Acoustic bubble detection – II: The detection of transient cavitation

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This paper follows Part I, which described primarily acoustic techniques for the detection of stable bubbles (gas bodies). Other techniques, based on optical, chemical, and erosive effects, were discussed. This paper describes techniques in the same four categories, though again concentrating primarily on acoustic methods, for the detection of transient cavitation, which is characterised by the sudden growth of a bubble from a small seed nucleus, followed by an energetic collapse and rebound.

In Part I the range of techniques available for the detection of bubbles was subgrouped into three categories: acoustic, optical, and those associated with damage or chemical effects. Optical techniques (and those acoustic techniques involving shadowgraphy or the bubble resonance) were shown to be particularly appropriate for the detection and sizing of stable gas bodies. This paper concentrates primarily on the detection of less stable entities involved in more energetic processes, typified by significant bubble expansion and subsequent collapse. Such cavities might be formed by the sudden dumping of energy into a liquid, for example through the action of lasers, sparks, or ionising radiation. Alternatively small bubbles in high-amplitude pressure perturbations (usually tensions, either acoustic or hydrodynamic in origin) may grow to attain a radius several times the 'equilibrium' one; if, during this process, there is insufficient diffusion of dissolved gas from the liquid into the body, then the permanent gas pressure inside the 'cavity' when it is so expanded will be insufficient to prevent subsequent rapid collapse. This behaviour is termed "transient cavitation" or "unstable cavitation". During collapse, the gas compressed within the bubble may reach temperatures of several thousand degrees [**1-4**], and gas shocks may propagate, reflecting and focusing within the bubble [**4**§4.4.8; **4**§5.2.1b; **5-7**]. Both these processes can in principle generate free radicals and electronically-excited chemical species. Electrical effects may also occur [**4**§5.2.1c; **8,9**]. The collapsing bubble may rebound, emitting a pressure pulse into the liquid. The rebound may leave the bubble intact, or cause fragmentation. Alternatively, if the bubble environment contained directional influences (for example through the presence of another bubble or a boundary, or the passage of a shock wave), the bubble may involute on collapse and form a liquid jet [**10, 11**]. These processes are illustrated in Fig 2 of Part I.

Detection through optical, erosive and chemical effects

Whilst optical studies of 'transient cavitation' are not unknown [**12**], their implementation can sometimes be difficult. Some drawbacks, such as opacity of media or containers, are outlined in Part I. Others are specific to the nature of transient cavitation. An optical image of a size large enough to encompass the fully-expanded bubble will rarely be able to resolve the initial pre-existing gas pocket which seeds this growth. Often the precise location of the event is difficult to determine in advance, depending on the spatial and temporal coincidence of a suitable seed and an appropriate excitation (though focused acoustics and the use of sparks and lasers can influence the siting).

In contrast techniques which exploit chemical and erosive effects of transient cavitation by definition can interrogate all of the sample which contains cavitation sufficient to cause that effect. However continuous real-time monitoring by these techniques can sometimes be difficult, measurements often requiring a cessation of cavitation; and interpretation of such effects in terms of cavitation characteristics may be limited. Such techniques usually involve the detection of effects associated with physical changes (eg erosion) due to jet impact [**13**] and rebound pressures (either from single or cloud cavitation events [**14**]); and resulting from adiabatic heating of gas compressed within the bubbles, or the chemical and biological effects of the radical species which may be associated with both this heating and the propagation of gas shocks. These effects are discussed in chapter 5 of reference [**4**]. The range of effects available can be illustrated through the hotspot resulting from the heating of the bubble gas. This can be detected through its ability to ignite explosives [**15**]. Additionally it can be monitored through effects resulting from the generation within the bubble gas of unstable chemical species, some of which may diffuse into the liquid. These include: (i) sonoluminescence (Fig 1), a faint light emission [**3**] which results from the radiative recombination of free radicals; and (ii) specific sonochemical reactions [**16**], including techniques such as spin trapping which are designed to identify the nature of the short-lived free radicals by allowing them to react with diamagnetic nitroso or nitrone species to produce a longer-lived radical which can be identified through electron spin resonance (ESR) spectroscopy [**17**].

Chemiluminescence may be produced through the radiative reaction of free radical species generated within

Fig 1 A photograph, taken through the image intensifier, of sonoluminescence in a 90% glycerine/10% water mixture, occurring within a cylindrically-symmetric sound field of 2 atm. pressure amplitude (see [4] for details). Each point of light represents the detection of a single photo. The bright sources of light arise from sonoluminescence from bubbles, originating from two regions. Connecting these is a string of bubbles, which may be luminescing or simply reflecting light from the main sources of sonoluminescence. The frame measures 11.75 mm x 17 mm.

the collapsing bubble with a chemical dopant (called luminol, also triaminophthalic hydrazine) which is dissolved into the liquid. Photomultiplication of this has demonstrated that ultrasonic pulses containing essentially one cycle at 1 MHz and a 1:10 duty cycle could generate cavitation with a 20 atm. threshold acoustic pressure amplitude [**18**]. Roy and Fowlkes [**19**] have demonstrated that the sensitivity of the chemiluminescent technique is lower than that of passive acoustic detectors of the type discussed later. Luminol luminescence persists for about 1/20th second after the cavitational processes which generate the excited species has ceased [**20**]. The other disadvantage of the chemiluminescent system is that it necessitates considerable interference with the liquid, making it less suitable for use with, say, biological systems [**4**§5.2.2a(ii)].

Biological effects of cavitation may be related to either the sonochemical [**21**] or physical [**22**] mechanisms. Cavitation can produce a great many effects which have proven to be so complicated in mechanism as to make them as yet unsuitable for use in measurement beyond the most rudimentary indication of the type of cavitation occurring. Examples of these include the bioeffects associated with gas depletion when bubbles grow by rectified diffusion [**23**], microstreaming [**24**] and with the highspeed translation of bubbles under radiation forces [**25**].

As discussed in Part I, whilst sufficiently energetic cavitation may be detected through the chemical effects and damage it will generate, the process of relating such observations to gain information about the bubble field can be difficult, as the mechanisms by which these effects can arise are complicated (particularly, as is often the case, when many bubbles are involved). However the volume changes associated with transient cavitation may be coupled to acoustic fields, and provide a more readily interpretable signal.

Detection through acoustic emissions

Bubble volume changes can cause the emission of acoustic waves [**26,27**]. In Part I some of the emissions associated with the nonlinear oscillations of stable bubbles were discussed. Transient cavitation can also generate acoustic emission. Degeneracy does exist: line spectra and broadband signals, for example, have been detected from both transient and stable cavitation. In certain circumstances some signals prove to be unsuitable for bubble detection since it is not always possible to determine the type of bubble activity responsible for its generation.

In 1980 Neppiras [**1**] summarised the available experimental data as a progression of emissions. If a liquid containing a bubble population is insonated at low power levels, continuous-wave at the fundamental frequency v, the detected acoustic emissions are at v only. At higher intensities, but below the threshold level required to generate transient cavitation, harmonics are emitted at integer multiples of v up to high order. The 2v emission is prominent, its amplitude being proportional to the square of the fundamental. Low-level broadband continuum noise is present, which becomes very strong as the transient cavitation threshold is approached. The v/2 subharmonic appears intermittently, the duration of the emission being much shorter than the 'off-times'. Other subharmonics and ultraharmonics can be detected.

The increases in the broadband continuum, harmonic, ultraharmonic and subharmonic emissions above that encountered in stable cavitation, have the potential for indicating the onset of violent cavitation. The increase in the subharmonic prompted some workers in the past to use it as an indicator of the onset of transient cavitation. However this is not recommended, as it does not provide a unique and unequivocal indicator [**3,4**]. If conditions allow the assumption that only transient cavitation is occurring, then subharmonic generation may occur through the prolonged expansion phase and delayed collapse which can occur during transient cavitation, the bubbles surviving for one, two or three acoustic cycles before collapse [**2**]. Neppiras [**1**] also suggests that a form of periodic unstable oscillation of a bubble driven at twice its resonance near threshold, might emit at subharmonic frequencies. However, as Vaughan and Leeman [**5**] stated in 1986, "the generation of fractional harmonics, in particular the first and third half-harmonic, is a general characteristic of nonlinear bubble pulsation and does not specifically indicate the occurrence of transient cavitation". The excitation of harmonics, subharmonics, and ultraharmonics through the nonlinearity inherent in the pulsation of individual bubbles was described in Part I.

Three other theories for the generation of the subharmonic in stable cavitation were summarised in 1980 by Neppiras [**1**]. The first is parametric amplification by the liquid: if propagation through the medium itself is nonlinear, parametric excitation can occur in the absence of bubbles. The second is that the lower frequency emissions might originate from surface waves on the bubble wall. In 1969 Neppiras [**28**] wrote "Although it has never been found possible to relate subharmonic signals with surface-wave activity on the bubbles, surface waves would be expected to contribute to the v/2 signals – the possibility is not yet ruled out." The third theory suggests that such emission might be the result of the acoustic field acting on a population of bubbles containing, either wholly or in part, bubbles with an equilibrium radius twice the size of the radius which would be the pulsation resonance of the acoustic field [**29**]. Similar mechanisms involving progressively larger bubbles to account for the lower subharmonics were proposed. This mechanism does not explain the subharmonic emissions from single bubbles.

When one has a poorly-characterised sound field acting upon an unknown bubble population, the fact that a signal may arise by more than one mechanism makes it unsuitable as a sizing tool. For a large bubble population in a powerful acoustic field, commonly some bubbles will be undergoing stable, and some transient, cavitation. All the above mechanisms may operate. In addition broad band contributions may arise. Stable bubbles may contribute to this noise through the emission of microbubbles from large-amplitude surface waves [**30**], or to reversion of the oscillations of shocked bubbles out of the steady-state, so that their own natural frequencies appear in the spectrum [**1**]. Such 'noise' would then be dependent on the bubble population, and have structure. Random frequencies, unconnected with v, were thought to be the result of the shock excitation of large bubbles which would then oscillate at their own natural pulsation resonances [**28,31**]. Broad band contributions from transient cavitation are discussed in the next section.

Attempts to correlate acoustic with other indicators of transient cavitation range from investigations on the relationship between acoustic emission and cavitation erosion [**32**], to attempts to monitor the effect of ultrasound on biological materials. Examples of the latter include the exploitation of subharmonic acoustic emissions [**33**], and additionally broad band acoustic signals [**34**], to relate the acoustic emission to the biological effects and damage induced by the ultrasound in cells suspended *in vitro*. Eastwood and Watmough [**35**] measured the sound produced when human blood plasma was made to sonoluminesce. Attempts to correlate the presence of the half-harmonic with the onset of violent cavitation and sonoluminescence are inconclusive [**4**§4.4.7]. Experiments have shown that the appearance of the half-harmonic is not correlated to the onset of sonoluminescent activity [**36-39**].

The most effective methods for acoustic detection of transient cavitation usually employ acoustic signals that can be more readily interpreted in terms of cavitation characteristics. These are discussed in the following section.

The acoustic detection of transient cavitation

The concept of a steady-state resonant response, as discussed in Part I, is inappropriate for the dynamics of transient cavitation, the detection of which following insonation by short, microsecond pulses of ultrasound being of particular importance. This is because of the clinical relevance of such pulses, and the need to assess the likelihood of cavitation-induced effects during exposure. By the mid 1980's calculations suggested that transient cavitation could occur in liquids in response to such ultrasonic pulses [**40-44**]. Free radical production in response to the high temperatures generated within the collapsing bubble had also been indicated for microsecond pulses in experiments [**18, 45-48**]. Of particular importance for clinical applications are techniques which are minimally invasive beyond the effect caused by the primary cavitating beam. These fall into two broad types: those which detect the pressure wave emitted into the liquid on rebound, and those which exploit the enhanced reverberation of the primary beam which the bubbles cause.

Detection through rebound pressure pulses

In 1968 West and Howlett [**49**] set up a 20.25 kHz (continuous-wave) standing-wave condition in a cylindrical transducer filled with degassed tectrachloroethylene. At certain times, related to the phase of the continuous-wave field, they nucleated the medium using a pulsed neutron source. The shock waves emitted by the collapsing bubbles were used to count the number of cavitation events (up to 25 bubbles per second, compared to about one per minute when no neutron source was used).

The short duration of rebound shocks indicates a broad frequency content. Negishi [**50**] found a continuum in the acoustic spectrum occurred when he detected sonoluminescence. Kuttruff [**51**] confirmed these results by examining the circular shock waves produced by cavities undergoing transient collapse using schlieren optics.

Roy et al. [**52**] subjected liquid to a continuous-wave spherically-symmetric stationary acoustic field generated at 61.725 kHz within a spherical resonator. The acoustic pressure amplitude was automatically ramped until cavitation sufficient for the detection of sonoluminescence through photomultiplication just occurred. In addition the operator could listen in on the liquid contained within the spherical cavitation cell via headphones connected to a microphone. Audible 'clicks' or 'pops' were taken to indicate transient cavitation. They found that the pressure threshold for audible sound emission was always less than or equal to that for sonoluminescence. They deduced that "sound and light emission indicate thresholds for two different types of phenomena associated with transient cavitation", and concluded that "if one desires a threshold for 'violent' cavitation, then sonoluminescence is a fitting criterion", and that "light emission may serve as an ideal indicator of what Apfel [**40**] calls the 'threshold for transient-violent cavitation'."

The resonant bubble detector (RBD) described in Part I has been employed in attempts to detect the 'stable' bubble products following shock-induced transient cavitation downstream from the exposure site of a Dornier System lithotripter [**53**]. Extracorporeal shock wave lithotripsy (ESWL) is a technique by which short pulses of high pressure are focused into the body in order to break kidney or gall stones [**54**]. The 1.65 MHz continuous-wave RBD was sensitive to bubbles of radius $R_o = 2 \pm 0.5$ µm at the detection site. Though bubbles were detected *in vitro* in water and blood, and in blood pumped by the heart through a plastic arterio-venous shunt, cavitation was not detected *in vivo* in the canine abdominal aorta. The RBD effectively detects through the stable oscillations it excites in relatively long-lasting bubbles.

Since lithotripter cavitation is characterised by transient cavitation and rapid changes in bubble size, the stable bubbles being the remnants from collapse, a more direct form of detection would employ the energetic emissions associated with rapid changes in bubble size. Coleman et al. [**55, 56**] detected cavitation produced by an electrohydraulic lithotripter using, as a passive remote hydrophone, a focused bowl lead zirconate titanate (PZT) piezoceramic transducer, of 100 mm diameter and 120 mm focal length in water, the focus measuring 5 mm (on axis) by 3 mm wide. This transducer had a 1 MHz resonance: the detection process would therefore correspond to its response to the broad band signal characteristic of the rebound pressure pulse. It is interesting to note that a skilled operator can, by listening to the secondary acoustic emissions during clinical lithotripsy, determine whether or not the shock has hit the stone [**57**].

Passive acoustic detection through reverberation

If an ultrasonic pulse causes transient cavitation, the sudden extensive bubble growth associated with the event may cause enhanced reverberation of the ultrasonic pulse, both through the presence of the expanded bubbles, and through the increase in gas bodies present after the collapse (the net volume of gas in the fragments may be greater than that contained within the seeds owing to the exsolution of previously-dissolved gas during the expansion). Atchley et al. [**58**] used this enhanced scattering to find acoustic pressure thresholds for the cavitation as a function of pulse duration τ_p and pulse repetition frequency ν_{rep} at insonation frequencies of 0.98 and 2.30 MHz, for distilled, degassed, deionized water, filtered to 0.2 μm and seeded with hydrophobic carboxyl latex particles (l μm diameter). Similar experiments [**52, 59, 60**] used short tone bursts of ultrasound to find the threshold acoustic pressure amplitude for transient cavitation in a fluid which was carefully prepared to remove uncontrolled seed nuclei (eg solid particles) before the introduction of a known nuclei population (including Albunex™ spheres, and hydrophobic polystyrene spheres of nominal radius 0.5 μm).

Fig 2 shows the output of the passive detector of Roy et al. [**61**] with and without cavitation. The top trace (a) illustrates the primary pulse from the 757 kHz transducer, followed by a stable low-amplitude background resulting from multiple-path scattering and reverberation in the fluid-filled test chamber. Its stability is testament to the stationary nature of the scattering surfaces. In the lower trace, (b), this scattered background contains a perturbation indicative of a time-varying scatterer, which Roy et al. showed to be at the focal region of the primary transducer. Such signals indicate cavitation. The apparatus used to obtain this result in addition contained an active detection system, and is described in the next section.

Active acoustic detection

A system which is more sensitive to bubbles in the micron size range is the active detector described by Roy et al. [**61**]. Subsequent to their production by the pulse from the first transducer (ν=757 kHz; ν_{rep}= 1 kHz; τ_p =10 μs), the cavitation bubbles then backscatter high frequency pulses (ν=30 MHz; τ_p=10 μs) from a second transducer. Roy et al. [**61**] in fact deployed both active and passive acoustic detectors in their system, which is illustrated in Fig. 3. The cavitation cell and the electronics associated with the transducers are shown in (a), whilst in (b) the fluid management apparatus (a closed-flow system) is illustrated.

From the reservoir, where the fluid is degassed using a vacuum pump, it is then sequentially deionized, cleared of organics, and filtered down to 0.2 μm. The fluid is then passed into the cell, which can be flushed of impurities using flow rates up to 5 litres/minute. Acoustic streaming, generating flows of up to 3 cm/s, could be used to convect nuclei into the focal region. The cavitation cell is substantially similar to those employed in the passive detection studies described above. Separated from the test chamber by a 9 μm thick stainless steel acoustic window is a second chamber containing an identical fluid and an absorber of rho-c™ rubber (so-called as it is impedance matched to water).

This is done to minimise spurious reflections and inhibit the introduction of standing-waves (the steel window prevents contamination of the test liquid by dirt or small particles from the rubber). The 757 kHz transducer, which generates the cavitation and is therefore labelled the 'primary', is mounted on a two-dimensional translating stage to enable its focus to coincide with that of the 30 MHz pulse-echo detector. The 1 MHz unfocussed transducer is cou-

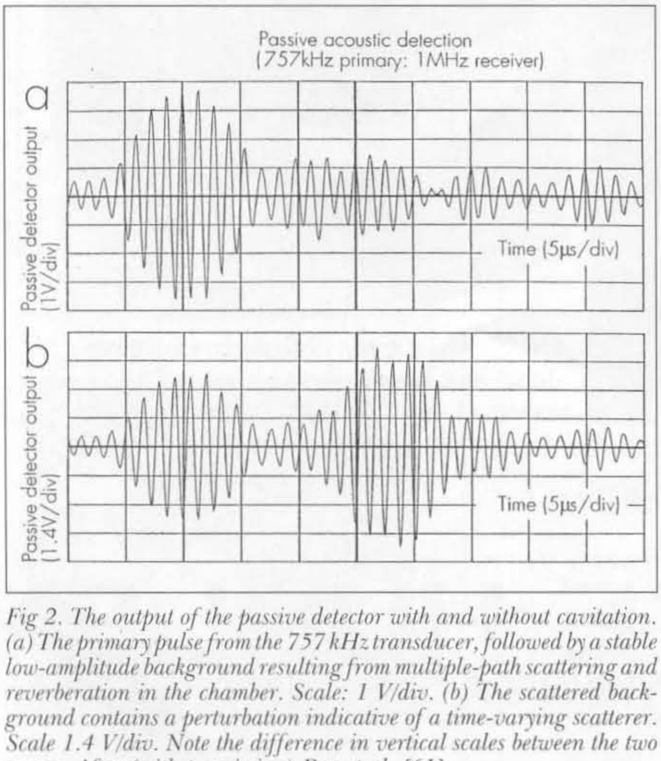

Fig 2. The output of the passive detector with and without cavitation. (a) The primary pulse from the 757 kHz transducer, followed by a stable low-amplitude background resulting from multiple-path scattering and reverberation in the chamber. Scale: 1 V/div. (b) The scattered background contains a perturbation indicative of a time-varying scatterer. Scale 1.4 V/div. Note the difference in vertical scales between the two traces. After (with permission) Roy et al. [61].

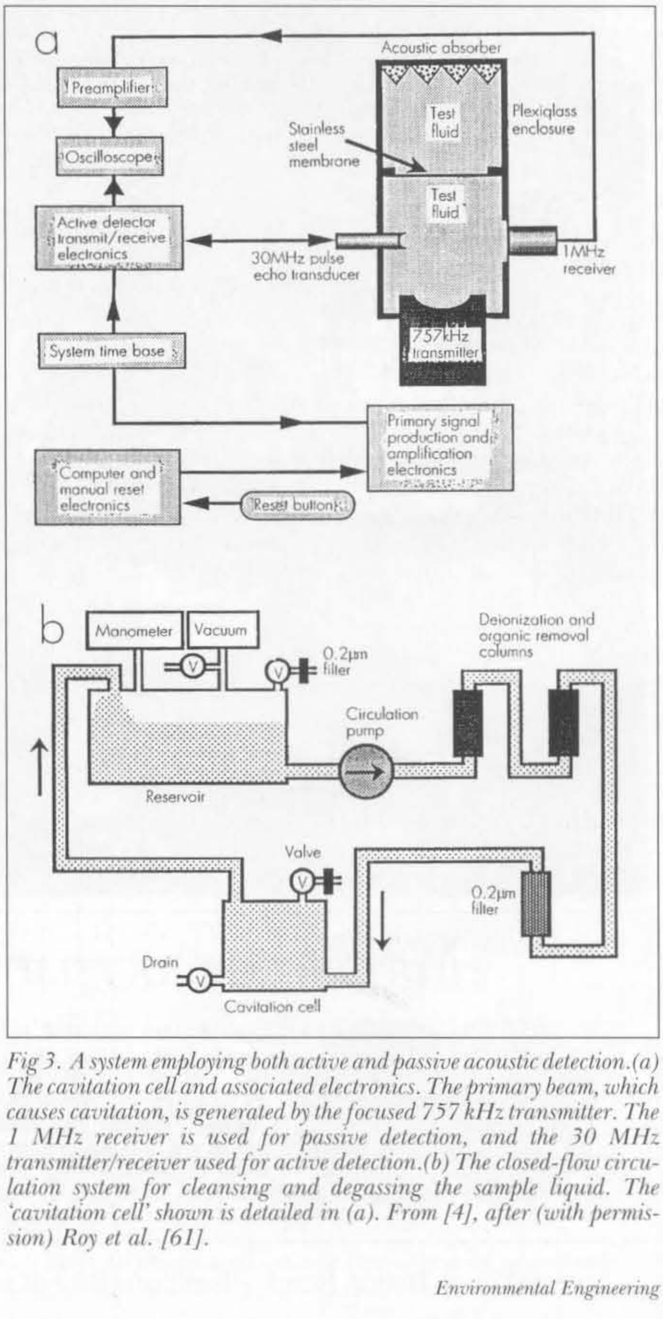

Fig 3. A system employing both active and passive acoustic detection.(a) The cavitation cell and associated electronics. The primary beam, which causes cavitation, is generated by the focused 757 kHz transmitter. The 1 MHz receiver is used for passive detection, and the 30 MHz transmitter/receiver used for active detection.(b) The closed-flow circulation system for cleansing and degassing the sample liquid. The 'cavitation cell' shown is detailed in (a). From [4], after (with permission) Roy et al. [61].

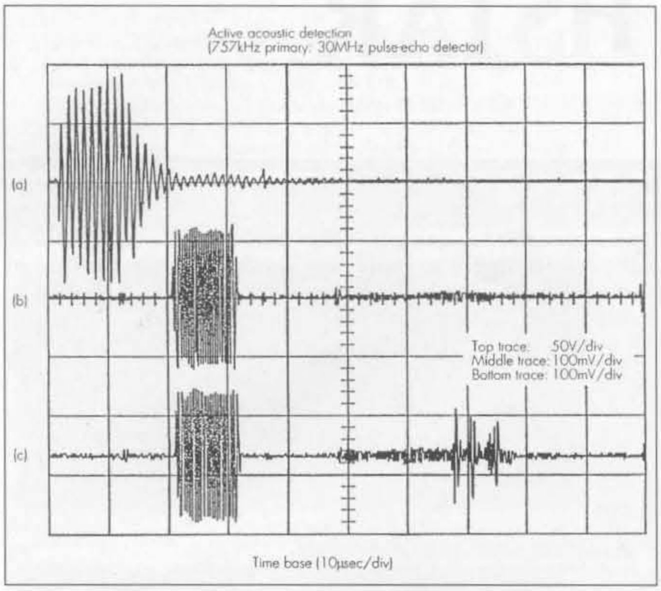

Fig 4. The active acoustic detection of bubbles. (a) The electrical signal which drives the 757 kHz primary transducer (scale: 50 V/div). (b) The signal from the active detector system in the absence of cavitation, the main pulse representing the interrogating 30 MHz signal (scale: 100 mV/div). (c) The signal from the active detector in the presence of the cavitation generated by the 757 kHz transducer (scale: 100 mV/div). The reflected signal from the bubbles is clearly evident in (c). The oscilloscope digitizing rate was 100 Msamples/s. After (with permission) Roy et al. [61].

pled with gel to the cell, opposite the 30 MHz active detector.

Fig. 4 shows (a) the electrical signal which drives the 757 kHz primary transducer, which in turn generates the cavitation. Trace (b) shows the signal from the active detector system in the absence of cavitation, the main pulse representing the interrogating 30 MHz signal. Trace (c) shows the signal from the active detector in the presence of the cavitation generated by the 757 kHz transducer. The reflected signal from the bubbles is clearly evident.

Holland and Apfel [**59**] comment that, when the thresholds measured by the active and passive systems for very similar fluid systems (eg having the same polystyrene sphere and gas concentrations) were compared, they were found to be very close, suggesting that the acoustic pressure threshold to cause one bubble to go transient in this system is the same as that required to make a cloud of bubbles go transient.

Holland et al. [**62**] used the active detection system alone to investigate *in vitro* transient cavitation from short-pulse diagnostic ultrasound, both imaging M-mode and Doppler, under conditions comparable to the clinical situation, with ν=2.5 or 5 MHz. Cavitation was detected in water seeded with 0.125 μm mean radius hydrophobic polystyrene spheres at 2.5 MHz, with a threshold peak negative pressure of 1.1 MPa, in both M-mode and Doppler insonations. No cavitation was detected at 5 MHz even at peak negative pressures as high as 1.1 MPa. No cavitation was detected in water seeded with Albunex™ at either frequency.

Madanshetty et al. [**63**] discuss the effect of the sensor system on the cavitation threshold for water containing a microparticle suspension. With the active system turned off the passive system detected a cavitation threshold at around 15 bar; with the active system on the latter detected a threshold at only 7 bar. This implies that either the active system is more sensitive, or that it encourages cavitation itself, or both. The fact that the passive system also detects a reduction in threshold (to around 8 bar) suggests that both might be true, and that the cavitational effect of the active system might be considerable. To investigate this Madanshetty et al. [**63**] reduced the peak negative pressure from the active transducer to only 0.5 bar, so that its influence on transient cavitation should be negligible, and found that the active detector alone at this intensity could not generate cavitation events. They found that the active detector could effect cavitation through the streaming it develops (which is promoted by its high frequency and focused nature). This flow convects nuclei into the cavitation zone. A second mechanism by which the active detector can affect cavitation is through the accelerations it imparts to particles. The combination of these accelerations with the density contrast generates kinetic buoyancy forces. These cause tiny gas pockets on the surface of solid particles to aggregate into gas patches of a size suitable to act as nuclei for cavitation. Another possibility suggested by Madanshetty et al. [**63**] is that potential nuclei, which would normally be driven by the 0.75 MHz transducer, would be detained in the region where the fields are strongest by cross-streaming with the flow generated by the active transducer, so enhancing the likelihood of a cavitation event at lower insonating pressures. Citing simulation results of Church [**64**], Madanshetty et al. [**63**] rule out the possibility of the active detector influencing cavitation through rectified diffusion.

In comparison with the passive detector, the active detector seems to be more sensitive, may affect cavitation itself, and is sensitive predominantly in its focal region, so may more readily be used to give a degree of spatial information. On the other hand, the passive detector is sensitive to a larger region of space, and remains continually alert for cavitation: for the active detector to operate its interrogating pulse must arrive at its focus at the instant when the transient bubbles are present. There are implications regarding the strain placed on the user of these systems inherent in the nature and display of the detected signals [**63**].

Conclusions

Transient cavitation can produce a number of effects which may be used to detect and, to some extent, quantify it. Chemical, biological, and erosive effects may be difficult to interpret in order to obtain information about the cavitational field and bubble dynamics. Acoustic emissions, based on the rebound shock, and the excess scattering by the cavitation bubbles either of the field that induced the growth, or of a second field applied specifically for detection purposes, may be particularly useful in detecting the transient growth and collapse phases. However it should be noted that transient cavitation often is seeded by a pre-existing nucleus, which may itself be stable [**4**§2.1; **65, 66**]. After the event there will be gas body remnants, which again might be stabilised or not. Fragmentation will produce daughters, increasing the number of such bodies. Therefore the detection of stable bubbles, as discussed in Part I, may be employed to characterise the origins and products of transient cavitation.

Acknowledgements

The author wishes to thank NERC (SIDAL 00670) and SERC for funding. This article contains material from The Acoustic Bubble (T G Leighton), Academic Press, London, 1994.

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