Fundamental studies on cavitation melt processing

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Abstract. The application of ultrasound to industrial casting processes has attracted research interest during the last 50 years. However, the transfer and scale-up of this advanced and promising technology to industry has been hindered by difficulties in treating large volumes of liquid metal due to the lack of understanding of certain fundamentals. In the current study experimental results on ultrasonic processing in deionised water and in liquid aluminium (Al) are reported. Cavitation activity was determined in both liquid environments and acoustic pressures were successfully measured using an advanced high-temperature cavitometer sensor. Results showed that highest cavitation intensity in the liquid bulk is achieved at lower amplitudes of the sonotrode tip than the maximum available, suggesting nonlinearity in energy transfer to the liquid, while the location of the sonotrode is seen to substantially affect cavitation activity within the liquid. Estimation of real-time acoustic pressures distributed inside a crucible with liquid Al was performed for the first time.

1. Introduction
In recent years, there has been an increased interest in fundamental and applied investigations on metal solidification with the use of ultrasound. Ultrasonic treatment of metallic alloys is driven by cavitation and bubble dynamics and it has been proven to be effective and promising for degassing and refining grain structure. However, its industrial application remains rather limited. Commercial-scale ultrasonic melt treatment is hindered by the lack of fundamental knowledge and practical models needed to optimize the ultrasonic treatment conditions, particularly those concerning, (i) the interaction between ultrasound-induced cavitation and the processed volume as well as (ii) the characteristics of cavitation zone in molten metals, which currently limits this advanced processing to laboratory scale. The development of such knowledge is required for a major technological breakthrough.

Pioneering research work using vibrations in liquid metals can date back to 1878, when Chernov [1] was trying to improve the quality of cast metal by elastic oscillations. Since that time, a significant amount of relevant research [2–5] has been conducted showing that the use of ultrasound in liquid melt alloys can refine material’s microstructure by the formation of nondendritic structures by enhanced nucleation, improved wetting, dispersing and distributing solid or immiscible phases, and reducing porosity by degassing. As a result of these effects the downstream properties of metallic alloys and their products are significantly improved [3]. Cast components with refined grain structure...
have many advantages including significant improvement of product quality and mechanical properties, structural integrity and reduced grain size. At the same time, the mechanisms of ultrasound-induced grain refinement are still under scrutiny [3, 6]. Further experimental work is required in order to clearly understand the fundamental mechanisms of sono-crystallization in liquid metals.

In the current study, two different approaches were used. The first one is related to the characterisation of cavitation activity within a particular water vessel; while the second one investigates cavitation activity within molten Al. Ultrasonic excitation for both cases is achieved by utilising an ultrasonic transducer and a horn (sonotrode) submerged into the melt. Measurements at various locations across the water vessel revealed noticeable spatial variations in the cavitation activity. The local cavitation phenomena in the vessel were explained based on the spectral characteristics of acoustic emission recorded by a high-temperature cavimeter sensor calibrated as an acoustic pressure sensor under free-field conditions, at specific frequencies at the National Physical Laboratory [7].

2. Methodology

In this study two different experimental setups for water and Al were deployed, as shown in figure 1. In the case when water was treated by sonication a titanium sonotrode with 40-mm tip diameter was driven by a 1-kW piezoelectric transducer which oscillates at a nominal fundamental frequency of 20 kHz producing maximum peak-to-peak amplitude of 17 μm (Hielscher/Germany) at the tip. The tip of the sonotrode was submerged 20 mm below the water surface. A 3 litre cylindrical vessel filled with 2 litres of deionised water was used. The reason for choosing this geometry is that the geometrical features of this water vessel are very close to the crucible’s geometry where Al melt is treated (table 1). Water temperature was adjusted at 22±1 °C. Experiments were performed at various distances across the central axis of the sonotrode, with the sonotrode placed at two different positions i) in the centre of the vessel and ii) near to the vessel’s wall in order to investigate the effect of positioning on the cavitation intensity. The relationship between cavitation activity and depth change of the sonotrode is also examined. All experiments were carried out with sonication amplitude adjusted at 50% (8.5 μm) and 100% (17 μm).

Table 1: Geometrical characteristics of the experimental vessels a) water beaker b) Al crucible

| Fluid Phase | Diameter (cm) | Height (cm) | Treated Volume (cm³) |
|-------------|--------------|-------------|----------------------|
| Water       | 16           | 15          | 2000                 |
| Aluminum    | 15           | 21          | 1925                 |

Pure Al was selected as a material for study because it has been extensively studied and widely used in metallurgical, automotive and aerospace industry as an alloy base. Additionally, Al and water liquid phases have very close kinematic viscosities while their Newtonian behaviour is similar [8] making water a frequently used candidate in physical modelling of liquid Al. The charge of 5.2 kg of Al was melted in a clay–graphite crucible coated with boron nitride (BN) from inside, with the size and geometry being close to the corresponding water vessel. The molten Al charge heated to 760 °C. The ultrasonic equipment consisted of a 5-kW generator and a 5-kW water-cooled magnetostrictive transducer (Reltec/Russia). The sonotrode consisted of a Ti concentrator and Nb tip driven by the transducer which oscillates at a nominal fundamental frequency of 17.5 kHz. The peak-to-peak amplitude was measured by a contactless vibrometer (BSUIR/Belorussia) in air and then was divided by a factor of 1.5 [3]. The sonication was performed by dipping the Nb tip (sonotrode) from the top of the melt to a depth of approximately 5 mm. The tip was preheated and the melt temperature was controlled during the process. There was no controlled atmosphere. The input power of the generator was adjusted at 3.5 kW with the corresponding amplitude at the Nb tip at 43 μm. The melt temperature was continuously monitored by a K-type thermocouple.
The intensity of cavitation and the extent of the cavitation region were directly measured using an advanced high-temperature cavitometer which is stated by the manufacturer to measure equally well in water and in liquid Al. The cavitometer used in this study is primarily designed for immersion into molten metals (made at the Belorussian State University of Radioelectronics, Dr N.V. Dezakunov) [7]. It 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. The receiving area of the tungsten waveguide at the tip of the cavitometer probe clearly forms a surface which is potentially sensitive to cavitation signals over a wide area. The receiver is then connected to analogue electronics that process the electrical signals in response to acoustical excitation. Various settings and outputs are available for optimising the detection regime. The signal acquisition and processing was carried out on a dedicated external digital oscilloscope device “Picoscope” (Pico Technology/UK) that allowed real-time signal monitoring of the cavitometer sensor’s data and ultrasonic parameters. Measurements were set to acquire 200 snapshots of the acoustic spectrum at 1 ms period of time. Each experiment was repeated to assess reproducibility of results.

3. Experimental Results and Discussion

Figure 2 shows a typical acoustic spectrum for water and liquid Al as received by the cavitometer. The acoustic source in water and in Al working at 20 and 17.5 kHz frequency, respectively, produces broadband signals well into the MHz region, and so examining acoustic signals up to MHz frequencies might be appropriate for analysing cavitation activity, however this is not within the scope of this paper. The range of signals obtained by the cavitometer for both cases are marginally different although the general shape of the spectrum plot is similar reinforcing further the opinion that water and Al can share similar behaviour under ultrasonic treatment. Specifically, the prominent fundamental frequency component is clearly shown in both graphs with further contributions from sub- and ultra-harmonic frequencies. The determinant is that harmonics in molten Al are not prominent implying that cavitation activity has not been fully developed yet (unlikely, judging from the amplitude of vibrations), or a more mild regime has been established with mainly stable cavitation bubbles present (likely) or acoustic signals received are much lower than expected due to the attenuation effects from the liquid (very likely). Additionally, another reason is that in molten Al there are not as many nuclei or pre-existing bubbles as in water, making the inception and development of powerful cavitation more difficult [3]. However, at higher frequencies, specifically in the range of 200–250 kHz prominent peaks are shown, suggesting nonlinear activity from numerous cavitation bubbles. This leads us to a preliminary conclusion that in the studied melt the activity of bubbles with resonant sizes of 10–15 μm (according to Minnaert equation [9]) prevails in the cavitation regime. This outcome is in agreement with our recent in-situ study of cavitation in Al melts, performed in the Diamond Light Source Synchrotron (National UK facility) jointly with University of Manchester,
showing after the analysis of more than 30,000 bubbles at a 30-kHz driving frequency, the majority of cavitation bubbles can be also found in that particular range [10]. For both graphs in figure 2, the broadband signal is generated by the collapsing bubbles of a wide range of sizes with their shock emissions and liquid jets to contribute further. The shape of the spectrum with this hump in the area of 160-170 kHz is attributable to the variation in the sensitivity response of the cavitometer.

Figure 2: Typical example of acoustic spectrum generated by a) 20-kHz piezoelectric ultrasonic transducer in water and b) 17.5-kHz magnetostrictive ultrasonic transducer measured with the cavitometer tip positioned about 3–4 cm below the sonotrode tip surface.

Figure 3: Cavitation activity measured in deionised water for two positions of the sonotrode a) near to the side wall b) in the centre of the cylindrical vessel, as a function of distance between the centre tip of the cavitometer and the sonotrode respectively under mechanical amplitudes of 50% and 100%.

Figure 3 shows the spatial variation data of the cavitation activity for different positions of the sonotrode within the water vessel: in the centre and near to the wall. Each point is represented by the minimum, maximum and average cavitation intensity value measured with the cavitometer at a particular distance from the tip of the sonotrode. It can be clearly seen that less power is more effective in producing higher cavitation activity. In the case where sonotrode was placed in the centre of the cylindrical vessel, 50% power was producing slightly higher cavitation activity levels than 100%. However, when sonotrode was moved near to the side wall the 50% output produces about 30% more cavitation activity as compared to 100%. These amplitude differences regarding the power output and the positioning of the sonotrode are attributable to two things: a) to the shielding and scattering effects; where the higher output level, i.e. 100%, produces an intense region of cavitation very close to the sonotrode tip surface, which prevents the further propagation of ultrasound into the liquid, thus attenuating the signal received by the cavitometer sensor and b) to the fact that activity can be higher near to the wall as bubbles can collapse easier due to the solid interface and also cavitation nuclei or many microscopic air bubbles are more available due to the surface roughness of the glass wall. Results are in a good agreement with the work of Hodnett et al. [11]. Additionally, when cavitometer was placed below the sonotrode, intensity was measured to be similar to that at 100%.
Collected data in liquid Al was numerically filtered into two groups, at 1 kHz higher and 1 kHz lower the fundamental frequency of 17.5 kHz, ensuring that all the prominent peak amplitudes will be captured. Then using the calibration data for the cavitometer obtained in the National Physical Laboratory in water at room temperature, under free-field conditions at the frequencies of interest (assuming that sensitivity does not change at temperatures up to 800 °C, as indicated by the manufacturer) these amplitudes were converted into acoustic pressures. The environment in which the sensor is calibrated is clearly significantly different from that in which it used (high temperatures, non free-field) and so computing the pressures according to the measured voltages received at 17.5 kHz is done with caution. The computed peak negative acoustic pressures were revealed across different areas of the crucible giving physical meaning to the likelihood of cavitation activity in this extreme environment (figure 4a). Acoustic pressures gradually increase as cavitometer approaches the sonotrode. This is in a good agreement with the experiments in water as intensity was also observed to be higher under the sonotrode (figure 3). Interestingly, it can be seen that the existing cavitation regime generates pressures which are relatively small compared to previous investigations that showed that the cavitation threshold is close to 0.5 MPa [6, 12]. A possible reason is that for multi bubble structures the signal level can be significantly reduced due to scattering and shielding effects occurring in the bubbly cloud along with the fact that molten Al is very dense with high surface tension.

Figure 4: a) Top view of the crucible showing locations of the acoustic measurements and the peak negative acoustic pressures at these locations b) Cavitation activity versus temperature measured at various positions of the cavitometer within the melt as depicted in figure 4a.

Also another interesting feature is that cavitation activity significantly increases with the drop of temperature as shown in figure 4b. This clearly appears in the areas away from the main cavitation zone. Below the sonotrode the intense region of cavitation maintains high levels of cavitation activity regardless of temperature variation.
4. Conclusions
Investigation of cavitation activity in liquid metals is a very complicated task due to high temperatures, opaqueness and chemical activity of the melts however with the use of appropriate equipment, as shown in this study, these obstacles can be overcome and useful data can be drawn. The main conclusions are:

1) Molten Al has presented acoustic spectrum graph comparable to water. Areas with higher cavitation activity were revealed showing that the majority of the cavitation bubbles in Al melts can be found in the range of 10-15 μm.
2) Lower power outputs i.e. 50%, seems to be the best setting for producing the higher cavitation activity. Advantages, in most of the cases, are related with the generation of the maximum cavitation intensity utilizing low power/energy. This in turns can potentially have an impact on environmental savings and on economic benefits.
3) To the best of our knowledge this is the first time when characterisation of real-time acoustic pressures inside liquid Al volume was performed. Additionally, cavitation intensity significantly increases with the drop of temperature except in the area below the sonotrode where intense cavitation is maintained regardless temperature variations.

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References:
1. D. K. Chernov “Nauka o Metallakh” Moscow: Metallurgizdat, 1950.
2. G.I. Eskin. “Ultrasonic Processing of Molten Aluminium” Moscow: Metallurgiya, 1965.
3. G. I. Eskin “Ultrasonic Treatment of Light Alloys Melts”, Amsterdam, Gordon and Breach Science Publishers, 1998.
4. J. Campbell “Effects of vibration during solidification” Intern. Met. Rev. 26 (1981) 71-108.
5. O. V. Abramov “Ultrasound in Liquid and Solid Metals” Boca Raton: CRC Press, 1994.
6. T.V. Atamanenko, D. G. Eskin, L. Zhang, L. Katgerman “Criteria of Grain Refinement Induced by Ultrasonic Melt Treatment of Aluminum Alloys Containing Zr and Ti” Metall. Mater. Trans. A, 41A (2010) 2056–2066.
7. I. Tzanakis, M. Hodnett, D.G. Eskin, B. Lebon “Advanced calibration of a high-temperature cavitometer” the 14th meeting of the European Society of Sonochemistry (ESS 2014), Avignon, France, 2-6 June 2014.
8. D. Xu, W.K. Jones, Jr., J.W. Evans “The Use of PIV in the Physical Modeling of Flow in EM or DC Casting of Aluminum: Part I. Development of the Physical Model” Metall. Mater. Trans. B, 29B (1998) 1281-1288.
9. M. Minnaert “On musical air bubbles and the sound of running water.”Phil. Mag., 16 (1933) 235–248.
10. W. Wu, I. Tzanakis, P. Srirangam, S. Terzi, W. Mirihanage, D. G. Eskin, P. Lee “In situ Synchrotron Radiography of Ultrasound Cavitation in a molten Al–10Cu alloy”, TMS Annual Meeting, Orlando, USA, 15-19 March 2015 (submitted).
11. M. Hodnett, R. Chow, B. Zeqiri “High-frequency acoustic emissions generated by a 20 kHz sonochemical horn processor detected using a novel broadband acoustic sensor: a preliminary study” Ultrason. Sonochem. 11 (2004) 441–454.
12. T.J. Mason. Advances in Sonochemistry, Volume 4, JAI Press, 1996.