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In situ synchrotron radiography and spectrum analysis of transient cavitation bubbles in molten aluminium alloy

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Abstract

The melt processing of conventional and advanced metallic materials with high-intensity ultrasonic vibrations significantly improves the quality and properties of molten metals during their solidification. These improvements are primarily attributed to ultrasonic cavitation: the creation, growth, pulsation, and collapse of bubbles in the liquid. However, the development of practical applications is limited by the lack of fundamental knowledge on the dynamics of the cavitation bubbles; it is very difficult to directly observe ultrasonic cavitation using conventional techniques in molten metals due their high temperature and opaqueness.

In this study, an *in situ* synchrotron radiography experiment was performed to investigate bubble dynamics in an Al-10 wt.% Cu alloy under an external ultrasound field at 30 kHz. Radiographs with an exposure time of 78 ms were collected continuously during the sonication of molten alloys at temperatures of 660±10 °C. To the best of our knowledge, this is the first time that transient cavitation bubbles have been observed in liquid aluminium. Quantification of bubble parameters such as average size and time of collapse were evaluated from radiographs using advanced image analysis. Additionally, broadband noise associated with the acoustic emissions from shock waves of transient cavitation bubbles and estimation of the real-time acoustic pressure at the driving frequency were assessed using an advanced high-temperature cavitometer in separate bulk experiments.

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Keywords: molten metals; ultrasound cavitation; transient bubbles; acoustic pressure

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1. Introduction

In recent years, there has been an increased interest in fundamental and applied investigations of liquid metal processing with ultrasound. Ultrasonic treatment offers sustainable, economical, and pollution-free solutions to melt processing, resulting in improved quality and downstream properties for the treated metallic materials [1]. However, this technology has not been extended to the treatment of large melt volumes due to the absence of fundamental knowledge of the cavitation bubble dynamics and characterization of the acoustic pressure fields in liquid metals; hence its industrial application remains rather limited. This research aims to address this knowledge gap by coupling the dynamic behaviour of acoustic cavitation bubbles in molten metal monitored using X-ray radiography with their intensity, spectral characteristics and pressure distribution measured using an advanced high-temperature cavitometer. The development of such knowledge is a prerequisite for any major technological breakthrough.

The mechanisms of ultrasonic cavitation include the formation, oscillation and collapse of cavitation bubbles accompanied from high-speed liquid micro-jets and powerful shock-waves [2]. Ultrasonic treatment has been considered as a tool to promote grain nucleation [1] and intermetallic fragmentation during solidification [3], deagglomeration of solid particles [1, 4] ,metallic filtration [1, 5], as well as melt degassing [1, 6]. Cast components with refined crystal structure have many advantages including significant improvement of product quality and mechanical properties, structural integrity and reduced grain size [1]. However, direct observations of ultrasonication processes in liquid metals have been hindered until recently by obvious limitations such as high temperatures, opaqueness and chemical activity of the melts. Nowadays, X-ray imaging technology, available through third generation synchrotron radiation sources, is extensively applied in the in situ and real-time investigations of liquid metals and their solidification processes [5, 7-10].

In the current study, two different ultrasonic transducers were deployed, having different operating driving frequencies but the same pressure outputs in the melt. The first one was used for X-ray radiography experiments while the second transducer was used for batch tests in liquid Al inside a clay-graphite crucible. In the first case, image analysis was performed to obtain statistical data only for the monitored transient cavitation bubbles (bubbles which exhibit a non-linear behaviour, with regular collapses and re-growths at random time steps within a sonication pulse). In the second case, cavitation phenomena mainly generated by similar cavitation bubbles were explained based on the spectral characteristics of acoustic emissions recorded by a calibrated high-temperature cavitometer. By bridging these different studies a more comprehensive picture of the phenomena governing the cavitation process within a liquid metal environment will be constructed.

2. Methodology

In this study two different approaches were used: a) visual observation of the real-time behaviour of the cavitation bubbles (microscale) and b) characterisation of the acoustic spectrum and the pressure field distribution in a bulk molten Al sample (macroscale). For this reason, two different experimental setups for X-ray radiography and batch tests in a crucible were deployed, as seen in Fig. 1. A custom-made in situ rig was constructed to fit the requirements of synchrotron X-ray radiography tests.

In situ synchrotron X-ray radiography studies of a molten Al-10 wt% Cu alloy subjected to ultrasonic processing were carried out using the Diamond-Manchester Branchline at the Diamond Light Source (National Synchrotron Facility in UK). A bespoke ultrasonic test rig, consisting of an ultrasonic processor coupled with a Ti sonotrode and a PID-controlled resistance furnace [11] was used (Fig. 1a). A full characterisation of the test rig can be found in [5]. A 50 W ultrasonic piezoelectric transducer (Hielscher UP50H) was deployed to introduce ultrasound into the liquid melt with a temperature of 660±10 °C. A sonication pulse was generated in a way such that the power was charged for 0.5 s then discharged for 0.5 s in every 1.0 s (i.e., the cycle is 1.0 s). A constant peak-to-peak amplitude of 28 μm and a frequency of 30 kHz were maintained. The transducer generated an intensity of 8 W/mm^2 and a pressure of 5 MPa at the 1 mm radiation face of the sonotrode which was immersed to a depth of ~4 mm in the melt. The duration of the test was 30 s and recordings of bubbles dynamic performance were obtained using a PCO-Edge camera. The rate of radiographs was 13 frames per second.

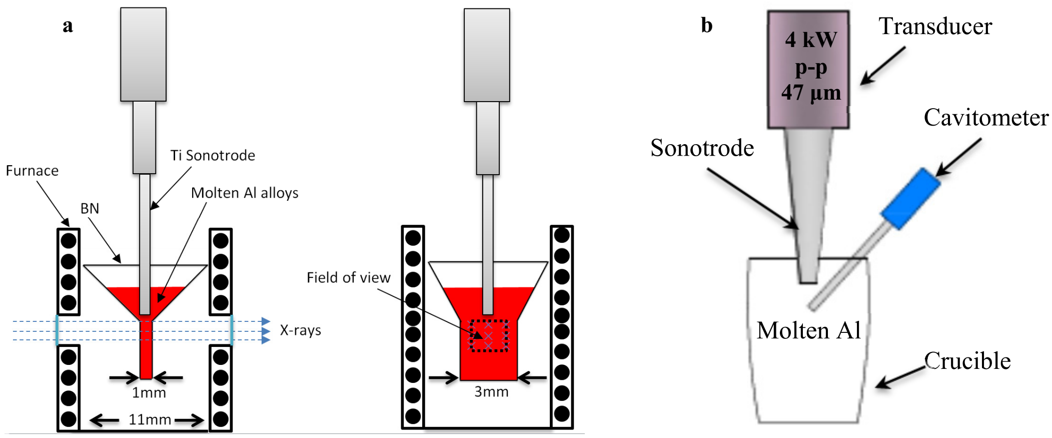

Fig. 1. a) Experimental test facility used for the ultrasonication in the beamline b) Test rig used for measuring cavitation activity in molten Al with the use of cavitometer

A similar Al alloy was selected as the material for experiments inside the crucible. The charge of 5.2 kg was introduced in a clay-graphite crucible and was melted using an electric furnace. The ultrasonic equipment consisted of a 5 kW magnetostrictive transducer (Reltec, Russia) with a Nb sonotrode of 20 mm in diameter. Experiments were performed at a driving frequency of 17.5 kHz only for the case of 4 kW (peak-to-peak 47 µm inside Al melt) as the acoustic pressure produced at the tip of the Nb sonotrode submerged to a depth of ~4 mm in the melt was the same, 5 MPa, to that of the synchrotron experiments. The intensity of cavitation inside the cavitation zone was directly measured with the use of a calibrated high-temperature cavitometer placed under the sonotrode at an angle as shown in Fig. 1b. The cavitometer used in this study is primarily designed for immersion into molten metals and consists of a tungsten probe with a diameter of 4 mm and length of 500 mm connected to a piezoelectric receiver mounted within a metallic enclosure (Belorussian State University of Informatics and Radioelectronics). A full account of the cavitometer can be found in [12]. Signal acquisition and processing was carried out using a dedicated external digital oscilloscope device Picoscope that allowed real-time signal monitoring of the cavitometer sensor's data and ultrasonic parameters. A Blackman window was applied to the raw voltage signal, which was then transformed to the frequency domain with a Fast Fourier Transform. The majority of the measurements were set to acquire 30 averages of the acoustic spectrum using a resolution bandwidth of 95 Hz. The time for this signal acquisition was approximately 30 × 1.3 (time gate) = 39 ms. In both cases there was no controlled atmosphere, and each experiment was repeated several times to ensure reproducibility of results.

3. Results

In Fig. 2 the typical behaviour of a transient cavitation bubble in molten Al is shown. The life cycle of a typical acoustic cavitation bubble in water i.e. vapour bubble, is usually determined by the acoustic cycles; the bubble reaches a maximum size and then collapses [13]. In contrast, in the case of liquid Al, we did not observe the collapse of bubbles within an acoustic cycle i.e. ~33 µs, which could not be captured due to the 78 ms integration time required to achieve contrast in the radiographs. Any bubbles only lasting 33 µs would not be imaged as they only last for 1/2000th of a frame. Instead we captured a new type of bubble formed, those that are stabilised and last many acoustic cycles, growing during each until final collapse. Interestingly, these bubbles have a resonance size that is in a good agreement with the theoretical size of a resonant Al bubble at 30 kHz estimated by the Minnaert equation [14] and found to be in the range of 60-70 µm in radius. Another interesting feature is the fact that these enhanced stability bubbles do not disintegrate into smaller satellite bubbles generating cloud of clusters or complicated bubbly structures. This could possibly reduce the maximum pressure emitted by the Al bubbles as in the presence of a cloud the pressure in the cloud centre becomes much higher than that of a single bubble [15]. The reason may be related with the relatively low power setting used to generate bubbles in this particular set up with a tiny sonotrode surface area (the number of bubbles generated is much less compared to the case of Fig. 1b), or with the relative distance of the observation window from the sonotrode tip (further away from the sonotrode tip, thus less chances of clusters to be observed, see also [5]).

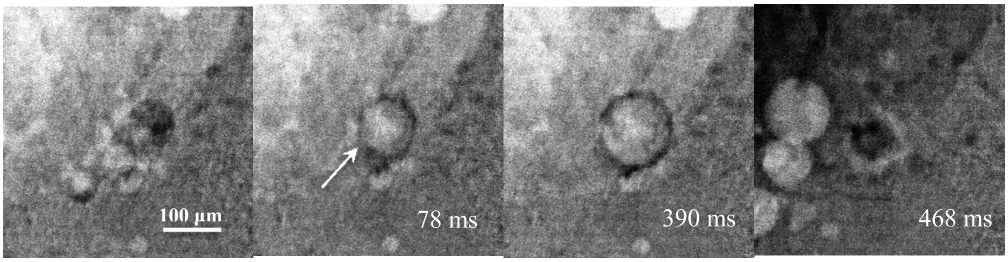

Fig. 2. Characteristic transient behaviour of an acoustic cavitation Al bubble inside liquid melt.

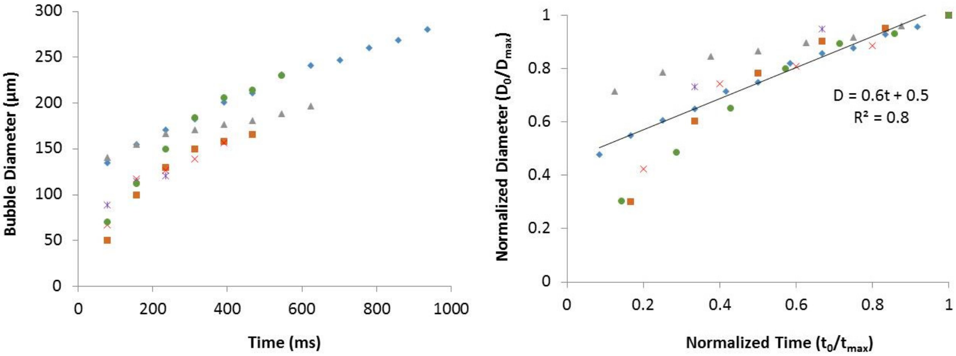

Fig. 3. Statistical measurements of cavitation Al bubbles in the synchrotron experiments showing a) evolution of bubble diameter with time until the final collapse stage b) normalization of diameter with respect to normalised time.

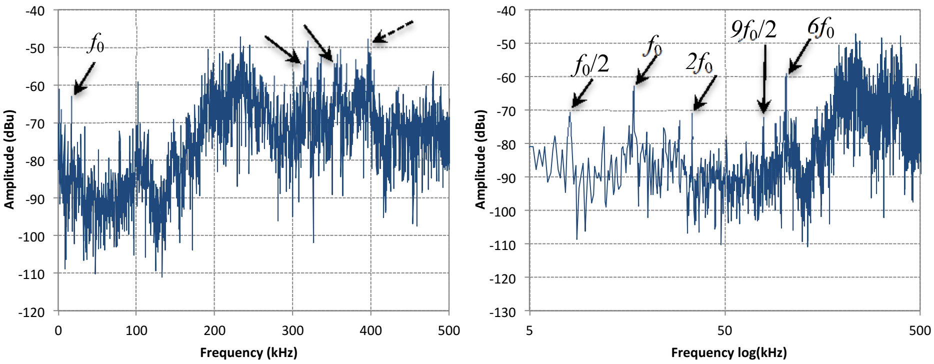

Fig. 4: a) Typical example of acoustic spectrum measured below the sonotrode's tip surface in the laboratory experiments. b) Discretization of sub- ultra- harmonics of Fig 4a.

In Fig. 3a, the variation of the cavitation bubble's diameter with respect to time is shown. It can be seen that final collapse takes place in various time steps although it seems that most of those collapses occurred near the end of the sonication cycle, at 0.5 sec. However, in some cases, bubbles can be sustained for a full cycle (1 s) and then, when sonication starts again, they violently collapse. This observation is in agreement with our previous study where the majority of the Al cavitation bubbles sustain a stable cavitation behaviour without regular collapses or rapid changes in their size [9].

Fig. 3b presents all the Fig. 3a data normalized to the maximum value of diameter against normalised time. This was achieved by dividing each of the measured values with the maximum measured value for each case. The normalised data (Fig. 3b) for both the quantities illustrates a very good match among the different cavitation bubble sizes. The average growth of bubble radii until the point of their collapse is well fitted by a linear relationship as indicated by the solid line with a good cross correlation factor of R^2=0.8.

A typical acoustic spectrum for liquid Al, as received from the inside of the cavitation zone by the cavitometer, is shown in Figure 4a. Specifically, the fundamental frequency component at 17.5 kHz (f_0) is apparent with further contributions from harmonics nf_0, subharmonic f_0/n and ultraharmonic nf_0/m (where n and m are integers and n>m) frequencies (Fig. 4b). Since cavitation bubbles survive for significant periods of time in liquid Al, it is reasonable to say that their corresponding acoustic emissions would affect the acoustic spectrum at higher frequencies. Thus, for example, single (dashed arrow) or populated (solid arrows) discrete peaks found in the range of 300-400 kHz could be possibly associated with transient behaviour (non-linear pulsations) of cavitation bubbles. The shape of the spectrum, with the rise at 160 kHz, is attributable to the variation in the sensitivity response of the cavitometer. Finally, RMS pressures were found to be at ~75 kPa at the driving frequency f_0 using the methodology from [12].

4. Conclusions

We devoted this work to the fundamental dynamics of cavitation bubbles and their spectral characteristics in a liquid Al alloy. To the best of our knowledge, this is the first time ever when the transient cavitation behaviour of liquid Al bubbles was captured on X-ray camera and analysed. Results showed that a) cavitation bubbles survive in the sonicated liquid aluminium for prolonged period of time prior to collapse, b) these longer stability bubble do not tend to cluster or disintegrate into satellite bubbles, c) evolution of their growth prior to the collapse can be expressed by a linear relationship, d) acoustic emissions associated with cavitation bubble transient behaviour contribute to the higher frequency domain of the measured acoustic spectrum, and e) RMS acoustic pressures under the tip of the sonotrode and inside the cavitation zone were found to be ~75 kPa at the driving frequency 17.5 kHz.

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