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GRAIN REFINEMENT IN ALUMINUM ALLOYS BY ACOUSTIC CAVITATION PHENOMENA

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ABSTRACT

In this article, ultrasonic method of transmitting forced vibrations to solidifying aluminum-alloy melts is presented. In the presence of well developed cavitation situations, a fine and homogeneous microstructure has been observed throughout the irradiated ingots. The effects produced when high-intensity sonic or ultrasonic waves are propagated through molten metals can be listed under three main categories: grain refinement, dispersive effects, and degassing resulting in reduced porosity. It has been found that vibrations of a mechanical origin are effective in increasing fluidity by as much as a factor of three and consequently, favorably influence the mold-filling ability of aluminum alloys. There appear to be two distinct views regarding the mechanism, which may be explained by the cavitation effects and the influence of the fluid-flow phenomena.

KEYWORDS: solidification, grain size, ultrasonic field, acoustic streaming, cavitation

1. Introduction

Owing to their impact on industrial casting processes, increasing interest has been shown in fundamental and applied investigations on metal solidification, either in the presence of free convection or when various dynamic treatments generating forced convection are applied in the melt during freezing. A number of examples can be found in the literature where external forces have been applied to induce fluid flow during solidification in order to refine grain size. These methods include rotation of the mold, mechanical or electromagnetic stirring of the melt, and rheocasting [1-6]. Under these conditions, grain structures of castings and ingots change from columnar-dendritic to equiaxed dendritic or globular when they are solidified in the presence of a sufficiently intense forced convection, which generally promotes both the evacuation of superheat and the homogenization of the melt temperature. The phenomena occurring during these various treatments are currently well understood. Several investigators have found that mechanical vibrations of both sonic and ultrasonic character, when applied during the solidification of metals and alloys, modify conventionally obtained macrostructures and microstructures [7,8]. The most commonly observed effect is the suppression of undesirable dendritic and columnar zones and the development of a fine-grained equiaxed structure. In fact, the effects produced when high-intensity sonic or ultrasonic waves are propagated through molten metals can be listed under three main categories: grain refinement, dispersive effects, and degassing resulting in reduced porosity. In addition, it has been found that vibrations of a mechanical origin are effective in increasing fluidity by as much as a factor of three and consequently, favorably influence the mold-filling ability of aluminum alloys. There appear to be two distinct views regarding the mechanism, which may be explained by the cavitation effects and the influence of the fluid-flow phenomena.

Cavitation is the term used to describe the formation of bubbles, or cavities, in liquid. These cavities may be filled with air or vapor or may be almost empty; they can be produced in liquids by the passage of sonic or ultrasonic waves, provided they are of suitable frequency and intensity. Due to the oscillation of the medium, regions of compression and rarefaction are formed. In the rarefaction regions, a negative pressure (tension) may exist and air or vapor bubbles can form. In most liquid metals, a considerable amount of gas is present in the form of very small bubbles, which are, in most circumstances, seeded from pre-existing gas pockets [8,9]. Liquid vapor may also evaporate into the partial void produced by the sudden expansion of the undissolved gas bubbles. The usefulness of cavitation in processes such as cleaning, dispersion, and grain refinement is largely due to the very high pressure produced locally on the collapse of the cavities. During this collapse time, the walls of the bubble are forced inward until they impinge on the small nuclei of gas or vapor contained in the cavity, which is severely compressed at the time. It has been demonstrated that the pressure in the bubbles immediately prior to their final collapse can reach several thousand atmospheres [8-10]. Thus, when the bubbles finally disappear, extremely powerful shock waves occur, which are responsible for most of the phenomena brought about by cavitation. In particular, during metals and alloys solidification, the forces associated with cavitation result in the dislocation of growing crystals. This splitting up of crystals effectively produces many more nuclei around which new crystals can form. Thus, in this manner, the crystals never grow beyond a certain size.

The other view is that vibrations also give rise to considerable agitation of the melt and result in the newly formed nuclei being distributed throughout the solidifying pool, so that crystallization takes place uniformly inside the entire volume. Moreover, the vibrations have much the same effect as turbulence, dispersing small crystals so that more of them grow, resulting in reduced grain size.

2. Experimental setup

The sonic or ultrasonic irradiation of molten metals is mainly carried out with magnetostrictive, or piezoelectric, transducers. Coupling rods made of quartz, graphite, and various ceramic materials have been used to communicate vibrations to a molten metal, and these materials are attached to the transducer by special cements. However, such a technique presents several disadvantages. The oscillating rods are very rapidly dissolved when they are immersed into molten aluminum alloys, and this circumstance provokes an undesirable contamination of the metal. Moreover, the intensity of cavitation is greatest near the transducer or the coupling rod face; thus, the use of such a system is principally justified for the treatment of metal mixtures on a small scale.

In our experiment, the ultrasonic field was generated using a magnetostrictive transducer and an ultrasound generator. We preferred to introduce the ultrasound continuous longitudinal waves through the bottom of the crucible.

In this arrangement, there is no barrier between the ultrasound source and the melted metal. The stepped stainless steel horn was used to transmit the ultrasound to the molten and it is completely resistant to ultrasonic erosion.

Typical operating parameters were frequency of 20.338 kHz and the nominal input power of 600 W. The acoustic power dissipated by ultrasonic probe in 1000ml-deionized water at ambient temperature and pressure as a function of electrical input power was determined by calorimetry.

These data were used to allow selection of the appropriate input power to give constant transmitted power. After the completion of measurements, the ultrasonic horn was examined microscopically. No attack of the stainless steel by liquid metal samples was observed in either case, so there was no evidence of contamination of the liquid metal by alloying.

We used samples solidified in presence and in absence of ultrasonic field.

The metallography results are obtained by means of TEM (Transmission Electron Microscopy) and EDX (X-ray Photoelectron Diffraction) techniques. The TEM technique is used in the analysis of thin foils of aluminum alloys and allows obtaining important information about the substructure. The measurements were done in Physical Laboratory of **Alcan Děčín** Extrusions s.r.o., The Czech Republic.

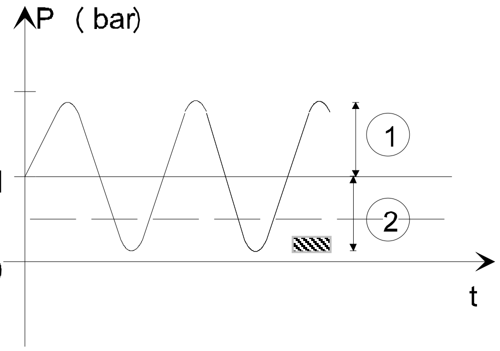
3. Results and discussions

From the hydrodynamic standpoint, the melt flow may be considered as the superposition of three motions: two alternating flows of N and $2N$ frequency, respectively, and an unstable recirculating flow that may be resolved into a steady component and a random component.

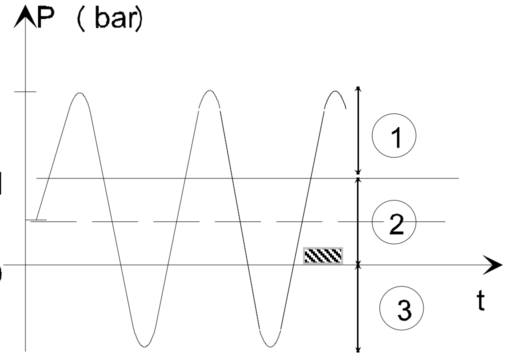
In these experiments, the pressure in the solidifying liquid metal is the sum of static and oscillating terms (Figure 1). Actually, the metallostatic pressure $p\,g\,h'$ at depth h' and the fluctuating dynamic pressure $\rho u^2/2$ can be neglected here. Accordingly, if the ultrasonic pressure amplitude is greater than the atmospheric pressure, then the pressure in the liquid will be negative (tension) for part of each period (Figure 1-b).

Cavitation takes place with full efficiency during the negative pressure part of a cycle or a series of cycles (Figure 1-b), and nucleation follows either as a result of the modification in equilibrium temperature, caused by the pressure change during the collapse of the cavitation bubbles, or as a result of cooling the surface of the bubbles by evaporation during its growth. Under this circumstance, cavitation may occur at several points inside the liquid and on the wall mould.

On the other hand, velocity measurements [11] showed that the fluctuating flow is relatively intense. When the ultrasound pressure is optimal, the shear rate reaches a value comparable to the magnitude order obtained in the rheocasting operation. It appears that during the irradiation of the semisolid alloy, the apparent viscosity remains practically unchanged up to a solid fraction on the order of 0.5, whereas the velocity decreases dramatically when the solid fraction exceeds 0.55.



a)



b)

Fig. 1. *The influence of the ultrasonic pressure amplitude on the grain refinement at **(a)** the simultaneous influence of shear rate and cavitation and **(b)** the predominance of cavitation effects, producing a fine microstructure. 1-ultrasonic pressure amplitude, 2-static pressure, and 3-tension generated in liquid (negative pressure). The dotted line corresponds to the onset of a significant grain refinement provoked by the oscillating flow, and the hachured area corresponds to the critical threshold zone of cavitation.*

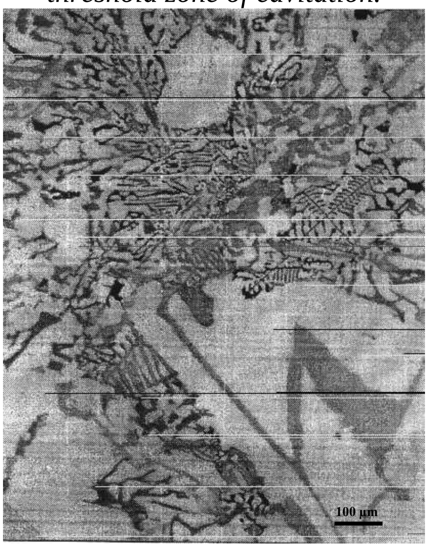


Fig.2. *Optical micrographs of the cast structure of aluminum alloy exhibits a typical dendritic structure.*

The production of cavitation in liquid metals strongly depends on the most volatile undissolved gas content of the liquid, and it has been established that in the case of aluminum melts, the hydrogen content is the controlling factor [8,12]. The solubility of hydrogen in aluminum is dependent on both the partial pressure of gas and the melt temperature. At a constant temperature, the equilibrium concentration of gas in solution is proportional to the square root of the partial pressure [13,14].

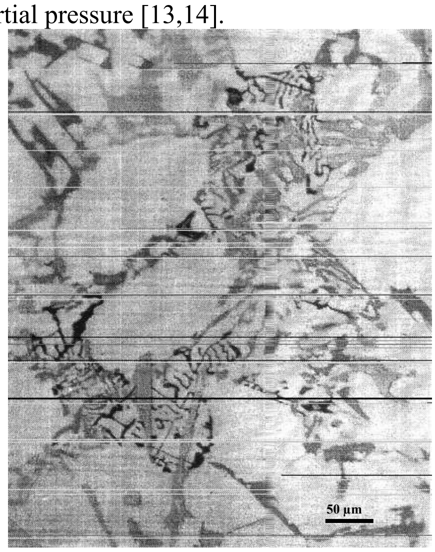


Fig.3. *The micrograph exhibits a typical nondendritic crystals (with smaller size) due to cavitation conditions.*

Figure 3 relates to the following test: an ultrasonic pressure field was applied to the solidifying alloy for five minutes from the liquidus temperature, with a cooling rate of 6°C /min. In this experiment, the operation corresponds to grain refinement, consisting mainly of the presence of an agglomeration of nondendritic crystals (with smaller size) due to cavitation conditions. This observation is understood by the disintegration of a cluster, probably under the action of a single cavitation event producing impressive mechanical damage over a microscopic region. Identical repetitive tests have confirmed this observation. In such a case, cavitation in the solidifying two-phase mixture does not take place uniformly, and systematic trials showed that the regions of intense cavitation are quite random from one experimental run to the next.

Examination of Figure 3 reveals that the grains are much finer and globular than those produced by the hydrodynamic effect in the absence of cavitation (Figure 2). Moreover, it is seen that the cavitation effects result in the total disappearance of the numerous large clusters.

It appears that contrary to the case of natural cooling melts, the ultrasonic irradiated semisolid alloy samples exhibit uniform grain refinement across the entire section. This is due to the fact that in this experiment, the ultrasonic forces that induce acoustic flow motion act uniformly throughout the semisolid melt. Moreover, it should be noted that the cavitation causes efficient degassing, resulting in tremendously reduced shrinkage and porosity [12]. Consequently, the compactness of the irradiated material is considerably improved. Under ultrasonic conditions, the acoustic flow takes place in the liquid metal. It is clearly demonstrated that the acoustic flows are associated with the ultrasound absorption, whatever its nature. However, the absorption coefficient is quite small for the liquid metals, so the increase in the temperature of the melt caused by absorption process has been eliminated. In these conditions, the reason for this prominent change in solidification kinetic is assumed to be large-scale acoustic streaming. Its effect is a permanent stirring of the melt so the effects of thermal and mass homogeneity of the melt are quite obvious. The increasing in the intensity of fluid flow can give rise to grain multiplication, which can be attributed to the increased effective nucleation rate caused by the extremely uniform temperature and composition fields in the bulk liquid at early stages of solidification. Also, the forced convection increases the growth rate. The solidification starts by heterogeneous nucleation at the crucible wall through the so- called "big-bang" mechanism. Only a fraction of the nuclei formed at this stage contributed to the formation of the chilled zone and the majority of the nuclei are transferred into the hotter bulk liquid and remelted. The final solidified microstructure depends largely on the amount of nuclei surviving after the big-bang nucleation. Under the ultrasound action both the temperature and composition fields of the liquid metal are extremely uniform. The nuclei formed will survive due to the uniform temperature field, resulting in an increased effective nucleation rate. In addition, the intensive stirring may also disperse the cluster of potential nucleation agents, giving rise to an increased number of potential nucleation sites. Also, under forced convection, the nucleation and the growth at the chilled wall were suppressed, while the nucleation and growth in the bulk liquid were enhanced [15].

It has been suggested that the forced convector fluid flow induced by ultrasound may be sufficient to break small dendrite arms and distribute them throughout the melt. If a high energy boundary is formed in a metal in contact with its liquid then the condition indicates that the grain boundary will be wetted by the liquid phase, i.e. replaced by a thin layer of liquid and thus the dendrites break appear. Further these broken dendrites act as nucleants and grow as globular nondendrite structures.

The acoustic streaming produced the change in possibility that hydrodynamic force to cause breakage of dendritic arms under the solidification conditions. In the same time, due to supplementary energy contribution, the ultrasonic field presence hinders the long-range ordering processes of atoms. At this moment, they act as nuclei for the growth of more particles and the relatively small dendrite spacing are created.

The possibility that fluid flow could disrupt the crystal bonding is also considered [16,17]. The shear forces resulting from natural convection flow of the melt are too weak to disrupt the crystal bonding during solidification. However under ultrasonic field, these forces are dramatically increasing. The accuracy of sonic measurements is reasonable taking into account the difficulties associated with getting the ultrasound into the melted metal.

The ultrasonic field presence into a liquid causes cavitation phenomenon [15]. This imposes a sinusoidal variation in pressure on a steady state ambient pressure. One new question of this study is the problem of cavitation and its microstreaming effect. The effect of ultrasound increases with increasing power, but not indefinitely since there is an optimum value beyond which the effect diminishes. When 20.338 kHz high-intensity ultrasound was applied to the molten system, a mixing of the melted metal close to the solid-liquid interface and the crucible wall due to cavitation was produced. Near the solid surface, cumulative jets can be generated and the diffusion layer is thinned due to enhanced mass transport resulting from microstreaming. In our experiment, these optimum conditions in cavitation were studied in deionized water at ambient temperature. The ultrasonic treatment of liquid metals differed essentially from that of aqueous solutions and organic liquids. This is due to the different nature of cavitation nuclei and, hence different conditions required for the initiation and development of acoustic cavitation. Only fine solid particles (mainly oxides, e.g. Al_2O_3 in aluminium melt) can act as cavitation nuclei in metallic melts. At the same time, because the molten metals feature light opacity, the cavitation cannot be studied directly.

4. Conclusion

The paper present the way how mechanical vibrations of ultrasonic character applied during the solidification of metals and alloys, modify conventionally obtained macrostructures and microstructures. The most commonly observed effect is the suppression of undesirable dendritic and columnar zones and the development of a fine-grained equiaxed structure.

Also, the ultrasonic field presence into a liquid causes cavitation phenomenon. The usefulness of cavitation in grain refinement is largely due to the very high pressure produced locally on the collapse of the cavities. When an ultrasonic pressure field was applied to the solidifying alloy the grain refinement, consisting mainly of the presence of an agglomeration of nondendritic crystals (with smaller size) due to cavitation conditions, appears. This observation is understood by the disintegration of a cluster, probably under the action of a single cavitation event producing impressive mechanical damage over a microscopic region. Identical repetitive tests have confirmed this observation.

This process presents a serious drawback for very important potential industrial applications (which consist of alloy-grain refinement), since, for the large-scale ingot castings, substantial energy consumption is required to reach the cavitation threshold.

Another technique consisting of the continuous maintenance of the melt at resonance can be considered. Under this condition, the ultrasonic power would be markedly lowered, and, consequently, the facility cost and energy expense would be decidedly reduced.

References

[1]. C. Vivès and C. Perry, 1986, Grain Refinement in Aluminium Alloys by Means of Electromagnetic Vibrations Including Cavitation Phenomena, *Int. J. Heat Mass Transfer*, 1, p. 21.

[2]. C. Vivès, 1989, Grain Refinement in Aluminium Alloys by Means of Electromagnetic Vibrations Including Cavitation Phenomena, *Metall. Trans. B*, 20B, p. 623.

[3]. C. Vivès, *Metall. Trans. B*, 20B (1989), p. 631.

[4]. M.C. Flemings, 1991, *Metall. Trans. B*, 22B, p. 269.

[5]. C. Vivès, 1992, Effect of low frequency electromagnetic field on microstructures and macrosegregation of horizontal direct chill casting aluminium alloy, *Metall. Trans. B*, 23B, p. 189.

[6]. C. Vivès, 1993, Grain refinement induced by electromagnetic stirring: A dendrite fragmentation criterion, *Metall. Trans. B,* 24B, p. 493.

[7]. J.-P. Gabathuler et al., 1992, Processing of Semi-Solid Alloys and Composites, ed. S.B. Brown and M.C. Flemings (Cambridge, MA: MIT, p. 33.

[8]. O. Abramov, 1994, Ultrasound in Liquid and Solid Metals Boca Raton, FL: CRC Press, p. 289.

[9]. T. Leighton, 1994, The Acoustic Bubble, London: Academic Press Ltd, p. 531.

[10]. J. Szekely, 1979, Fluid Flow Phenomena in Metals Processing, New York: Academic Press Ltd, p. 305.

[11]. C. Vivès, (1990) *Int. J. Heat Mass Transfer*, 33, p. 2585.

[12]. L. Moraru, S Macuta, 2006, Acoustical degassing of molten aluminium, SISOM 2006 (Annual Symposium of the Institute of Solid Mechanics), Bucuresti, 17-20 Mai, CD-ROM publication

[13]. C. Vivès, 1996, Effects of Forced Electromagnetic Vibrations during the Solidification of Aluminium Alloys: Part I. Solidification in the Presence of Crossed Alternating Electric Fields and Stationary Magnetic Fields, *Metall. Trans. B*, 27B, p. 445.

[14]. C. Vivès, 1996, Effects of Forced Electromagnetic Vibrations during the Solidification of Aluminium Alloys: Part II. Solidification in the Presence of Colinear Variable and Stationary Magnetic Fields, *Metall. Trans. B*, 27B, p. 456.

[15]. Abramov O.V., 1993, Ultrasound in liquid and solid metals, Russian Academy of Sciences, Moscow, (in English).

[16]. Moraru L. 2005, The effect of fluid flow on solidification of light metal alloy, *Transaction of the University of Kosice*, Vol 5, pp70-75.

[17]. Moraru L., **Tudose C.,** 2004, Analytic model of bonding forces in liquid metals and ultrasound influence, *Proc 2nd Int Conf Romanian Acoustical Society*, Impuls Publishing House Ed., Bucureşti, pp 75-80.