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ULTRASONIC TREATMENT OF ALUMINIUM ALLOYS

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ABSTRACT

During the last years aluminium alloys have been gaining increased acceptance as structural materials in the automotive and aeronautical industries, mainly due to their light weight, good formability and corrosion resistance. However, improvement of mechanical properties is a constant in research activities, either by the development of new alloys or by microstructure manipulation.

This presentation focus a novel effective dynamic methodology for degassing metallic melts and to perform microstructural refinement / modification of light alloys, namely aluminum alloys, by applying ultrasonic vibration after melting and during solidification. This technique improves significantly the mechanical properties of those alloys, avoiding the use of traditional chemical based degassing and refining methodologies which are less effective and present significant environmental impact.

Ultrasonic vibration has proven to be extremelly effective in degassing, controlling columnar dendritic structure, reducing the size of equiaxed grains and, under some conditions, producing globular non-dendritic grains and modifying the eutectic silicon cells in AlSi alloys.

The mechanisms of ultrasonic degassing and microstructure refinement are discussed.

INTRODUCTION

The development of modern automotive and aeronautical industres lead to an intense search for light weight products with high mechanical performance. Intensive research effort aiming the development of new alloys or inovative technologies that may lead to the development of existing materials by microstructure manipulation has been carried out on the last years. Ultrasonic processing presents a new effective dynamic method for treating a molten and solidifying metal (Eskin 1998; Xu, Jian et al. 2004) in order to improve their mechanical behaviour. Ultrasonic vibration can be used to improve alloys density and to modify the solidification patterns in order to achieve fine and globular grains.

The main source of gas porosities in aluminium castings is hydrogen, which is the only gas with significant solubility in molten aluminium (Gruzleski and Closset 1990). At the freezing point, a large drop in solubility occurs, leading to hydrogen precipitation and development of gas porosities. For this reason, the hydrogen content of a molten alloy must be kept as low as possible, especially when dealing with high-strength casting alloys (Meidani and Hasan 2004; Xu, Jian, Meek and Han 2004).

Several methods are usually used to reduce the hydrogen content of aluminium melts, all of them generating great amount of slag and presenting an important environmental impact. One of the most efficient, fast and environmentally friend degassing method of Al alloys is based on the supply of ultrasonic energy to the molten metal in order to induce cavitation.

When a liquid metal is submitted to high intensity ultrasonic vibrations, the alternating pressure above the cavitation threshold creates numerous cavities in the liquid metal (Eskin 1998) which intensifies mass transfer processes and accelerates the diffusion of hydrogen from the melt to the developed bubbles. As acoustic cavitation progresses with time, adjacent bubbles touch and coalesce, growing to a size sufficient to allow them to rise up through the liquid, against gravity, until reaching surface (Eskin 1998).

Besides degasing, the development of cavitation promotes significant undercooling and consequent formation of numerous solidification nuclei. Moreover,

during cavitation strong accoustic streams develop breaking the solidifing phases and distributing them throughout the liquid metal, which will also contribute to homogenous solidification.
This effect avoids the development of dendritic structures leading to the formation of globular grains of reduced size of primary aluminium, and modify the eutectic silicon phase.

The majority of the traditional ultrasonic applications are based on fixed-frequency, well-tuned ultrasonic sources, where a large number of design and matching parameters must be respected (Abramov, Abramov et al. 1998; Xu, Jian, Meek and Han 2004). Extensive field tests conducted by experts in ultrasonics have demonstrated that in order to achieve high efficiency, the ultrasonic systems must be well tuned to the load. Since most ultrasound units work inherently in non-stationary conditions, they have to, in theory, continuously adapt themselves to the load to maximize the efficiency, which is difficult to achieve with the fixed-frequency units. To meet this challenge, novel MMM signal processing techniques have been developed by MP Interconsulting . This technique has the potential to synchronously excite many vibrating modes through the coupled harmonics and sub harmonics in solids and liquid containers to produce high intensity multimode vibrations that are uniform and repeatable (Puga, Barbosa et al. 2009). Such sonic and ultrasonic driving creates uniform and homogenous distribution of cavitation and acoustical activity on a surface and inside of the vibrating system, while avoiding the creation of stationary and standing waves, so that the whole vibrating system is fully agitated.

EXPERIMENTAL SET-UP AND PROCEDURE

Ultrasonic degassing unit

The main components of the MMM ultrasonic system used in this research consist in a high power ultrasonic converter, an acoustic wave-guide and radiator, a sweeping-frequency, adaptively modulated waveform generated by an MMM ultrasonic power supply (Figure 1a). The equipment is fully controlled through Windows compatible software developed by MPI. Optimal ultrasonic parameters (sweeping and fswm = frequency shift with modulation) for the selected resonant frequency and electric power are adjusted in order to produce the highest acoustic amplitude and largest frequency spectrum in the metal, which is monitored with a specifically implemented feedback loop. Figure 1 presents the current ultrasonic degassing unit. Besides the ultrasonic apparatus, a low frequency mechanical vibrator was coupled to the acoustic radiator, to induce a gentle stirring action in the liquid alloy (Figure 1b)

Ultrasonic solidification unit

Two different ultrasonic solidification set-up’s were used on the research - one to perform isothermal processing and another to aplly ultrasonic energy during cooling. In both cases a high power ultrasonic converter coupled to an acoustic wave-guide was used. For the isothermal processing a Ti based sonotrode was coupled to the wave guide and imersed into the liquid alloy inside a melting crucible which was an integrating part of the melting unit (Figure 2a) (Puga 2010). For the ultrasonic solidification during cooling, the wave guide was

a)

b)

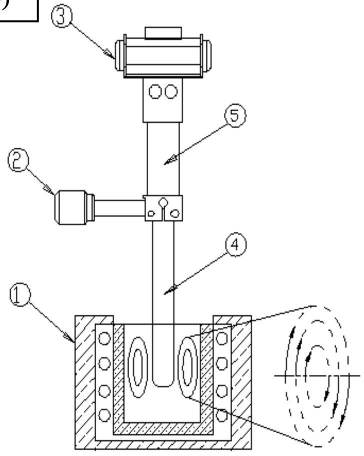


Figure 1: a) Degassing laboratorial unit; b) components of unit of degassing: (1) furnace, (2) ultrasonic transducer, (3) low frequency mechanical vibrator, (4) radiator, (5) helicoids interface.

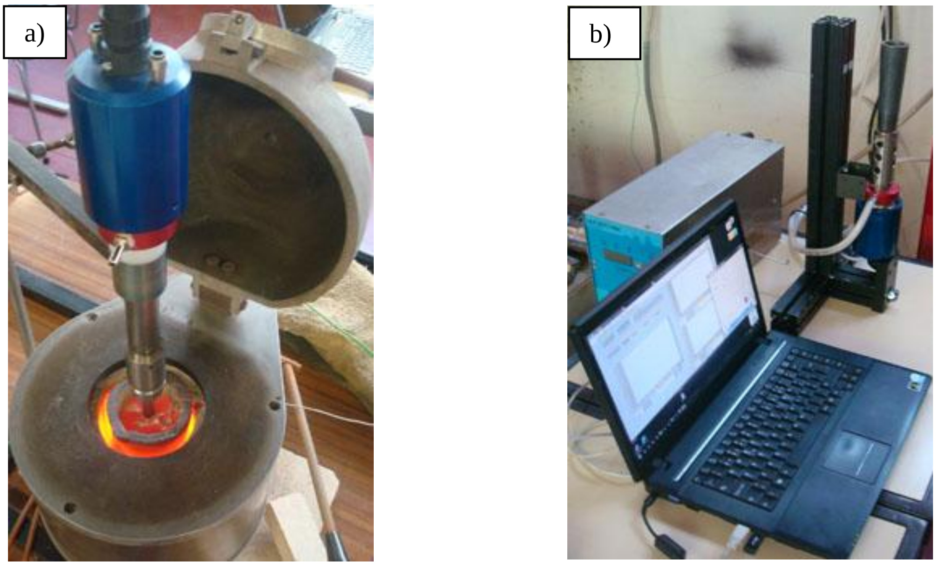


Figure 2: Ultrasonic laboratorial unit: a) isothermal processing by vibration of melt; b) by ultrasonic vibration applied to the mould.

coupled to a metallic mould were the liquid metal was poured after melting and allowed to solidify (Figure 2b).

Experimental procedure

Degassing

An AlSi9Cu3 alloy was used in this work which composition is presented in Table 1. Melting stocks weighing 4 kg were melted in a resistance furnace equipped with a 170 mm diameter and 180 mm height SiC crucible. The melt temperature was controlled within an accuracy of ±10ºC. Degassing was carried out under different processing conditions according to Table 2 using a 60 mm diameter radiator made of a titanium-based material. Due to the well-known difficulty to measure accurately the hydrogen content of aluminium alloys, results evaluation was based in the measurement of samples density, which is directly related to the samples hyrogen content.

Table 1: Chemical composition of AlSi9Cu3 alloy obtained by optical emission spectrometry.

Elem.	Cu	Fe	Mg	Mn	Si	Ti	Zn	Al
Wt %	2.9	0.7	0.24	0.26	8.3	0.03	0.4	Bal.

For every experiment and processing parameters combination, the Reduced Pressure Test (RPT) and the apparent density measurement method were used to evaluate the samples density. Tests were carried out on liquid metal, by combining the effect of low and high frequency vibration. The efficiency of ultrasonic degassing was evaluated as a function of the vibrating parameters (power and frequency), melt temperature and processing times.

Table 2: Ultrasonic degassing conditions

Ultrasonic Frequency [Hz]	19800 ± 100
Mechanical vibration [Hz]	15 – 20
Electric Power [W]	250 - 450 - 750
Degassing Time [minute]	0 – 1 – 3 – 5
Melt Temperature [ºC]	650 – 700 – 750
Ambient Humidity [RH]	58 ± 5

Solidification

The alloy was melted in a electrical resistance furnace equiped with a 1kg capacity crucible. As referred before, these experiments were carried out in two groups: isothermal processing (Figure 2a) and by ultrasonic vibration applied to the mould during cooling (Figure 2b).

In first group of experiments, the AlSi9Cu3 alloy was melted to a temperature of 700ºC, degassed with argon and allowed to cool to 615ºC. After temperature stabilization, the acoustic radiator (pre-heated to 615ºC) was dipped into the melt. The acoustic energy was then supplied for 2 minutes at different electric power levels, After ultrasonic processing, the melt was poured into a metallic mold to solidify. In the second group of experiments, the AlSi9Cu3 alloy was cast at 700ºC, degassed with argon and poured into a metallic mould with geoemtry and dimensions similar to that used for the isothermal experiments pre-heated to 250ºC. During cooling, accoustic energy was supplied to the mould at 200 W and 500 W electric power, until the metal attained a temperature 10ºC above (584ºC) the alloy *solidus* temperature.

After cooling to room temperature, samples were cut and prepared for microstructure and mechanical characterization using standard samples preparation techniques.

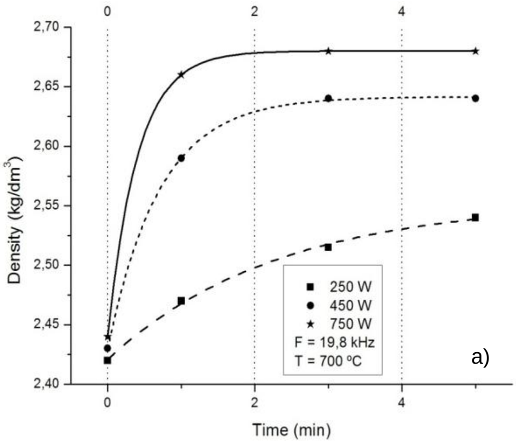
Results and Discussion

Ultrasonic degassing

i) Effect of electric power

In Figure 3 the effect of electric power in the samples density increasing rate is presented for different degassing times at 700ºC, for a frequency of 19800±100 Hz and different electric power values. It seems that the degassing efficiency is power dependent, and that this has a significant influence in both the degassing rate and the maximum alloy density.

For 750 W, a density steady-state plateau of 2.68 kgxdm^{-3} was obtained after 2 minutes ultrasonic processing. Lower electric power inputs not only decrease the maximum alloy density, but also slow down the hydrogen removal rate. For 450 W, the density steady-state plateau was about 2.64 kg x dm^{-3}, which was achieved only after 3 minutes ultrasonic processing. For lower power values the hydrogen removal efficiency is very low, as it happened using 250W, for which 4 minutes where not enough to reach the density steady-state plateau, and the alloy density did not increase for more than 2.55 kg x dm^{-3}, which is unacceptable in foundry practice.



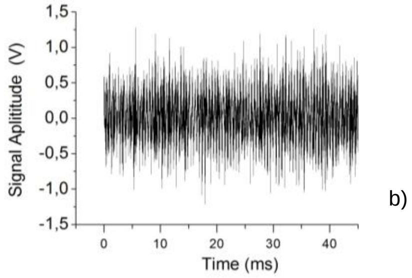


Figure 3: a) Density of the AlSi9Cu3 alloy as a function of electric power and processing time, for 700ºC melt temperature and 19800±100 Hz resonant frequency; b) acoustic waves in the molten metal for 750 W electric energy.

The influence of the electric power level in the kinetics of hydrogen removal can be easily understood by observing the amplitude of the acoustic waves inside the molten alloy presented in Figure 3b), which is a valuable indicator to the effective development of cavitation in the liquid metal (Eskin 1998). In those melts treated in a well-developed cavitation regime, which is suggested by Figure 3b for 750 W, the high density of cavitation bubbles and the partial displacement of drops of liquid in the treated volume develop strong acoustic streams that improve the coagulation of separate bubbles of hydrogen and its floating to the surface of the pool (Eskin 1998), and a consequent raise of the degassing and density increasing rates.

ii) Effect of Temperature

For temperatures below 750ºC, the degassing rate was found to be sensitive to melt temperature, increasing directly with it (Figure 4). Experiments above 750ºC were not carried on because above that temperature the hydrogen solubility in Al alloys is very high, thus increasing too much the hydrogen content of the molten alloy. On the other hand, in industrial practice aluminium alloys are usually poured at lower temperatures, thus melt treatment above 750ºC would be high energy consuming and inadequate for foundry practice.

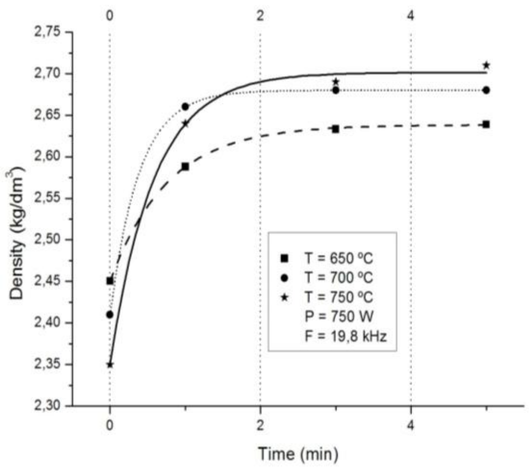


Figure 4: Density of the AlSi9Cu3 alloy as a function of melt temperature and processing time, for 19.8 kHz resonant frequency and 750W electric power.

Experimental results show that for melt temperatures of 700 and 750ºC the hydrogen removal and density increasing rates, as well as the density steady-state plateaus are quite similar (2.68 and 2.7 kg x dm^{-3} respectively). For lower melt temperatures the degassing rate slows down, and the maximum achieved alloy density is about 2.63 kg x dm^{-3}. This behaviour can be explained by the decrease of the diffusion coefficient of hydrogen in liquid metals as temperatures drops which slows down the diffusion of hydrogen from the liquid to the cavitation bubbles, and also by the decrease of the melt viscosity which makes more difficult the pulsation of cavitation bubbles, their coagulation and floating to the surface.

iii) Effect of mechanical vibration frequency

Figure 5 shows the effect of mechanical vibration to promote melt stirring on the samples density and degassing efficiency, for different processing times at 700ºC, using simultaneously ultrasonic frequency 19800±100 Hz on the acoustic radiator.

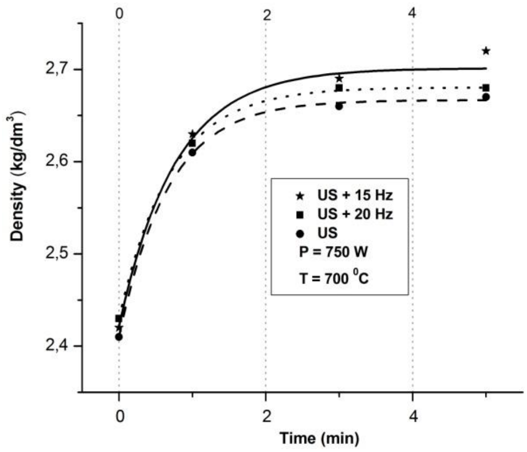


Figure 5: Density of the RPT samples and degassing efficiency of the AlSi9Cu3 alloy as a function of mechanical vibration frequency and processing time, for 700ºC melt temperature.

The maximum alloy density (2.72 kg×dm^{-3}) is 0.05 kg×dm^{-3} higher than that obtained by ultrasonic degassing (US) and was achieved after 5 minutes degassing, using 15 Hz for melt stirring (US + 15 Hz). That value is very close to the theoretical alloy density indicated by the alloy supplier (2.74 kg×dm^{-3}), and represents a degassing efficiency of 94%. For 20 Hz frequency (US + 20 Hz), the maximum alloy density was 2.67 kg×dm^{-3} which represents 83% degassing efficiency. In both cases, an improvement over single ultrasonic degassing (US) is clear although much more significant for 15 Hz vibration frequency (Figure 5). The difference to US becomes more important as the processing time increases and the dissolved hydrogen content decreases. In single US the approach of remaining H atoms to form H_2 gas molecules or its approach to cavitation bubbles is very difficult, because they are just a few and the distances between them is very high. However, the particular liquid motion profile induced by the mechanical vibrator promotes its approach to the ultrasonic radiator making it easier its diffusion into cavitation bubbles and escape to the melt surface, as show the Figure 1 b). When degassing starts this effect is not so notorious because the quantity of dissolved hydrogen atoms is much higher and distances between them and the cavitation bubbles are much smaller, making it easier the diffusion process, thus degassing.

As referred above the improvement in the degassing efficiency is due to the motion of the liquid metal that forces a greater volume of liquid to pass at distances from the acoustic radiator where cavitation develops, making it easier hydrogen removal. Best results achieved for 15 Hz mechanical vibration deal with the higher dislocation amplitude of the acoustic radiator when compared with 20Hz vibration frequencies that promoted more intense liquid motion and faster loop cycles (Puga, Teixeira et al. 2009).

Ultrasonic Solidification

i) Isothermal processing by vibration of melt

Figure 6 shows the microstructure of the AlSi9Cu3 obtained in different conditions. In Figure 6a) a fine and dendritic structure is clearly perceptible, which corresponds to a sample obtained without any refinement treatment. Figure 6b) shows a ultrasonic processed sample using 200W during 2 minutes. This microstructure still reveals a rosette-like structure and globular grains of primary α-Al with average grain size of aproximately 70 μm, suggesting that some incipient refinement was achieved. Figure 6c) shows the microstructure of a sample obtained after ultrasonic processing at 600 W for 2 minutes. The microstructure of this sample is almost totally gloabular, with an average grain size of 50 μm. This microstructure is clearly refined revealing the high refinning potential of the process.

When compared with the untreated alloy, those alloys processed by ultrasound show a much lower volume fraction of porosities, suggesting that the ultrasonic refining also promotes a significant melt degassing.

In Table 3 the mechanical properties of these alloys are presented, compared to those obtained for a similar alloy refined using traditional Al-Ti-B master alloy adition. The best results were obtained for the alloy treated at 600 W, for which a Tensile Strength of 340 Mpa and 2.8% Elongation was obtained. In what concerns to the alloy treated at 200 W, although its Tensile Strength (273 MPa) is similar to the traditionally treated alloy (277 MPa), the Yield Strength is higher in this case (200 MPa), suggesting a better homogenization of the alloy.

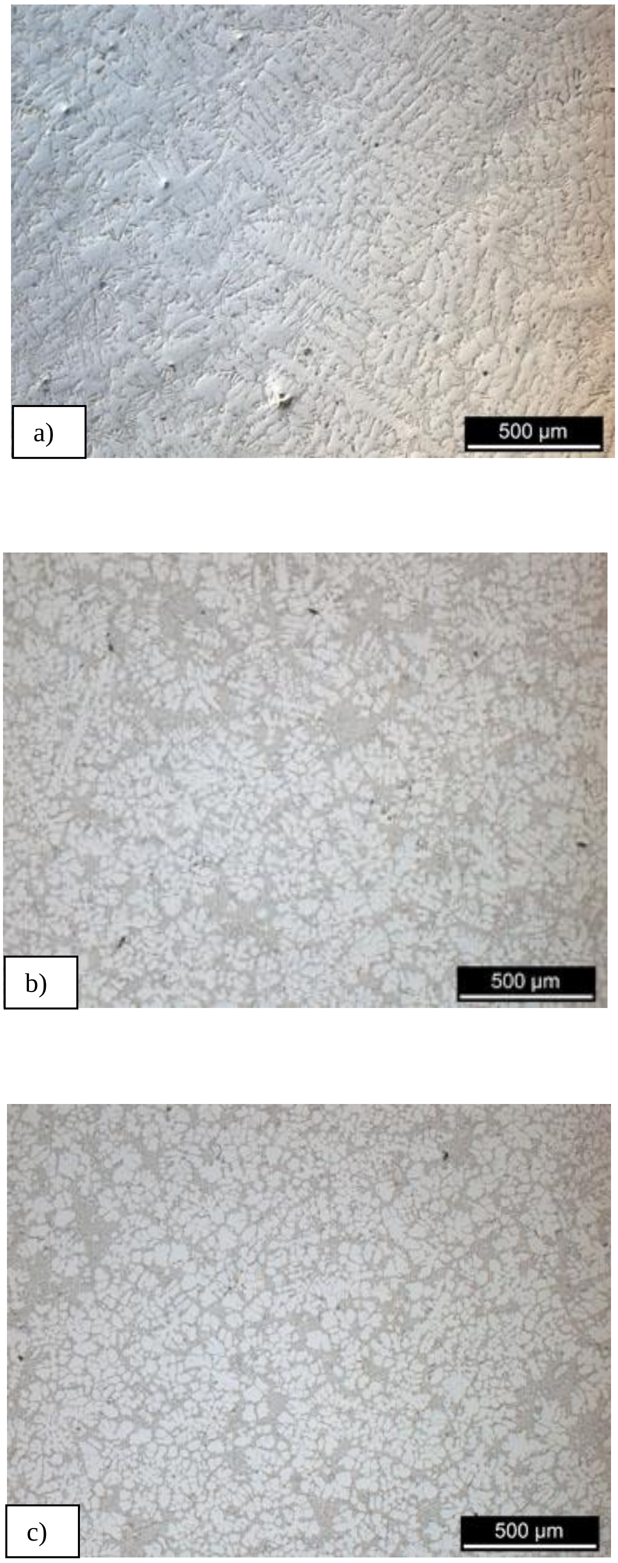
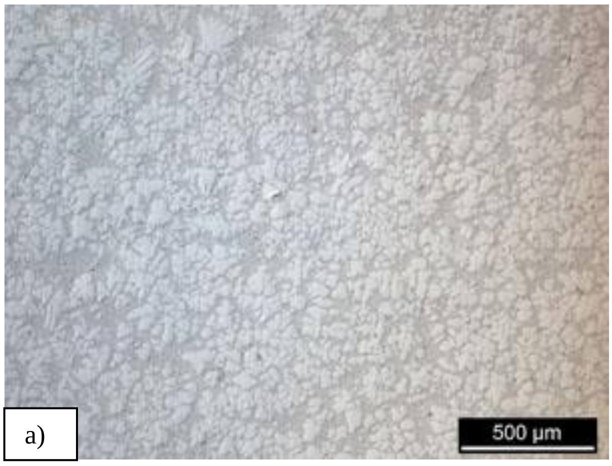


Figure 6: Microstructures of AlSi9Cu3 alloys obtained: a) Wihout US; b) with 200 W of US; c) with 600 W of US.

ii) Ultrasonic vibration applied to the mould

In Figure 7a) the microstructure of a AlSi9Cu3 sample ultrasonically treated at 600 W during cooling is presented. In this case, a small volume fraction of fine rosette-like dendrites of primary α-Al coexhists with a globular Al structure. Although the average grain size of this sample is slightly higher than that obtained with isothermal processing at 600 W, Figure 7b) reveals a strong eutectic silicon modification, which is a crucial parameter to obtain high mechanical properties. In fact, both the Tensile Strength (345 Mpa) and the Yield Strength (310 Mpa), as well as Elongation (2,9%) are much higher that those usually obtained for this alloy using traditional chemically grain refinement and Si modification (see Table 3).



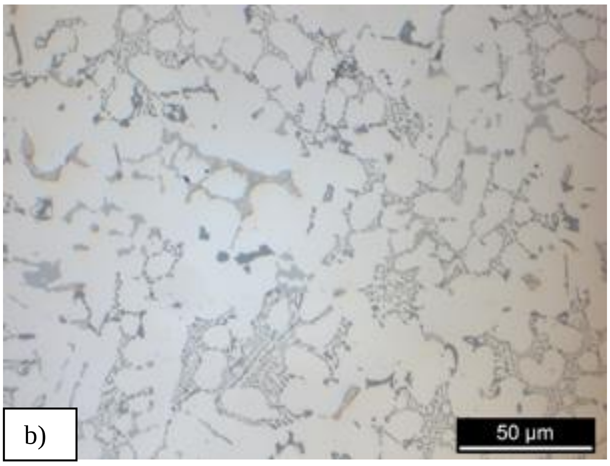


Figure 7: Microstructure of a AlSi9Cu3 sample ultrasonically treated at 600 W during cooling: a) 50 X; b) 500 x.

Although the high mechanical properties obtained with the isothermal ultrasonic processing, for industrial application the ultrasonic refinement during cooling is much easier to implement, thus future research on this field should mainly focus this technique

Table 3: Mechanical properties.

Processing	Yield strength (MPa)	Tensile strength (MPa)	Elongation (%)
UST	150	209	0.7
Al-Ti-B	170	277	1.4
Isothermal processing by vibration of melt			
200 W	200	273	0.8
400 W	250	308	1.7
600 W	280	340	2.8
Ultrasonic vibration applied to the mould			
200 W	210	301	1.6
600 W	310	345	2.9

CONCLUSIONS

- Ultrasonic degassing is an efficient technique to degas molten aluminium alloys.
- The degassing efficiency depends on the electric energy that is converted into acoustic energy, which affects the degassing rate and the final alloy density, but it is not affected by the resonant frequency.
- When compared with the traditional degassing techniques ultrasonic degassing leads to much higher degassing rates and alloys densities.
- Ultrasonic degassing is significantly improved when a stirring action is promoted in the melt
- Significant grain refinement is obtained by ultrasonic vibration either in isothermal conditions or during the cooling stage.
- High acoustic intensity promotes the formation of small and globular primary aluminum grains in Al alloys.

- Ultrasonic treatment increases significantly the tensile strength, yield strength and elongation of AlSi9Cu3 alloys.

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