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THE PULSE ENHANCEMENT OF UNSTABLE CAVITATION BY MECHANISMS OF BUBBLE MIGRATION

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When ultrasound passes through a liquid, bubbles may be produced. These can subsequently collapse, adiabatically heating the gas contained to several thousand kelvin, creating free radicals. The radicals may undergo radiative recombination, giving rise to sonoluminescence. Therefore the presence of sonoluminescence may indicate potential biohazard.

Experiments have shown that, when ultrasound is pulsed, the amount of sonoluminescence from unstable cavitation may increase, contrary to the expected result.Two theories for this are proposed, tested, and found to be satisfactory. Both rely on the migration of bubbles away from bubble aggregates during the off-time of the insonation. Such migrations would: a) reduce local degassing of the liquid at these aggregates, so promoting bubble growth by rectified diffusion there; b) remove regions which have an acoustic impedance very different to that of the pure liquid (and would thus otherwise attenuate the passage of sound to the bubbles). Experiments with agar gels of varying viscosity suggest that both mechanisms work together.

The driving force for these migrations could arise through (i) buoyancy, (ii) acoustic streaming forces, or (iii) a coupling of the acoustic pressure field with the bubble oscillation at the end of each sound pulse. The particular driving force dominating in a given regimen depends on the size of the active bubbles, and so is frequency dependent: for therapeutic ultrasound (operating at 1MHz), process (iii) is responsible for the migrations.

INTRODUCTION

The enhancement of cavitational effects that has been observed in some acoustic pulsing regimens has never been explained satisfactorily. The existence of a transient excitation mechanism has been established [1], and the effect of this when the sound is pulsed has been demonstrated at 10 kHz. In this paper, the sonoluminescence from pulsed clinical ultrasound is discussed, to see whether pulse enhancement occurs; and if it does, whether it is due to transient excitation alone or coupled with other, possibly dominant, mechanisms.

EXPERIMENTAL RESULTS

The experiment is described in Pickworth et al. [2]. Sonoluminescence is detected from a 1MHz therapeutic ultrasound field by photomultiplication. The results are in the form of a population distribution for the photomultiplier output pulses, shown as a function of the energy of those pulses (measured in arbitrary units). An example of such a graph is shown in figure 1a, where the sonoluminescence from aerated water at 22°C is shown for the continuous-wave insonation, and for the duty cycles 1:2, 1:4 and 1:7 (the duty cycle is the ratio of the on-time to the off-time). Since the Therasonic 1030 has a fixed pulse length of 2 ms, the duty cycles of 1:2, 1:4 and 1:7 therefore represent off-times of 4ms, 8ms and 14ms respectively. This is illustrated in figure 2.

The background count is negligible on this logarithmic scale. The fall-off in each curve for pulse heights of less than 1 unit is due to saturation of the counting system. Calculations show that such effects are negligible for pulse heights greater than this. Note that the count scale is logarithmic, so that differences between curves are in fact considerable, and were entirely reproducible.

DISCUSSION

1 Theories for pulse enhancement

Without pulse enhancement, one would expect the magnitude of the sonoluminescence to increase in ascending order for 1:7, 1:4, 1:2 and continuous-wave, and the count rate ratios to be respectively 1/8 : 1/5 : 1/3 : 1 (ie. in the ratios of the total insonation times). This can be seen from figure 2. If the light output were to depend only on the total insonation time, then the magnitude of the sonoluminescence expected in each case would be in the ratios of the shaded areas in figure 2 (taken over a sufficiently long time). These expected results are seen for the smaller pulse heights, but not for the larger pulses when the light output in increasing order is for continuous-wave and then for the duty cycles 1:2, 1:4 and finally 1:7 .

If transient excitation were the sole cause of this pulse enhancement, one would expect the count rate to increase with the number of starts of pulses contained within a given interval. From figure 2 it can readily be seen that this would lead to the magnitude of the luminescence occurring in increasing order for continuous-wave, 1:7, 1:4 and then 1:2.

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Figure 1. The luminescence count rate as a function of the pulse height.

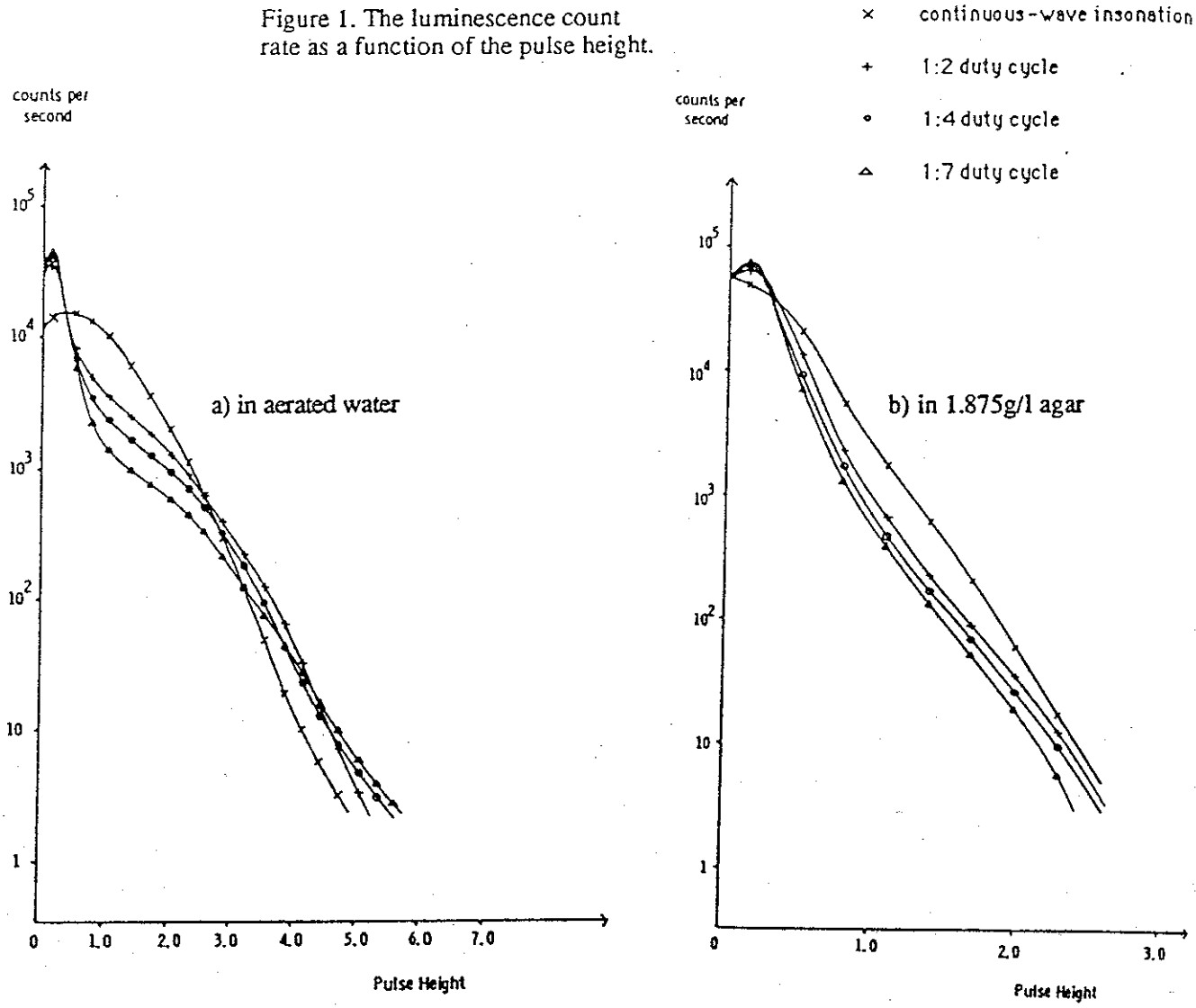
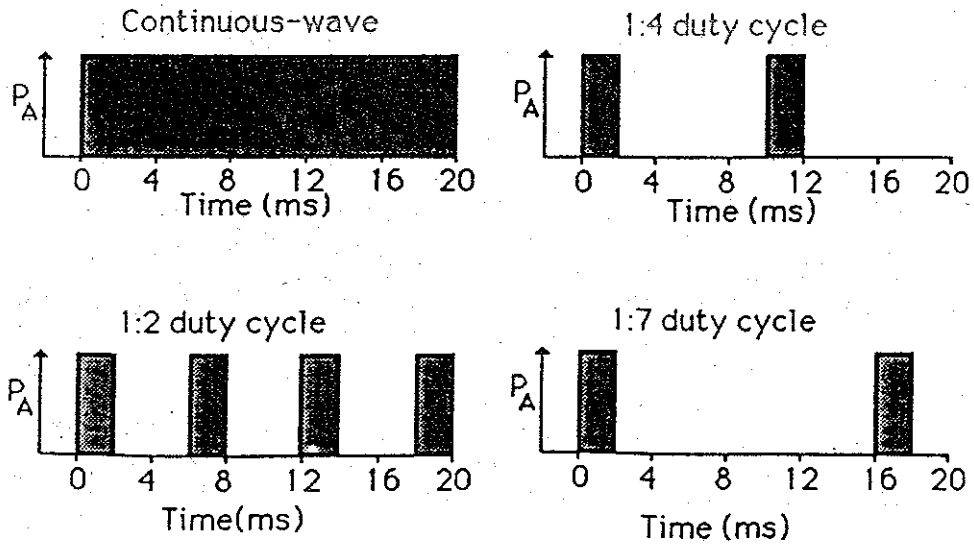


Figure 2. A schematic representation of the pulsing regimes of the Therasonic 1030.



This is indeed the case for pulse heights between 3.2 and 4.2 units. The fact that this is not the order for the larger pulses suggests that, while transient excitation is a potent mechanism in some acoustic regimens [1], its effects in this 1 MHz system are observable only in a limited range of pulse heights. Outside this region, other factors must dominate.

To explain the form of figure 1a beyond this range, two alternative mechanisms are postulated. Both rely on the reasonable assumption that the brightest light flashes are the product of the most intense cavitational collapses. They are referred to as the 'degassing' and 'impedance' mechanisms.

1.1 The degassing mechanism. The first mechanism (subsequently referred to as the 'degassing' theory) refers to the cyclic process of a bubble growing by rectified diffusion from a nucleus to resonant size (or to the transient cavitation threshold, if this occurs first), where it collapses unstably to produce bubble fragments. These fragments then grow to resonant size, to themselves collapse, and so on. Within each oscillatory cycle, there is a finite chance* of the bubble undergoing unstable collapse, a chance which increases as the bubble approaches resonant size. Therefore, the more cycles it takes for a bubble to grow to resonant size by rectified diffusion, the more likely it will collapse before reaching resonant size. Such a collapse would produce a lower-energy light pulse.

During a prolonged period of insonation, bubbles of less than resonant size, growing by rectified diffusion, cluster at the pressure antinodes under the influence of Bjerknes forces [3]. As they grow, they will deplete the water in the region of dissolved gases. This can be seen from the fact that the diffusion length of air in water, after a time t, is given by $\sqrt{(4Dt)}$, where D is the diffusivity of dissolved air in water, and equal to about 10^{-5} cm^2s^{-1} [4]. Thus in 2 ms (the length of each sound pulse) the gas diffusion length in water is 2.8 μm, which is very much less than the node-antinode spacing in a 1 MHz field (≈ 0.75 mm). Therefore during insonation the pressure antinodes will become partially degassed. This will make each bubble oscillation less efficient in drawing dissolved gas into itself from the liquid and so it will take more oscillations to grow to resonant size than would a bubble in water that remined fully aerated (figure 3a). Therefore, on average, this type of growth would result in more of the lower-energy light pulses.

If, when the sound was pulsed, the bubbles were to migrate during the off-time in such a way as to break up these antinodal clusters, and to disturb the liquid distribution at the antinodes, then at the start of the next pulse when the Bjerknes forces drove them back to the antinode, this region would be flushed with fresh aerated water. Rectified diffusion would consequently be more efficient, and the bubbles would be more likely to reach resonant size before collapsing unstably. This would result in more higher-energy flashes of sonoluminescence.

Thus the form of figure 1 would be explained: continuous-wave insonation causes bubble clustering which leads to antinodal degassing, resulting in more low-energy sonoluminescent pulses. On the other hand, when the sound is pulsed, degassing does not occur to so great an extent, bubble growth is thus more rapid, and so more of the higher-energy sonoluminescent pulses are detected. The longer the off-time, greater the effect.

This theory is substantiated by observations by Blake [6] who describes a phenomenon called 'ultrasonic degassing'. Here liquid in the locality of cavitating bubbles becomes degassed. Blake noticed that once strong cavitation was obtained in water which was continually stirred, the intensity of the cavitation would show a marked decrease a few seconds after the stirrer was switched off.

1.2 The impedance theory. The second theory (subsequently termed the 'impedance' theory) is schematically illustrated in figure 3b. During insonation, the bubbles of less than resonant size (ie. the source of sonoluminescence) cluster at the pressure antinodes. An acoustic impedance mismatch can occur at the interface between regions of high and sparse bubble concentrations (ie at the outer layers of the antinodal clusters), hindering the transfer of sound from one region to the next. Sound from the body of the liquid is attenuated before it reaches the bubbles at the heart of the cluster. The reduced acoustic pressure amplitudes will result in less cavitation, and so less sonoluminescence (particularly of the most intense type). However if bubbles migrate during the off-times when the sound is pulsed, these clusters will break up. At the start of the next pulse, the sound will reach all the bubbles at full amplitude, and the most intense sonoluminescence pulses will be detected (particularly when the effect of transient excitation is considered). Thus, the form of figure 1 is produced: antinodal aggregates form during continuous-wave insonation, and the bubbles in the interior are shielded by an impedance mismatch, and so fewer high-energy collapses occur; when the sound is pulsed, these aggregates are broken up, and more of the high-energy sonoluminescent pulses are recorded. The longer the off-time, the greater the effect.

* This *chance* arises from the fact that random features in the inhomogeneous acoustic field around a bubble can cause unstable collapse. A growing bubble can experience such violent surface oscillations that acoustic shear forces tear microbubbles from its surface; this may happen so rapidly that the bubble explodes in an unstable collapse [5]. Obviously, the larger the bubble, the weaker the surface tension which maintains sphericity, and so the more pronounced the surface oscillations.

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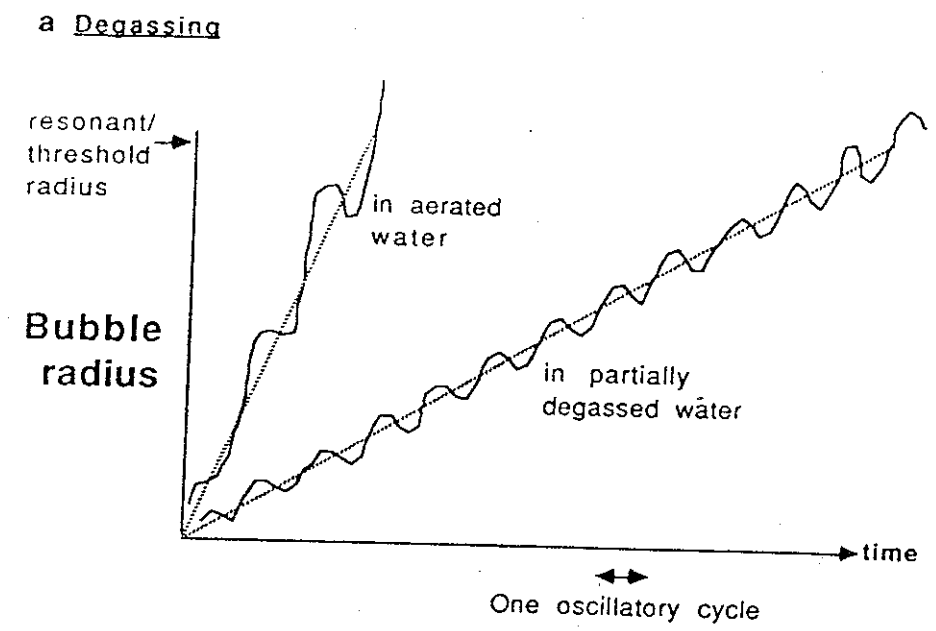
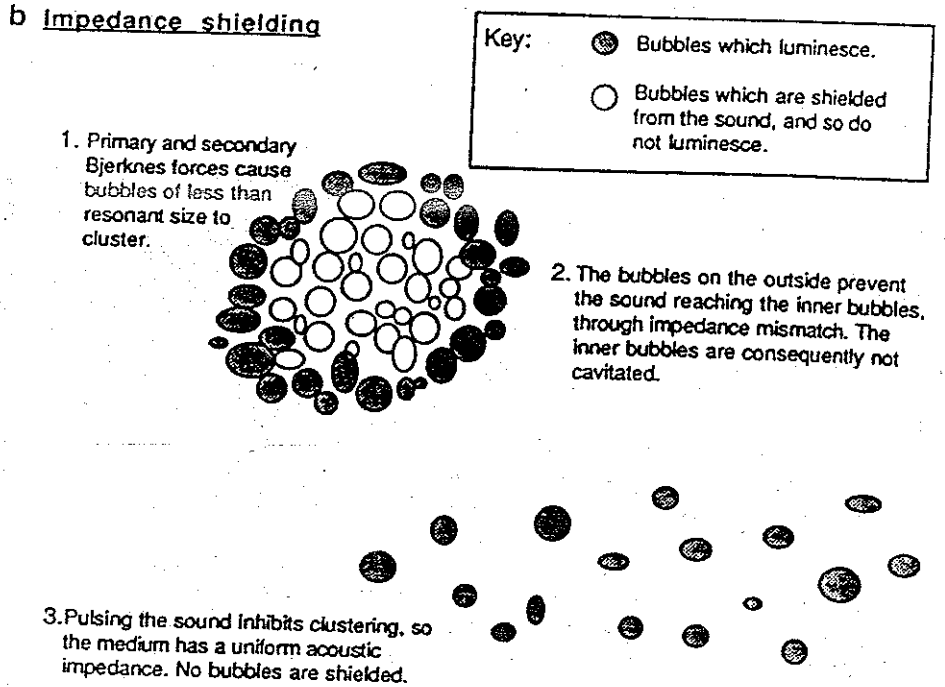
Figure 3. Diagrammatic representations of the two migration theories. a) The degassing theory: bubbles growing by rectified diffusion take fewer oscillations to reach resonance size (or the transient cavitation threshold) in aerated water than they do in partially degassed water. Therefore bubbles tend to collapse at resonance (or threshold) size in aerated water, whereas in partially deagassed water they collapse before reaching this size. Therefore there is more luminescence at the higher pulse heights in aerated water, and moreat the lower pulse heights in partially degassed water. b) The impedance theory.





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Both theories rely on the flushing of bubbles from the antinodal regions during the off-times. The further the migration, the more the enhancement, so both theories work to increase the luminescence as the off-time increases. If the experiment were to be performed in a more viscous medium, the migrations would be greatly reduced. One might expect the cross-over seen in figure 1 to disappear, and the count rates to be increase in the order of duty cycle 1:7, 1:4 and 1:2, with the most luminescence from continuous-wave insonation, the count rate now being simply a function of the total time of insonation. (Previous experiments suggested that transient excitation is inhibited by the greater viscosity of the agar.) Therefore to test these two theories, the pulse-energy distribution spectrum was obtained in exactly the same way as for water, but this time in 1.875g/l agar at 22°C. This substance had a viscosity of 170 ± 10 cp., and so was still fluid; since viscosity is a second-order effect when describing the actual bubble oscillations [7], then the actual cavitational behaviour will be very similar to that in water. What will be different is the scale of the migrations, which will be greatly reduced in agar (see section 2). The results are shown in figure 1b; the cross-over has disappeared, the count rates increasing in the order of duty cycle of 1:7, 1:4 and 1:2, with continuous-wave insonation the most luminescence. Therefore the hypothesis that cross-over is associated with bubble migrations is strengthened; in addition, if the spectrum is taken from a series of agar solutions starting with pure water and gradually increasing the agar concentration, the cross-over first disappears, then reappears, then finally disappears again. This suggests that the process is complex, probably involving more than one mechanism, ie. both the 'degassing' and 'impedance' mechanisms may be involved.

However, for either theory to be considered seriously, there must be a reason for the bubbles to migrate from the antinodal clusters. Some possibilities are discussed in the next section.

2 Mechanisms for bubble migration

2.1 Passive processes. Buoyancy forces can cause bubble migrations. When insonation occurs, bubbles are held in place by acoustic forces. Once the sound is switched off, the bubble can be moved by buoyant forces. If R_o is the equilibrium bubble radius (assumed constant since dissolution times are slower than the off-times employed here [8]), then the inertial mass m of the bubble is $2\pi\rho(R_o)^3/3$ (ie. half the mass of the displaced liquid [9]), where ρ is the liquid density. If x is a measure of linear distance, and η is the liquid viscosity, the buoyant drag forces on the bubble cause a net acceleration:

$$4\pi\rho g(R_o)^3/3 - 6\pi\eta R_o(dx/dt) = m(d^2x/dt^2) \quad . \quad 1$$

The solution to this is

$$(dx/dt) = v_t(1-\exp(-At)) \quad , \quad 2$$

where v_t is the terminal velocity, which is found by substituting this solution for x into equation 1 and setting $t=\infty$ to give

$$4\pi\rho(R_o)^3/3 = 6\pi\eta R_o v_t \quad , \quad 3$$

and hence

$$A = 6\pi\eta R_o/m = 9\eta/(\rho R_o^2) \quad . \quad 4$$

Integration of equation 2 with respect to time shows that the distance x travelled by a bubble in time t after the end of insonation is

$$x = v_t(t + (\exp(-At) - 1)/A) \quad . \quad 5$$

At 1 MHz, 3 μm is the resonant bubble radius. Visual inspection has shown that nodal bubbles have a typical radius of 0.3 mm, though there is much variation. Bearing in mind that at 1 MHz the node-antinode spacing is 0.375 mm, the migration distances obtained from equation 5 for such bubbles show that with the off-times used in this paper (ie. 14 ms maximum), only the buoyant migrations of bubbles of larger than resonant size have the potential to break up the antinodal clusters in water. In 1.875g/l agar, no significant migration occurs.

2.2 Active processes. Two such processes could occur. In both, the bubbles attain a certain velocity during some part of the acoustic pulse. Once the pulse ends, they continue moving, but with their velocity continually being decreased by liquid drag. Therefore before either process is dealt with in detail, it is necessary to formulate this deceleration of a bubble by viscous drag.

2.2a) Deceleration by viscous drag. If a bubble travels under no driving force through a liquid at speed v, drag forces decelerate it:

$$6\pi\eta R_o v = - m(dv/dt) \quad . \quad 6$$

Solving this equation shows that at a time t after the driving force (here, due to the sound) has ceased, the bubble is

moving with a speed given by

$$v = v_0 \exp(-At) \qquad 7$$

where A is as given in equation 4, and where v_0 is the bubble speed at t=0. Substituting v=dx/dt into equation 7 and integrating with respect to time gives x, the distance moved in time t to be

$$x = v_0(1 - \exp(-At))/A \qquad 8$$

This formulation may now be applied to the two acoustically-driven processes.

2.2b) The migration of bubbles from streamers. In a standing wave system, the Bjerknes forces drive bubbles of smaller than resonant size to the antinodes. These bubbles follow specific routes and from their consequent appearance are called streamers (figure 4). At the moment the sound ceases, a streamer bubble will have a certain speed v_0, which can be substituted into equation 8 to give the distance migrated by that bubble at times t after this point. To find the value of v_0, the magnitude of the Bjerknes force [10] can be equated to the viscous drag force to give the speed of the bubble v_0 if it is at a position y in the sound field when insonation ceases (assuming that the bubble speed responds rapidly enough to changes in the position of the bubble in the field):

$$[3 P_A k \xi_0 \sin(2ky)/2] \cdot [4\pi R_0^2/3] = 6\pi\eta R_0 v_0 \qquad 9$$

Here P_A is the acoustic pressure amplitude, k the acoustic wave number and ξ_0 the amplitude of the radial oscillations at the pressure antinode.

Having calculated v_0 from equation 9, the distance subsequently migrated by the bubble can be found from equation 8 and plotted as a function of its position in the sound field at the end of the acoustic pulse. This is done in figure 5 for air bubbles of resonant size in water and in 1.875g/l agar.

A bubble of radius 3μm travels no more than about 2μm in water. Therefore the migration of bubbles from streamers cannot cause pulse enhancement of sonoluminescence at 1 MHz. The migrations in agar are, of course, insignificant.

2.2c) Bubble migrations caused by acoustic impulses. Bjerknes forces arise from the time-average of the $-V\nabla P$ acoustic pressure forces. The instantaneous value of $-V\nabla P$ can be very much larger than its time average, because for any given value of y it will be positive for one half of each acoustic cycle and negative for the other half. Thus, when the pulse ends, the force acting on a bubble can be very variable, depending on the position of the bubble, the way in which the transducer ceases vibrating, and on the damping of the system. Whatever its value, an impulse will be delivered to the bubble whilst the system relaxes to zero.

An estimate of the magnitude of the impulse can be found by calculating instantaneous (rather than time-averaged) values of $-V\nabla P$, where $V=V_0(1-(3/R_0)(\xi_0\sin(ky)\cos(wt+\pi))$ and $\nabla P=2kP_A\cos(ky)\cos(wt)$ [10]. Here V_0 is the equilibrium bubble volume, and w the acoustic frequency. Then by assuming that this force acts for about one quarter of the sound cycle, the impulse given to the bubble can be found. Dividing this by the inertial mass m of the bubble (defined above) gives the velocity change of the bubble at the end of the pulse. If we assume the bubble was at rest during insonation (ie. it was either a bubble of smaller than resonant size near the pressure antinode, or of larger than resonant size near the node), then we have found the velocity v_0 of the bubble at the start of the off-time. [It should be noted that if the bubble is not at rest (for example, if it was in a streamer, as is often likely), then the initial velocity v_0, and consequently the distance migrated by the bubble, may be much greater than this]. Once an estimate of v_0 has been obtained, then the distance migrated in time t can be found as a function of the initial position of the bubble, by use of equation 8.

Such estimates have been made, and the results are shown in figure 6. Since there are so many variables involved, the estimate has been made using maximum typical values of $-V\nabla P$. This overestimate is, however, roughly compensated by the underestimate implicit in assuming that the bubble is initially at rest.

From figure 6a it can be seen that a bubble of radius 3 μm, in a 1 MHz sound field, could travel up to 8 μm in water so would be unlikely to cause pulse-enhancement. However, in the same sound field, there will be bubbles of greater than resonant size at the pressure nodes. These bubbles are clearly visible when the Therasonic 1030 is used, and their radii are estimated at being between 0.1 and 0.5 mm . The migrations of bubbles of 0.3 mm radius are shown in figure 6b. In a 1MHz sound field, such bubbles could migrate up to 8 cm in water under the acoustic impulse. This is equal to over 60 acoustic wavelengths. It should be stressed that, although the calculations used to plot this figure were exact, there were so many variables (eg. amplitude of the bubble oscillation), that large variations from this estimate can be expected for each individual bubble. However, it gives an idea of the magnitude of the migration, and since we are dealing with large numbers of bubbles, some will probably migrate such distances at the end of each pulse. If only some of these nodal bubbles migrated in this way when the sound was pulsed, the antinodal cluster

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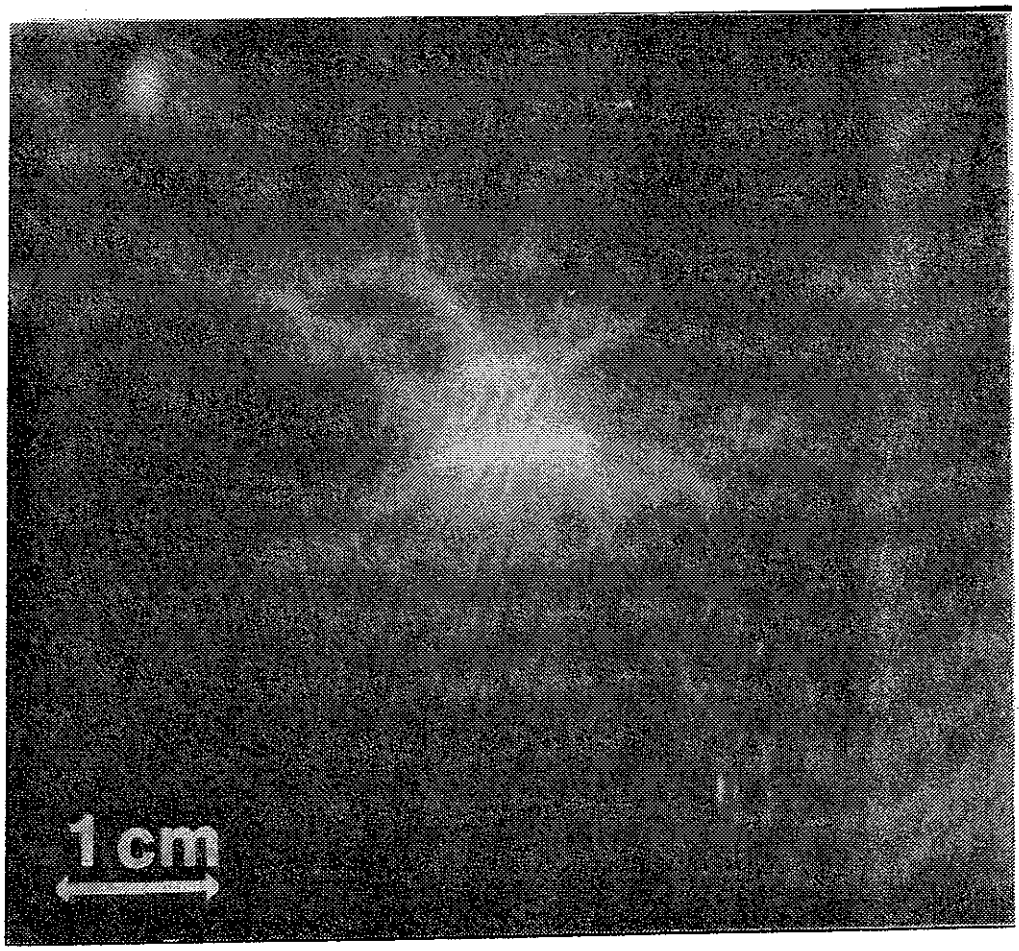
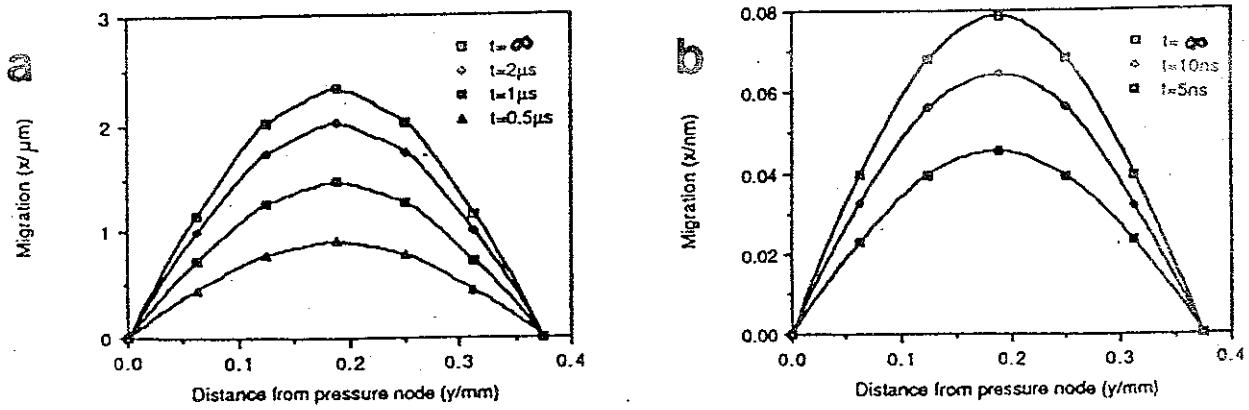


Figure 4. An example of 'streamers', seen here during cavitation in a 10kHz cylindrical, sound field (viewed from above). Small bubbles flow in the streamers to the focal pressure antinode at the centre of the photograph.

Figure 5. The distance migrated by a streamer bubble as a function of its position in the sound field at the end of the 1MHz acoustic pulse, plotted at various times after the sound has ceased in a) water and b) 1.875g/l agar.



would be disturbed sufficiently to enhance sonoluminescence. The maximum expected distances for migration in 1.875g/l agar solution are shown in figures 6c and 6d. It can be seen that neither antinodal nor nodal bubbles will migrate sufficiently in agar to enhance sonoluminescence.

3 Use of viscous drag calculations to verify that at 1MHz, pulse enhancement is due to the migration of nodal bubbles

The evidence suggests that pulse enhancement at 1 MHz is dominated by the 'degassing' and 'impedance' theories operating through a mechanism whereby the larger-than-resonance nodal bubbles migrate during the off-time after receiving a - $V\nabla P$ acoustic impulse, and in doing so flush smaller-than-resonance bubbles away from the antinodes. By examining the drag time-scales predicted in section 2.2a it can be verified that the migration of nodal bubbles is the primary cause of the pulse enhancement. Figure 1a shows that enhancement of the brightest pulses increases as the off-time increases, the maximum being for a 1:7 duty cycle (off-time of 14 ms). The salient results of section 2.2a can be summarised in the following table:

A bubble of radius 300 μm in water decelerates so that:

In 4ms it has migrated 33% of the total distance. This is equivalent to the off-time at the 1:2 duty cycle.
In 8ms it has migrated 55% of the total distance. This is equivalent to the off-time at the 1:4 duty cycle.
In 14ms it has migrated 75% of the total distance. This is equivalent to the off-time at the 1:7 duty cycle.

In comparison, a bubble of radius 3μm travels 90% of the total distance in 2 μs.

If the enhancement were due to the migration of antinodal bubbles (of radius ≤ 3 μm), then we would expect that pulsing the sound gave more sonoluminescence that continuous wave, all three duty cycles employed would give identical light outputs per pulse. This because after 2 μs no significant increase in migration occurs. The amount of luminescence would simply go in order of the total insonation time (ie. continuoos-wave>1:2>1:4>1:7). The fact that increasing the off-time does enhance the luminescence verifies that it is the migration of *nodal* bubbles which enhances the sonoluminescence.
The deductions leading to this conclusion are summarised in figure 7, which refers back to figure 1.

CONCLUSIONS

During continuous-wave insonation, bubbles of less than resonance size form aggregates at the pressure antinodes which decreases the sonoluminescence by the mechansisms outlined in the "degassing" and "impedance" theories. In a 1 MHz sound field, the migration of *nodal* bubbles (whose radii are greater than resonance) could in turn break up the *antinodal* bubble aggregates as they pass thgrough them, and so cause pulse enhancement (as measured in figure 1a). The migratory force is an impulsive one, proposed in section 2.2c .
Buoyancy may also contribute through the migration of nodal bubbles; however, these migrations are about one hundred times smaller than those due to impulse, so their effectiveness will likely be correspondingly less. These buoyant migrations will effect all the nodal bubbles; in all likelihood, the impulsive forces will not do so. Therefore the buoyant migrations will contribute, but probably not be dominant.
Transient excitation seems to influence the luminescence, though to a lesser extent. Its effects are seen only at the middle pulse-heights, where the cross-over means that the effects due to the two bubble migration mechanisms are hidden.

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Figure 6. The distance migrated by a bubble under the impulse. It is plotted as a function of the position of the bubble in the field at the end of the 1MHz acoustic pulse, and is shown at various times after the sound has ceased. a) Bubble radius 3μm, in water. b) Bubble radius 300μm, in water. c) Bubble radius 3μm, in 1.875g/l agar. d) Bubble radius 300μm, in 1.875g/l agar.

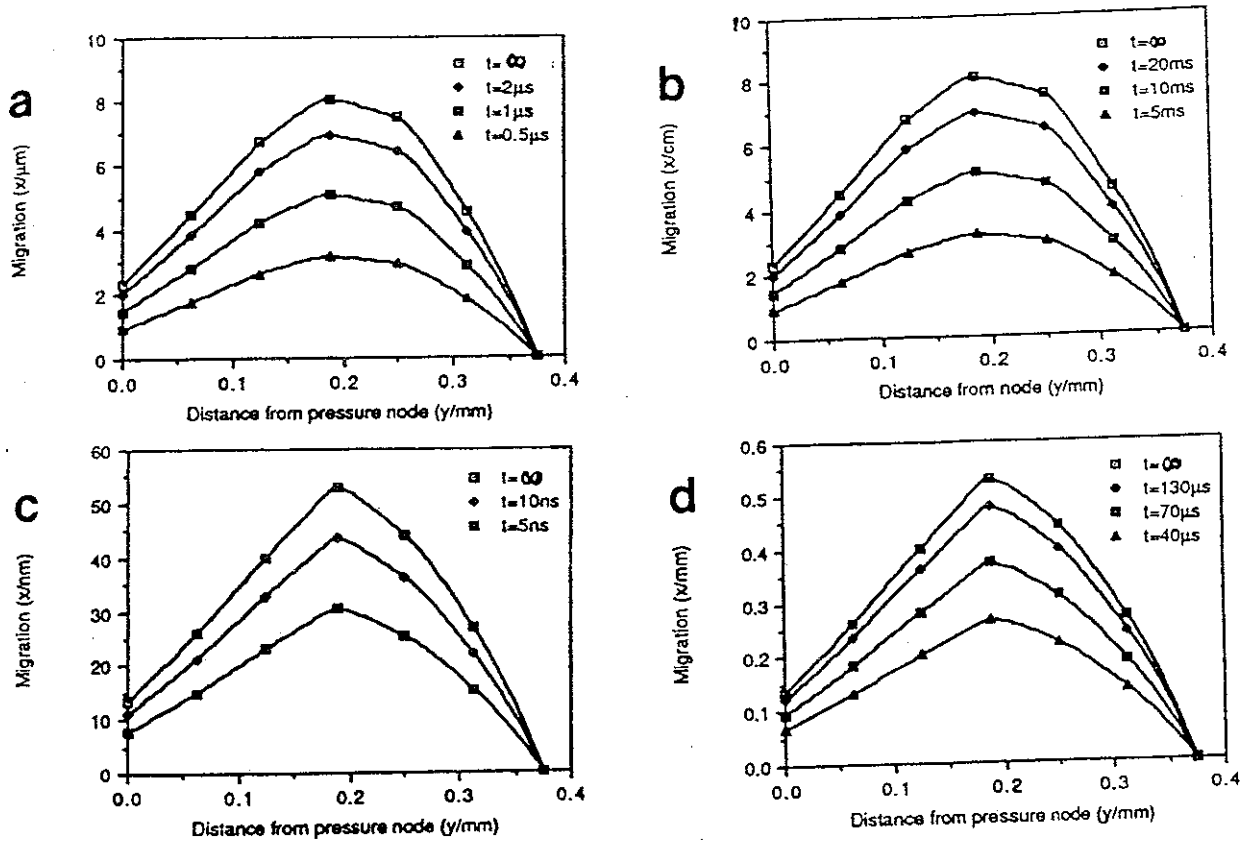

Figure 7. The observations and reasoning used in this paper to explain the form of figure 1.

Key: ICW - the light intensity from continuous-wave insonation; I1:2 - the light intensity from the 1:2 duty cycle I1:4 - the light intensity from the 1:4 duty cycle insonation; I1:7 - the light intensity from the 1:7 duty cycle

Reasoning:
1. If there were no pulse enhancement, we would expect ICW>I1:2>I1:4>I1:7 in water. This is so only at low pulse heights.
2. If the pulse enhancement were due to transient excitation, we would expect I1:2>I1:4>I1:7>ICW in water. There is some evidence of this in the middle pulse heights.
3. If the pulse enhancement were due to bubble migrations, we would expect ICW>I1:2>I1:4>I1:7 in agar. This is found to be so.
4. If the effect were due to the migration of antinodal bubbles, we would expect: ICW>I1:2>I1:4>I1:7 at both high and low pulse heights in water. This is not found to be so.
5. If the effect were due to the migration of nodal bubbles, we would expect: ICW>I1:2>I1:4>I1:7 at low pulse heights, and I1:7>I1:4>I1:2>ICW at high pulse heights, in water. This is found to be so.

Deductions: Pulse enhancement in this 1MHz system is dominated by the migration of nodal bubbles (ie. those greater than resonant size). Transient excitation contributes to a lesser extent.