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**Fragmentation of Argon gas bubbles in liquid steel**

by

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# ABSTRACT

Argon gas is used to clean and de-clog liquid steel as it is being cast, but an unwanted by-product of the process is the generation of 100  $\mu\text{m}$  to 1 mm radius bubbles in the final slab. These degrade the quality of the steel, and it is desirable that they be broken up into smaller bubbles of radius less than 100  $\mu\text{m}$  before setting.

It is postulated that acoustics might be exploited to bring about this bubble fragmentation in two ways: first, by exciting surface waves on the bubble wall of high enough amplitude that smaller bubbles are pinched off from the surface; or second, through generation of extreme volumetric pulsations which may couple with an asymmetry in the motion to then give rise to shape oscillations sufficient to bring about eventual fragmentation. Order-of-magnitude estimates of the acoustic pressure amplitudes required to bring about these processes are calculated.

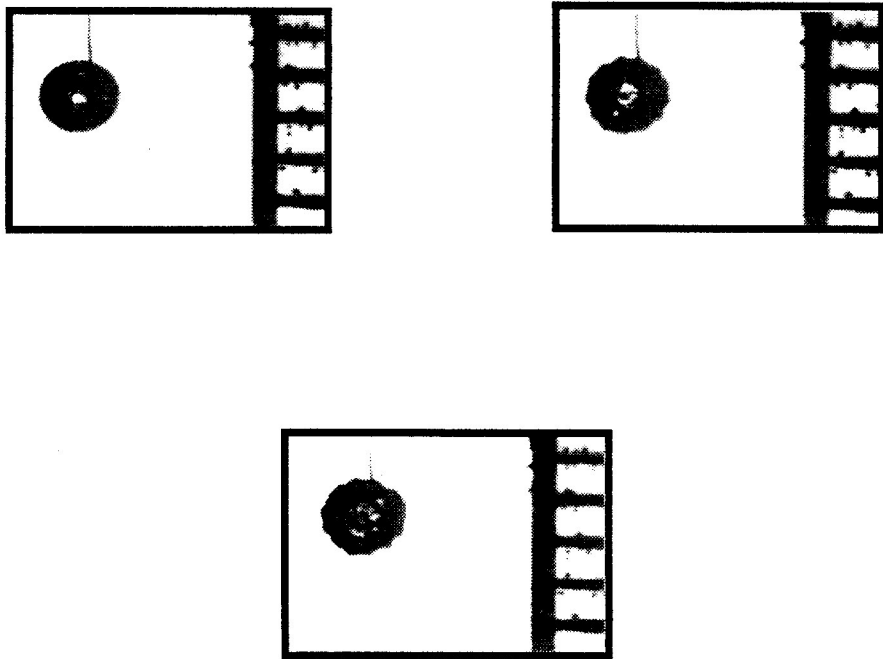
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# INTRODUCTION

Argon gas is used to clean and de-clog liquid steel as it is being cast, but an unwanted by-product of the process is the generation of 100  $\mu\text{m}$  to 1 mm radius bubbles in the final slab. These degrade the quality of the steel, and it is desirable that they be broken up into smaller bubbles of radius less than 100  $\mu\text{m}$  before setting.

It is postulated that acoustics might be exploited to bring about this bubble fragmentation in two ways: first, by exciting surface waves on the bubble wall of high enough amplitude that smaller bubbles are pinched off from the surface; or second, through generation of extreme volumetric pulsations which may couple with an asymmetry in the motion to then give rise to shape oscillations sufficient to bring about eventual fragmentation. The first of these fragmentation techniques is demonstrated in figures 1(a) to (c), photographed from a tethered bubble driven at its acoustic resonance frequency at three increasing amplitudes. The second of these techniques is demonstrated by Leighton (1994) in figure 4.29. Order-of-magnitude estimates of the acoustic pressure amplitudes required to bring about these processes can be made by determining the conditions in the liquid steel required to bring about a given amplitude of wall oscillation.



**Figure 1:** High speed photographs taken of a tethered bubble being driven at its resonance frequency at three increasing amplitudes. (a) shows a frame from the normal volumetric pulsation of the bubble, (b) shows the bubble as it is driven at an amplitude greater than that required to stimulate surface waves of the bubble wall, and (c) shows a microbubble pinched off due to the action of these waves. To the right of all three pictures is a ruler in 1 mm steps to allow an optical cross-check of the acoustic size estimate.



The dynamics of bubble fragmentation depend on the shape and stability of the bubble wall. All other things being equal, surface tension (which is numerically equal to the energy associated with a unit area of the interface) will cause a bubble to tend to a spherical form, since that minimises the surface area required to bound a given volume of gas, and so minimises the surface energy. There are various ways in which a bubble can depart from the spherical form. There is for example the effect of buoyancy which distorts the equilibrium shape. In addition, the bubble can be distorted by asymmetries in the environment, such as the proximity of other bubbles or boundary walls, gravity, shock fronts, and pressure gradients on scales small compared to the bubble radius.

The important phenomenon of transient cavitation is usually described as a procedure in which a small bubble nucleus might, when a tension is applied to the liquid, expand to a large sphere, and then undergo a violent collapse (creating, for example, gas and liquid shocks, sonoluminescence etc.). Towards the end of the collapse, instabilities might generate surface irregularities which, on rebound, could be accentuated to such an extent that the bubble would break up. Such surface distortions are intimately linked with the acceleration of the bubble wall.

Plesset (1954) studied the stability of a spherical surface using spherical harmonics to describe wall perturbations of a gas bubble in a liquid of surface tension  $\sigma$  and density  $\rho$ . The growth of these perturbations from an initially small size was the condition for instability. Plesset showed that, if  $a_n$  is the time-dependent amplitude of the perturbation described by the superimposition of the spherical harmonic of order  $n$  on a pulsating sphere of instantaneous radius  $R(t)$ , then, with the substitution  $a_n' = a_n R^{2/3}$ , the dynamics of the perturbation described by the  $n^{\text{th}}$  spherical harmonic are given by

$$\ddot{a}_n' + a_n' \left\{ (n-1)(n+1)(n+2) \frac{\sigma}{\rho R^3} - \frac{3\dot{R}^2}{4R^2} - \frac{(2n+1)\dot{R}}{2R} \right\} = 0 \quad (1)$$

The resonance frequency,  $\omega_{0n}$ , of the  $n^{\text{th}}$  spherical harmonic mode ( $n \geq 2$ ) of a gas bubble (of equilibrium radius  $R_0$ ) in a liquid is given by (Leighton, 1994):

$$\omega_{0n}^2 = (n-1)(n+1)(n+2) \frac{\sigma}{\rho R_0^3} \quad (2)$$

The small-amplitude behaviour of the surface oscillations on a pulsating bubble can be found by formulating the pulsation mode and retaining only linear terms (Hsieh, 1972, 1974). This procedure generates a Mathieu relation from equation (1), and allows the stability of the bubble wall when undergoing shape oscillations to be analysed. However, the potential for bubble fragmentation in this manner depends on the proximity of the bubble to surfaces, other bubbles etc. to provide an asymmetry in the motion, which when coupled with large amplitude pulsations would cause the bubble to become unstable. Thus the important variable in analysing the pulsation is the maximum radial displacement, as if it is high enough it would suggest that an unstable shape oscillation could be generated on the bubble, with therefore the potential for fragmentation.

The second of the possible fragmentation methods involves generating surface waves on the bubble wall, which at high amplitude would then pinch microbubbles off. The stability of interfaces undergoing periodic motion has been studied by Benjamin and Ursell (1954), who investigated the waves on the free surface of a vertically-vibrated cylinder of liquid. These waves were at half the exciting frequency, a phenomenon first recognised by Faraday (1831), and it was discovered that waves at such a frequency can be excited on a bubble wall. Surface oscillations on bubbles undergoing stable cavitation were originally observed by Kornfield and Suvarov (1944). They often manifest as an added 'shimmer' seen on the bubble surface, and can be detected acoustically (Phelps and Leighton, 1996). These surface waves are parametrically excited (i.e. there exists a driving pressure threshold below which the waves are not excited) at half the driving frequency, and are strongly coupled to the pulsation mode. Sorokin (1957), Eisenmenger (1959) and Phelps (1995) evaluated the radial displacement amplitude  $R_e$  necessary to excite these surface waves on a plane surface to be:

$$R_e = \sqrt[3]{\frac{16\eta^3}{\omega\sigma\rho^2}} \quad (3)$$

where  $\eta$  is the viscosity of the liquid. If the order of the mode is high, this plane-surface result is applicable to perturbations on a sphere. Neppiras points out that for large bubbles in pulsation resonance, the amplitude of pulsation is approximately related to the acoustic pressure by

$$R_e = \frac{p_A R_0}{3\gamma p_0 d_{tot}} \quad (4)$$

where  $p_0$  is the equilibrium pressure in the liquid,  $d_{tot}$  is the dimensionless damping constant and  $\gamma$  the ratio of the specific heat at constant pressure to that at constant volume for the gas. Neppiras (1980) combines equations (3) and (4) to give the pressure threshold to excite surface waves on a resonant air bubble in water at 20 kHz to be only 0.0025 bar, and to be 0.037 bar at 500 kHz. Phelps and Leighton (1996) use this formulation to confirm the mechanism for the production of combination-frequency waves involving the subharmonic of the bubble resonance, when two sound fields (one being close to the bubble resonance) are used to insonify a bubble. They give, as the acoustic pressure amplitude required of the resonant sound field to drive the bubble sufficiently to generate Faraday waves on the wall, as:

$$p_A = \frac{6\mu\kappa p_0 d_{tot}}{R_0} \sqrt[3]{\frac{2}{\omega\sigma\rho^2}} \quad (5)$$

## 2. METHODS

A bubble may therefore fragment through the shedding of microbubbles from surface waves, or through break up as a result of a large amplitude shape oscillation. The first technique will tend to produce many bubbles, all much smaller than the mother bubble,

which may or may not survive; the second will tend to produce a smaller number of bubbles, each of a similar size. Since both types of bubble fragmentation are associated with the displacement amplitude of the oscillation of the bubble wall, then estimates can be made of the acoustic pressure amplitudes required to generate such effects. This is done for the excitation of surface waves through calculation from the analytical solution, equation (5). However for fragmentation by shape oscillation, a numerical solution of the Rayleigh-Plesset equation is required to indicate the acoustic pressure amplitudes necessary to generate bubble pulsation amplitudes having the same order as those which, in water, have been observed to couple with shape oscillations and cause fragmentation. For both calculations key bubble parameters (such as the resonance relation) need to be estimated, requiring the quantification of the relevant gas and liquid parameters. Only some of these could be supplied by experts from the steel industry, at Hoogovens IJmuiden in The Netherlands. Others had to be estimated. The two parameter sets are given below:

(i) *Those provided in the problem specification:*

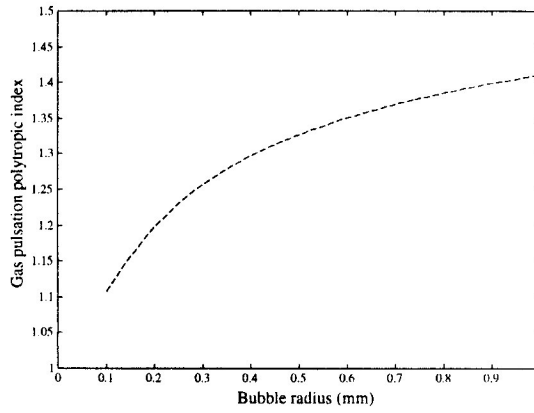
Depth of the bubbles	200 mm
Density of liquid steel	7003 kg/m <sup>3</sup>
Surface tension of liquid steel	1.865 N/m
Dynamic viscosity	6.8 x 10 <sup>-3</sup> Ns/m <sup>2</sup>
Ratio of specific heats of Argon	1.65
Temperature of casting	1550 °C

(ii) *Those assumed in the calculations:*

Speed of sound in liquid steel	6100 m/s
(Assumed to be equal to the speed of sound in solid steel)	
Heat capacity at constant pressure	0.52 x 10 <sup>3</sup> J/kgK
(Only data available was for Argon at 25 °C)	
Thermal conductivity	1.8 x 10 <sup>-2</sup> W/mK
(Only data available was for Argon at 25 °C)	

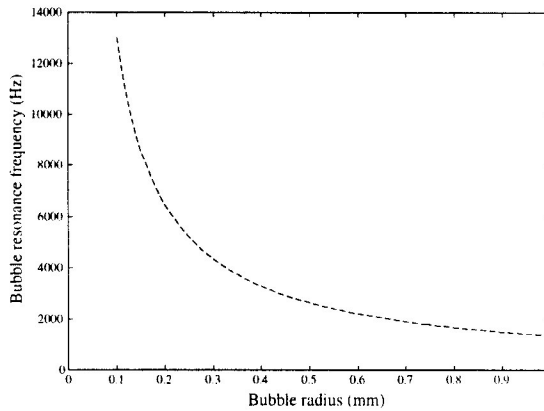
### 3. RESULTS AND DISCUSSION

Using both the accurate data provided for liquid steel and the less accurate assumed data, the first calculation involved the calculation of the likely polytropic relationship which best describes the compression of the gas inside the bubble. Unfortunately, this calculation requires the heat capacity and thermal conductivity values to be well defined, and so the accuracy of the estimates will be compromised somewhat by the use of the assumed parameters. The results are shown in figure 2 over the radius range considered.



**Figure 2:** Polytropic index of the gas pulsations when driven at their resonance. This, however, uses two of the poorly defined variables in its calculation.

It can be seen that the gas expands and contracts in a manner neither approximating adiabatic ( $\kappa = 1.65$ ) or isothermal behaviour ( $\kappa = 1.0$ ), and that the polytropic index changes considerably over the range of bubble radii used in the calculations. Using this information, the resonance frequency of the different bubbles can be calculated, and the results given in figure 3.

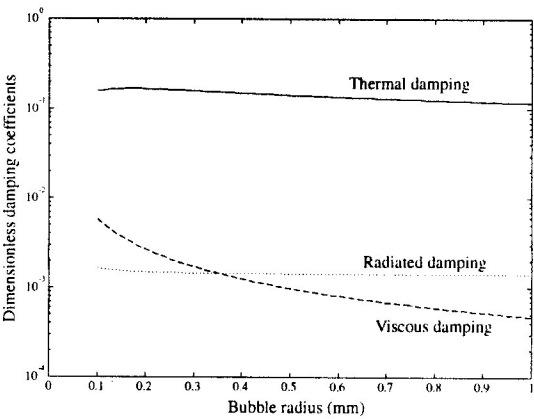


**Figure 3:** Resonance frequency of Argon bubbles in liquid steel.

The plot shows that over the radius range, the resonance frequency of the gas bubbles changes dramatically, from  $\sim 1300$  Hz at the largest bubble size to  $\sim 13$  kHz for the smaller bubbles. The calculation of these resonance frequencies does not require the use of the three poorly known variables, and thus the limits of its accuracy are solely dependant on the estimates for the polytropic indices. However, because of the high density of the liquid steel, the bubbles' resonance frequencies will change dramatically over a small change in the depth of their formation, and the estimates given in figure 3 will be accurate for bubbles at 200 mm depth only.

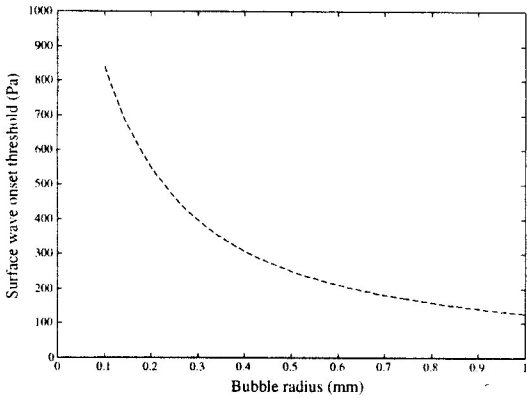
Having established the resonance frequencies of the individual bubbles, it is possible to examine how they would theoretically behave when excited by a driving sound field at their resonance. The first potential technique for breaking the bubbles up is to stimulate surface waves on the bubble which would, at high amplitudes, lead to the generation of daughter bubbles of much smaller size. This phenomenon is parametrically excited, i.e. a threshold exists for the driving pressure amplitude above which the surface waves

occur. From plane surface theory this threshold can readily be calculated, but it involves knowledge of the total damping of the bubble pulsations. This damping occurs through three mechanisms, by acoustic radiation into the liquid, by viscous losses at the surface and by thermal conduction into the liquid. As described earlier, the thermal properties of the pulsation are badly defined through a poor knowledge of the high temperature specific heat capacity and thermal conductivity parameters. However, the viscous and acoustic losses are both much better defined, and a comparison of the calculated values of the three damping coefficients is shown in figure 4 over the range of bubbles considered.



**Figure 4:** Magnitude of the three damping coefficients over the considered bubble range for an Argon bubble in liquid steel.

Unfortunately, it is clear that thermal losses account for the majority of the damping over the entire range, and so the results from the calculation of the surface waves threshold should be treated as an order of magnitude estimate only. The threshold for the onset of these waves is shown in figure 5.

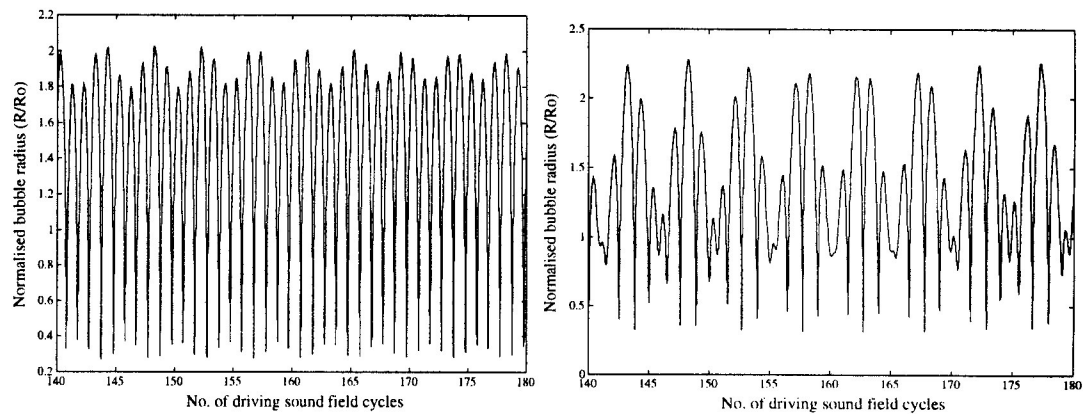


**Figure 5:** Acoustic pressure threshold for the onset of surface waves around an Argon bubble in liquid steel.

The very low pressure amplitudes (which are at largest around 1 kPa) suggest that this may be an effective way for fragmenting the bubbles. However, it should be noted that in order to pinch off smaller bubbles from the surface oscillations, the bubbles must be driven by a sound field considerably in excess of the threshold value. In addition, the daughter bubbles which are fragmented from the resonant bubble tend to be very small

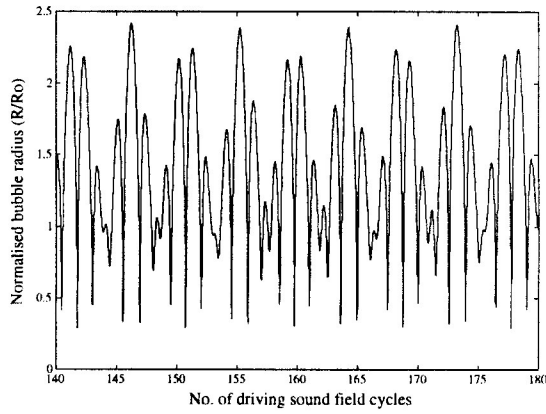
as they are formed from surface activity only, and so the technique may not yield the required reduction in the original bubble size.

The second potential fragmentation technique involves driving a trapped bubble suitably strongly at its resonance frequency to give rise to large volumetric changes, such that any asymmetry in the pulsations would become magnified and lead to shape oscillations, which in turn could cause the bubble to break up. This must be investigated numerically by solving the nonlinear Rayleigh-Plesset model for bubble dynamics, and this was performed on the two bubble sizes at either end of the considered radius range. The bubbles were driven at their resonance frequencies by sound fields of amplitude 100 kPa, and the radius time results are given in figures 6 (a) and (b). Unlike the earlier surface calculations, these results do not require the use of the poorly known thermal conductivity or heat capacity variables.



**Figure 6:** Radius-time curves for Argon bubbles in liquid steel driven at its resonance frequency by a 100 kPa sound field, with the radius normalised by dividing through by its initial value and the time converted into number of cycles of the insonifying sound field. Equilibrium bubble radius is (a) 100  $\mu$ m radius (b) 1 mm.

These two results show up some interesting points. The insonifying sound field, while being higher in amplitude than the theoretical surface wave threshold results, is still not beyond the ability of modern transducers to generate, and this sound field is shown to give rise to considerable volumetric pulsations. The two bubbles achieve maximum radii during the pulsations of 2.0 times and 2.3 times their equilibrium values for the 100  $\mu$ m and 1 mm bubbles respectively (or between 8 and 12 times their equilibrium volume). To investigate whether this radial deviation is sufficient to allow bubble fragmentation, the modelling was repeated with an air bubble in water, whose properties are better understood qualitatively. Air bubbles of 1 mm radius have been observed to fragment when excited at 100 kPa insonifying pressure (as evidenced in figure 4.29 from Leighton 1994), and the results from modelling this situation are shown in figure 7.



**Figure 7:** Radius time curves for a 1 mm radius air bubble in water driven at its resonance frequency by a 100 kPa sound field.

In this case it is apparent that the bubble radius amplitude reaches approximately 2.4 times its equilibrium value. This is only marginally higher than the 1 mm Argon bubble in liquid steel results of 2.3 times its equilibrium value, and it can be concluded that the absolute sound levels experienced by the Argon bubbles, if coupled with an asymmetry in the bubble motion, should lead to fragmentation. This break up method will give rise to daughter bubbles with a more random size distribution than of those formed by the extreme surface activity discussed earlier. However, the capability of a bubble to deviate from its spherical shape, which is also necessary for fragmentation, is determined by the relative strength of the surface tension forces. The surface tension of water is 25 times smaller than that of liquid steel, and so the shape deviation which is also necessary for fragmentation may require a greater insonifying amplitude to effect. The greater density of liquid steel will also tend to promote fragmentation through extreme shape oscillations.

## 4. CONCLUSIONS

The estimations suggest that, at sound pressure amplitudes which can be generated in liquid steel with current technologies, it should be possible to produce the fragmentation of injected argon bubbles.

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