent the initial sudden growth, and inertial cavitation does not occur. If it is too large, then the bubble may grow, but be too 'sluggish' to concentrate the energy sufficiently on collapse to generate free radicals, etc. There is therefore a critical size range in which, for a given sound field, the initial size of the bubble must fall if it is to nucleate inertial cavitation (Flynn, 1964; Flynn and Church, 1984; Holland and Apfel, 1989; Leighton, 1994a). The lower the frequency, the wider this range.

This is clearly shown in Figure 9.8, where the transition threshold between inertial and non-inertial cavitation is plotted, based on calculations by Apfel and Holland. They assumed that, in response to a single cycle of ultrasound, a bubble which is spherical at all times would grow and, on subsequent adiabatic collapse, the gas within the bubble should attain a temperature of at least 5000 K if the collapse is to be 'inertial' (Apfel, 1981; Apfel and Holland, 1991). Though there are clear approximations and the choice of such a criterion for defining inertial cavitation is not fundamental, this is nevertheless an extremely useful calculation, and illustrates that the acoustic pressure amplitude required to cause a bubble to undergo inertial cavitation is dependent upon the initial radius of that bubble. Since in most applications the frequency is the easiest of these three parameters to control, followed by the acoustic pressure amplitude at *the bubble,* with the radii of the nuclei present being the least accessible, then the graph can be interpreted in another manner. At a fixed frequency, say 10 MHz, an ultrasonic cycle with a peak negative pressure of 1.5 MPa (assumed to be constant throughout the field) will only generate inertial cavitation within a water sample if, according to this model, it contains bubbles having radii between 0.03 and 0.77 μm. As the pressure amplitude decreases, so does the range of bubble sizes which can nucleate inertial cavitation. The very lowest peak negative pressure which could give rise to inertial cavitation, according to this model, is P_{opt} (around 0.84 MPa at 10 MHz): and at this pressure

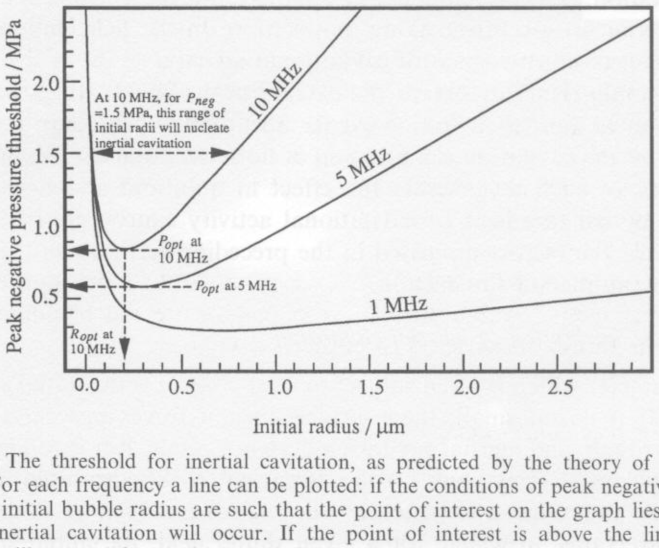

Figure 9.8 The threshold for inertial cavitation, as predicted by the theory of Apfel and Holland. For each frequency a line can be plotted: if the conditions of peak negative pressure and of the initial bubble radius are such that the point of interest on the graph lies below the line, non-inertial cavitation will occur. If the point of interest is above the line, inertial cavitation will occur (after Apfel and Holland, 1991). Reprinted by permission of Elsevier Science, Inc. from Gauging the liklehood of cavitation from short-pulse, low-duty cycle diagnostic ultrasound by Apfel R.E. and Holland C.K. *Ultrasound in Medicine and Biology*, Vol. 17, No. 2, pp. 179–185. Copyright 1991 by World Federation of Ultrasound in Medicine and Biology.

only bubble of a radius R_{opt} (0.2 μm at 10 MHz) could possibly nucleate inertial cavitation.

The theory assumes nucleation of inertial cavitation within the first acoustic cycle, so-called 'prompt cavitation', from a free-floating spherical bubble nucleus. In practice of course it is not a requirement that such nuclei be present before the start of insonation. Not only may bubbles which are initially too large to nucleate inertial cavitation enter the critical range (through, for example, dissolution, or fragmentation through a shape oscillation or microbubble shedding, as discussed in Section 9.3): bubbles too small to nucleate cavitation may enter it through rectified diffusion or coalescence. As discussed in Section 9.4, radiation forces can affect both size increases and size reductions. In Section 9.6 it will be shown how radiation forces in focused fields can convect suitable nuclei into the focus to nucleate inertial cavitation there.

The model employed to produce Figure 9.8 is based upon the dynamics of isolated, spherical bubbles, which *de facto* must be free floating. Other possible nuclei can be found naturally as gas pockets, stabilized against dissolution in crevices and cracks in the container wall or within free-floating particles within the liquid (Harvey *et al.*, 1944; Trevena, 1987; Atchley and Prosperetti, 1989; Leighton, 1994a). If inertial cavitation is undesirable, then such particles can be removed by filtering. However even this will not completely remove all suitable nuclei for inertial cavitation, which may be generated, for example, by the passage of cosmic rays through the sample (Greenspan and Tscheigg, 1967).

Crevice nucleation is illustrated in row 12 of Figure 9.1. High amplitude ultrasonic waves cause the gas pockets to either expand out of their crevice, or conceivably shed microbubbles through surface waves, to generate free-floating nuclei for cavitation. Such crevices are not the only mechanism through which a gas nucleus may be stabilized against dissolution: a hydrophobic 'skin' can collect on the bubble wall and cause free-floating nuclei to persist over long periods (Akulichev, 1966; Sirotyuk, 1970; Yount, 1979, 1982; Yount *et al.*, 1984).

The theory of Apfel and Holland has given rise to a measure called the 'mechanical index', which may be used to indicate the likelihood of exceeding the threshold required to nucleate cavitation.

9.5.2 *The mechanical index*

The model of Holland and Apfel investigates the threshold condition required to generate, within one acoustic cycle from the start of insonation, a collapse temperature of 5000 K. The threshold it predicts is relatively insensitive to the precise temperature criterion, and Holland and Apfel were able to compare their threshold predictions with those of Flynn and Church (1988), who used a different definition (based on the bubble achieving on expansion a maximum radius of at least twice its initial radius) for the onset of inertial cavitation, equivalent to the bubble achieving a maximum temperature of 960 K. A threshold for sonoluminescence of 1550 K has also been proposed (Sponer, 1990, 1991; Sponer *et al.*, 1990).

Apfel and Holland (1991) derive a *mechanical index,* which represents the likelihood that inertial cavitation will be nucleated, applicable (because of the assumption of 'prompt' cavitation) to microbubble growth in the limit of short-pulse, low duty cycle insonation. Clearly effects relating to longer insonation periods, such as growth by rectified diffusion, are not covered. However the index can be used to gauge the probability of inertial cavitation resulting from diagnostic ultrasound fields.

As described for Figure 9.8, the model demonstrates that for a given acoustic frequency there is a minimum in the curve. If the whole range of bubble size classes are present, then there is a specific initial bubble size for which the threshold acoustic pressure required to nucleate inertial cavitation is a minimum. As the insonation frequency increases, the bubble radius which requires minimum pressure to nucleate inertial cavitation decreases, since inertial and viscous forces increase with increasing frequency, and there is insufficient time to bring about the required amount of bubble growth. For the same reason the acoustic pressure required to nucleate inertial cavitation in all but the smallest bubbles increases with increasing frequency. Surface tension dominates the response of the smallest bubbles.

If one is interested in a worst-case assessment of the likelihood that inertial cavitation will occur when a liquid is insonated, clearly one must assume that the bubble population contains bubbles at the radius corresponding to the minimum in the threshold curve. At a given frequency, it is bubbles of this radius, R_{opt}, which the analysis predicts will require the smallest peak negative pressure, P_{opt}, to undergo prompt inertial cavitation in response to a single acoustic cycle. Apfel and Holland (1991) generate a plot of P_{opt} against frequency for water and whole blood, using pure fluid bulk property values for the σ, ρ and viscosity η relevant to the two fluids. The liquids are assumed to contain the relevant nuclei at size R_{opt}. Apfel and Holland employ a two-parameter least-squares fit to these plots in order to obtain a relationship between P_{opt} and the insonation frequency $f = \omega/2\pi$. They find a least-squares fit of:

$$\frac{(P_{\text{opt}})^a}{f} = b \tag{9.2}$$

where if P_{opt} is measured in MPa and f in MHz, the constant a takes values of 2.10 for water and 1.67 for blood, and b has values 0.06 for water and 0.13 for blood. For a given sound field with a maximum negative pressure of

P_{neg}, then by taking a value of $a \approx 2$ to approximate the appropriate physiologically relevant liquid, a mechanical index MI can be defined for the sound in that liquid

$$MI = \frac{(P_{neg}/\text{MPa})}{\sqrt{f/\text{MHz}}} \tag{9.3}$$

The mechanical index, *MI*, for prompt cavitation represents an approximate measure of the worst-case likelihood of nucleating inertial cavitation. As such it can be used to estimate the potential for nucleating inertial cavitation resulting from insonation by diagnostic ultrasound. Clearly it would be less appropriate to apply this index to tone-burst or continuous-wave insonations, during which a range of complicated processes, including rectified diffusion and enhancement of the cavitation by ultrasonic pulsing, can occur (Leighton, 1994a). Holland and Apfel recommend that the pulse length should not exceed 10 cycles, nor the duty cycle 1:100. The peak negative pressure output from the device, as measured in water, must be deaerated to give the appropriate peak negative pressure that would be attained *in vivo* at the location of the maximum pulse intensity integral (AIUM/NEMA, 1992). The centre frequency is used for f which, for accuracy, is expected to be of the order MHz. Apfel and Holland (1991) suggest that a mechanical index value below $\sqrt{0.5} \approx 0.7$ would indicate that, even in the presence of a broad size distribution of nuclei, the conditions are not sufficient to allow significant bubble expansion. If $MI \geq \sqrt{0.5}$, Apfel and Holland suggest that 'the user should be advised of the potential for bubble activity'. The AIUM, NEMA and FDA have adopted the mechanical index as a real-time output display to estimate the potential for cavitation *in vivo* during diagnostic ultrasound scanning (AIUM/NEMA, 1992).

Key points should be noted. First, the mechanical index gauges the likelihood of prompt cavitation, and nothing more: the effect of interest (e.g. a bioeffect) may be related to some other mechanism. Second, the model for the index is based on the assumption of a free-floating spherical nucleus of optimum size. In certain circumstances it may be that the nucleus is of a different type. Third, the effects of non-linear propagation are not included, and hence the index might underestimate conditions *in situ*. Fourth, when applied to diagnostic ultrasound instruments, the mechanical index describes conditions only at the focus, which is not necessarily the point of interest. Fifth, the mechanical index has arisen from a theory which gives smooth curves of the form shown in Figure 9.8: such curves may in fact show peaks when other effects are incorporated (Roy, 1996).

Lastly, the underlying theory is applied to derive the mechanical index in a way intended to elucidate the conditions which attain the threshold for nucleating inertial cavitation. The amount by which the mechanical index is exceeded is therefore only a guide to the degree of cavitational activity, and by no means an exact predictor. Consider the MHz range illustrated in Figure 9.8, where the range of nuclei size which can seed inertial cavitation is relatively narrow. In such a sound field of fixed frequency and increasing acoustic pressure, as one exceeds the threshold in Figure 9.8, the range of nuclei which may nucleate inertial cavitation increases. In a field containing a broad range of bubble sizes, with a uniform number of bubbles in each size class, the total number of nucleated events would be expected to increase. However if there is only a narrow distribution of bubble sizes, exceeding the threshold by increasing amounts in this manner would, to first order, have little effect on the *number* of inertial events which are nucleated. What might be expected to increase is the energy associated with each collapse. Similarly as, say, the acoustic frequency changes, it is no simple matter to predict how the mechanical index might correlate with the amount of cavitational 'activity' observed.

9.6 The 'activity' of cavitation

Whether a particular type of cavitation is desirable or undesirable in a given circumstance, or whether or not it is amenable to control, if cavitation does occur then the question of degree becomes important.

Consider inertial cavitation in a sound field which may show significant spatial variations. The relevant acoustic conditions, usually acoustic pressure and frequency, may be such that at any point within the sample there is theoretically no possibility of inertial cavitation occurring. To put it another way, at the frequency in question, the magnitude of the peak negative acoustic pressure is everywhere less than the P_{opt} calculated from the limits of the model described in the preceding section (Apfel and Holland, 1991). If however the peak negative acoustic pressure were to be greater or equal to P_{opt} at some point in the field, then inertial cavitation will only occur if suitable nuclei (i.e. having radii within the appropriate range) were present at that location. If such nuclei were not initially present, they may migrate to the point in question, perhaps through Bjerknes forces (Section 9.4) or convection by streaming, etc. (Madanshetty *et al.*, 1991). Alternatively, nuclei of a suitable size might be generated by the ultrasound in the region of interest, through for example, fragmentation of larger bubbles or the presence of suitable suspended agents (Madanshetty, 1995). Such considerations clearly become more important as the frequency becomes greater, because not only does the acoustic pressure threshold for inertial cavitation increase, but the range of allowable radii which a bubble must have to nucleate an inertial event becomes narrower (as illustrated in Figure 9.8 for the low MHz range).

Therefore knowledge of the sound field and the nuclei distribution can enable predictions of whether or not cavitation will occur, to within the accuracy of the model. Such threshold predictions are based on whether a given bubble will undergo inertial cavitation. However what is more difficult to predict is the 'amount of cavitation' that will occur if the threshold is exceeded, since this is clearly not amenable to models of single bubbles. This is partly because in referring to 'the amount of cavitation' what is actually observed is the magnitude of some effect which is brought about through cavitation, inertial or otherwise. The relationship between the 'amount of cavitation' and the ambient pressure is a case in point.

Inertial cavitation, as described in Section 9.5.2, involves two key phases: extensive initial growth, followed by a sudden, rapid collapse. Assuming that a bubble is to undergo inertial cavitation, increasing the static pressure will tend to affect a single bubble's inertial cavitation event in two ways (Walton and Reynolds, 1984): First, it makes it more difficult for the bubble to expand; and second, it increases the compressive forces that drive the bubble to collapse. The first would tend to decrease, and the second to increase, the energetics of the cavitation undergone by a single bubble. Whether the magnitude of the result (erosion, sonoluminescence, etc.) generated by the inertial cavitation of this bubble, and subsequently interpreted in terms of the energetics of the cavitation, increases or decreases depends on which effect has the greater influence. However there is a second factor to consider. The preceding discussion has been limited to the effect on a single bubble, which is in practice rarely the issue. In addition to altering the energy of each individual collapse, changes in the ambient pressure will alter the range of bubble nuclei that may seed inertial cavitation, and so affect the total number of collapses. It does this, not only through physically changing the size distribution of all bubbles present in the liquid, but also by altering the forms of the curves shown in Figure 9.8: in general an increase in static pressure would make the minima in the curves sharper, reducing the range of bubbles in the population that can nucleate inertial cavitation, primarily through inhibiting the growth phase.

Figure 9.9 shows the sonoluminescence measurements of Chendke and Fogler (1983) at $f = 20\,\text{kHz}$ for (a) nitrogen-saturated water, and (b) a saturated solution of water in carbon tetrachloride, containing dissolved nitrogen, for hydraulically increased pressures. The general trend is that small increases in static pressure (up to 7 bar) increase the violence of each collapse, which offsets the reduction in their number: as the pressure is increased further, the latter effect dominates, until the cavitation is suppressed. It has, as an aside, long been known by submariners that submerging the vessel will tend to reduce the noise emitted by flow-induced inertial cavitation at the propellers. However, when the cavitation is strong and the vessel is at high speed, increasing the depth of the vessel will initially cause an increase in the cavitation noise, before suppression occurs (Urick, 1983). This so-called 'anomalous depth effect' is due to the fact that, before the increasing pressure suppresses the growth phase, it first increases the violence of each collapse.

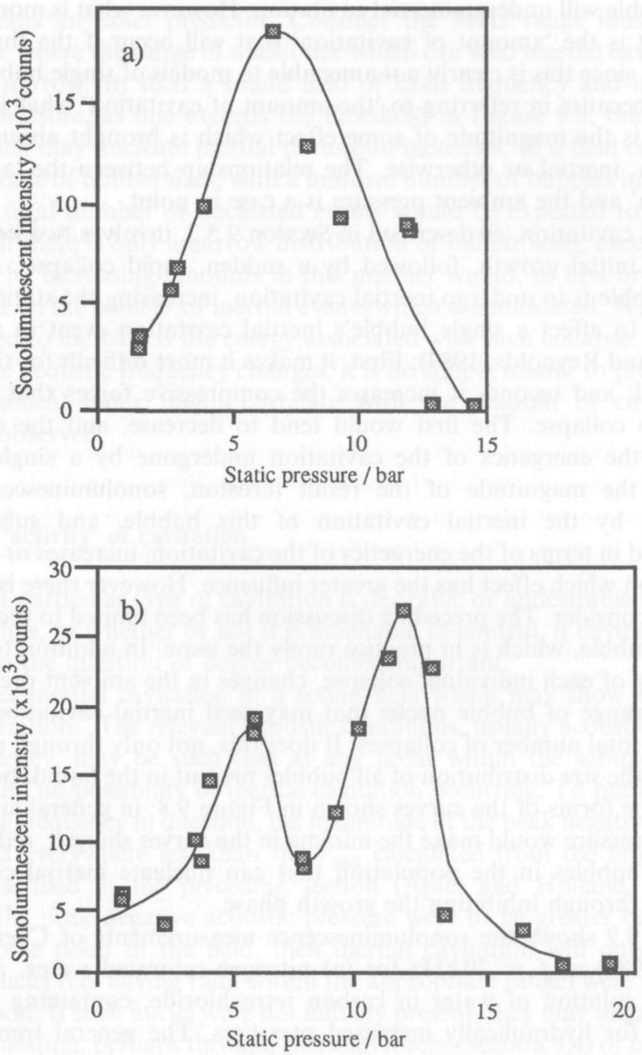

Figure 9.9 Variation of sonoluminescence with static pressure (hydraulically increased) at acoustic frequency of 20 kHz for (a) nitrogen-saturated water, and (b) a saturated solution of water in carbon tetrachloride, containing dissolved nitrogen (after Chendke and Fogler, 1983). Reprinted by permission *The Journal of Physical Chemistry,* 1983, vol. 87, pp. 1362; Copyright 1983 by the American Chemical Society and reprinted by permission of the copyright owner.

In Figure 9.9 the static pressure was increased hydraulically. If the pressure is increased by increasing a gas head, it will tend to increase the concentration of dissolved gas in the liquid. This will affect the cavitation further, as can be seen when the effect of increasing the gas concentration dissolved in the liquid (which can, of course, be achieved without increasing the ambient pressure) is considered in isolation. When the bubble expands during the growth phase of inertial cavitation, gas previously dissolved in the liquid will exsolve into the bubble, in a way similar to the first half-cycle of rectified diffusion in a bubble undergoing non-inertial cavitation. The greater the concentration of dissolved gas, the greater the exsolution for a given degree of expansion. This exsolution decreases the pressure reduction within the bubble which occurs just prior to collapse. The bubble volume change during the inertial collapse is so rapid that little gas re-dissolves, and the presence of a permanent gas component within the bubble will tend to 'cushion' the collapse. Therefore dissolving a gas such as carbon dioxide, which will readily come out of solution during the growth phase, will again cause a reduction in the violence of individual collapses. This is a technique often used to reduce the effects of inertial cavitation. (Clearly changing the gas content of the bubble will have other effects too, changing the acoustic, thermal and chemical properties of the gas content, although such effects will not be considered here.) However, although each individual collapse may be cushioned, the concentration of available nuclei is likely to have increased, increasing the total number of collapses. Similarly, though degassing the liquid will tend to increase the violence of each individual collapse, there will be fewer appropriate bubble nuclei, and therefore probably fewer inertial collapses (Flynn, 1964). The activity of inertial cavitation can also be manipulated through a variety of techniques which remedy the self-suppression effect that cavitation may generate in some modes of operation, as will now be discussed.

There is a range of mechanisms by which the suppression of cavitation is a by-product of the 'normal' method of insonating (e.g. fixed frequency, stationary transducer, etc.). In such circumstances it is therefore possible to enhance the cavitational activity by 'tweaking' the usual conditions. A simple example can be seen from extrapolation of the discussion of the primary Bjerknes forces on bubbles in standing wave fields (Section 9.4), where bubbles of smaller than resonance size migrate to the acoustic pressure antinode; and those of larger than resonance, to the node. The pattern of sonoluminescence from such a standing-wave field is shown in Figure 9.10. Hydrophone measurements in such fields show that the luminescence is clustered at the pressure antinodes, whilst the nodes are dark (Leighton *et al.*, 1988). In an almost identical field, Pickworth *et al.* (1989) demonstrated that the pressure antinodes were the dominant locations for cell lysis. This spatial distribution for cavitational effects is not surprising, since the larger bubbles are less common (Section 9.4), and tend to lie above the upper radius limit in the allowable size range for the nucleation of inertial cavitation (Section 9.5). The existence of regions of high, and of low, activity may be useful, for example with focused ultrasonic therapy (ter Haar *et al.*, 1991). However if the requirement is for uniform treatment by cavitation of a sample, then gradients in the acoustic pressure amplitude

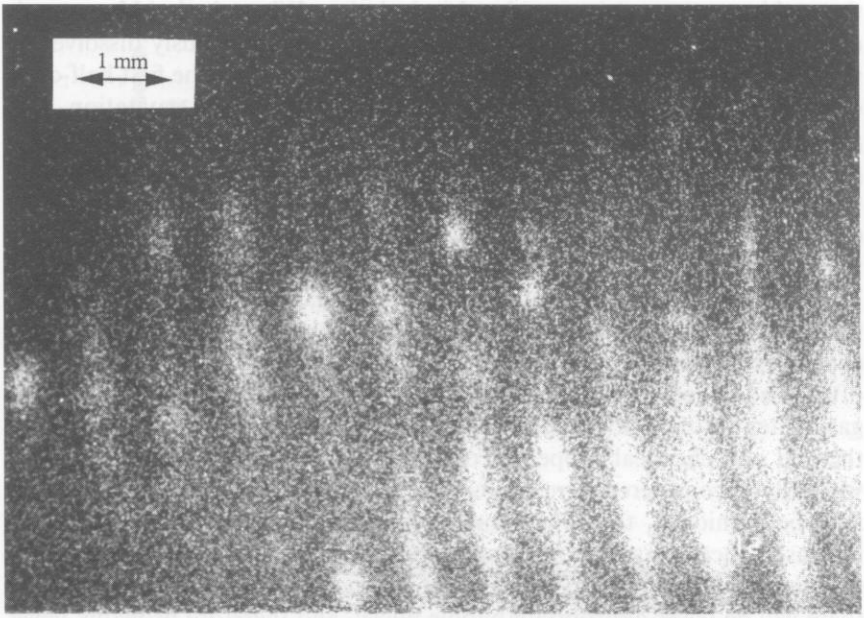

Figure 9.10 Bands of sonoluminescence generated by bubbles aggregating at the pressure antinodes in a standing wave field generated in water from a 1 MHz physiotherapeutic transducer generating a spatial-average acoustic intensity of 3 W/cm^2. The distance between centres of the bands of light is 0.75 mm, i.e. half an acoustic wavelength at 1 MHz in water (Image intensified photograph: T.G. Leighton and M.J.W. Pickworth).

(caused by reflections from the container walls, near-field effects, etc.) can lead to unwanted regions of inactivity (Walton and Reynolds, 1984). This problem can be reduced through a number of techniques which involve 'tweaking' the sound field by, for example, changing the ultrasonic frequency (Leighton, 1995a) or rocking the transducer (Leighton *et al.*, 1988).

Aggregations of bubbles may hinder sample treatment in other ways. If the transducer power is too great intense cavitation close to the transducer can prevent sound from reaching the bulk of the sample (Walton and Reynolds, 1984). The bubble clusters act as acoustic shields. This is the reason for some observations that an increase in power supplied to a continuous-wave transducer can result in a decrease in yield.

Acoustic shielding by bubble aggregations can also occur in continuous-wave standing-wave fields such as those described above (Leighton *et al.*, 1989a). Such aggregations may disperse if the sound is applied in tone-burst pulses. Though delivery of the acoustic energy in such pulses gives a lower time-average acoustic intensity than if a continuous-wave sound field of the same geometry and acoustic pressure amplitude were used, nevertheless there have been numerous reports of such 'pulse enhancement' of cavitation. These, and the mechanisms proposed, are reviewed by Leighton (1994a), as is the related phenomenon of the enhancement of a cavitational effect when the liquid sample is rotated in the ultrasound beam. In addition to shielding, other mechanisms by which sample rotation and acoustic pulsing may cause more thorough treatment of the sample may involve: the rise-time, saturation, and persistence of the 'activity' of the system (Henglein and Gutierrez, 1986; Leighton *et al.*, 1989a); the repeated cycling of a bubble through resonance size as it grows by rectified diffusion and subsequently dissolves when the sound is pulsed (Hill *et al.*, 1969; Clarke and Hill, 1970); the survival of unstabilized nuclei to seed further cavitation events (Ciaravino *et al.*, 1981; Flynn and Church, 1984); excitations associated with the start of insonation (Leighton *et al.*, 1989b); and with the local degassing of liquid (Pickworth *et al.*, 1988; Leighton *et al.*, 1989a). The role of the high-speed translations of the bubble through a sample was introduced in Section 9.2. Rotation of the sample will allow these bubbles to pass, not once, but repeatedly through the sample, enhancing their effectiveness (Miller and Williams, 1989; Miller *et al.*, 1991).

Finally, the relative importance of nucleation and activity to inertial cavitation needs clarifying. If, for example, no free-floating spherical nuclei exist, then one must look to stabilized gas bodies to seed cavitation (Section 9.5.1). If the threshold for the seeding from gas bodies is greater than the mechanical index for the free-floating bubbles generated from such gas bodies, then the key threshold to be exceeded to generate inertial cavitation is that seeding threshold, not the mechanical index. Once seeding has occurred, cavitation will occur; and once cavitation has occurred, it will through bubble fragmentation generate the free-floating nuclei for the next generation of inertial cavitation (providing off-times in the sound field are not too long). Indeed some workers have described the observation of two distinct thresholds: a 'nucleation' threshold, which indicates when cavitation first starts; and an 'activity' threshold, which indicates the onset of enhanced cavitational activity (Calabrese, 1996). For many industrial applications of power ultrasound, it is likely to be the latter, and not the former, which is critical.

9.7 Conclusions

There is a wide range of possible types of cavitation, many of which may occur simultaneously and interact in a power ultrasound field. Even though the effect of primary concern may be brought about through one form of cavitation (e.g. inertial), it is important to appreciate the role of other types since these may critically influence the main event by affecting either the sound field or the bubble size distribution.

When the effects of ultrasonically induced inertial cavitation are important, not only must the energetics of the individual collapses be considered, but also the number of collapses, features which may be interdependent.

Formulations are available to enable approximate calculation of the threshold conditions, particularly acoustic pressure amplitude, nuclei availability, and acoustic frequency. Once cavitation has commenced, the latter two may influence one another through feedback.

The amount of inertial cavitation 'activity' depends on both the energetics of each collapse and on the number of collapses. However it is far easier to monitor the *effects* of inertial cavitation (mechanical, chemical, biological), and these are in most practical situations the issue of prime importance.

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