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Effect of Ultrasonic Processing of Molten Metal on Structure Formation and Improvement of Properties of High-Strength Al-Zn-Mg-Cu-Zr Alloys

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Abstract. The ultrasonic treatment of molten and solidifying metal used along with other physical methods, such as thermal treatment, filtration, etc., enables one to obtain a nondendritic structure and uniformly disperse intermetallic particles in aluminum ingots. The structure and properties of ingots and extruded shapes from high-strength Al-Zn-Mg-Cu alloys (containing up to 0.3% Zr) are shown to be considerably improved after ultrasonic treatment.

Keywords: acoustic cavitation, intermetallic crystals, nondendritic structure, solidification nuclei

Abbreviations: F, fine filtration; HT, heat treatment; UST-1, ultrasonic treatment in a trough; T, undercooling of melt in a water-cooled tray; UST-2, combination of undercooling with ultrasonic treatment; UST-3, ultrasonic treatment in the liquid ingot bath; K, reference experimental results

Introduction

The reliability and service life of components made of aluminum alloys are determined mainly by such properties as fracture toughness, fatigue endurance, fatigue crack growth rate, and corrosion resistance. An improvement of the mechanical properties of high-strength aluminum alloys occurs with the addition of zirconium. As an example, zirconium additions resulted in a nonrecrystallized structure in extruded items without a coarse-grained rim; the static strength, impact toughness, and fatigue endurance being increased [1–3].

Zirconium, being a powerful antirecrystallizing agent, to a great extent determines the microstructure of mill products having a significant influence on their corrosion resistance, in particular resistance against segregating and intergranular corrosion [3]. It is known that mill products of aluminum alloys containing 0.15% Zr possess increased resistance to segregating corrosion and higher resistance against corrosion cracking as compared with similar mill products made of an alloy containing only 0.1% Zr.

However, the positive effect of zirconium is significant only when it is present as uniformly distributed fine intermetallic particles in a homogeneous solid solution. It is known that zirconium has a relatively low solubility in solid aluminum [1]. On conventional casting, Zr addition of more than 0.15–0.20% results in the appearance of coarse primary intermetallics in the structure of ingots and mill products. Thus, the maximum improvement of aluminum alloy properties with a zirconium addition is achieved when a sufficiently high concentration of zirconium is present to give the desired fine intermetallic dispersion, while avoiding formation of coarse intermetallic inclusions in the as-cast structure. The present paper will address this situation.

V.I. Dobatkin [4] considered the process of primary intermetallic crystals formation and proposed two basic methods to suppress primary intermetallic solidification: (1) decrease the number of intermetallic solidification nuclei, and (2) the introduction of additional solidification sites to create a situation where a lack of material for intermetallic solidification prohibits the growth of intermetallic particles.

Among the various methods of grain refinement and suppression of primary Al_3Zr solidification available in Al-Zn-Mg-Cu-Zr ingots, we have chosen the ultrasonic methods of molten alloy treatment under intensive acoustical cavitation [5–11]. Ultrasonic treatment of molten metal using multilayer screen filters (Usfirals-Process) results in a significant reduction in solid particles, and ultrasonic processing of molten metal in a liquid bath leads to grain refinement down to the level of nondendritic solidification.

In most cases of continuous casting, the primary solidification occurs on solid phase particles of a different type from that of the matrix melt, i.e., heterogeneously. It is well known that oxide particles and other microscopic nonmetallic compounds (so called "plankton") being wet by the molten metal and being crystallographically similar to the matrix operate as the solidification sites.

However, only a small proportion of the "plankton" are involved in the solidification, because nonmetallic particles as a whole are not wet by the molten metal due to surface defects (cracks, recesses, etc.) filled with gaseous phase, hence they do not participate in the solidification process. Previously [6–10], we have shown that a majority of the "plankton" particles are involved in the solidification if the molten metal is subjected to the acoustical cavitation during solidification.

The transition of inert particles into an active form is induced by acoustical cavitation with the following characteristics. When the ultrasonic intensity is higher that the cavitation threshold, a cavity containing a gaseous phase is formed close to the capillary surface, because precisely in this location the cavitation strength of the liquid metal is decreased due to the gaseous phase; i.e. the gaseous phase adjacent to the capillary surface of the non-metallic particles acts as the cavitation nucleation site. The cavity formed, as a rule, collapses after several cycles of nonlinear oscillations transform the stored energy either into high pressure pulses or powerful cumulative jets of liquid. This causes wetting of the impurities with molten metal despite the capillary limitations, and the molten metal solidifies after entering the capillary. This process can be considered as an activation of impurity particles, because the molten metal that solidifies in the capillary remains solid even at temperatures above the liquidus for a given alloy.

The solid phase formed in a capillary serves as a location for the formation of transition metal (TM) aluminides; the latter, in turn, acting as solidification sites.

Our studies have shown that the ultrasonic cavitation combined with the modification of the molten metal by the addition of a TM, e.g., $\geq$0.2% Zr, forms a sufficient amount of nuclei in the vicinity of the solidification front to result in the significant refinement of the grains and transition to nondendritic solidification which proceeds without primary solidification of coarse intermetallics.

Evidently, the nondendritic structure appears due to the presence of an extremely large numbers of potential solidification sites. In this case, the formation of solidification nuclei and the formation of dendritic arms occur at the same undercooling. As in the case of dendritic crystallization, the formation and growth of grains is controlled by the undercooling at the solidification front. The reduced undercooling adjacent to the growing nucleus prevents the formation and growth of new nuclei and formation of dendritic arms.

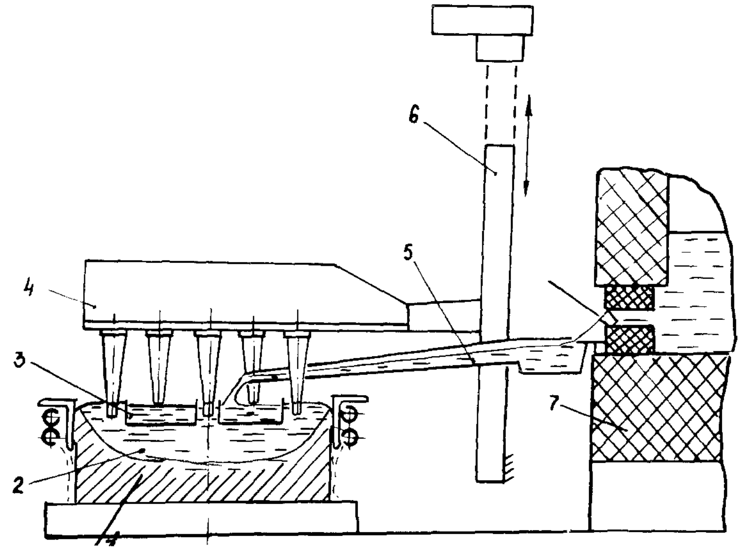

Figure 1. Schematic of large-scale ingot casting of aluminum alloys with ultrasonic processing of molten metal in the liquid bath: 1, ingot; 2, liquid bath; 3, distributing (casting) trough; 4, ultrasonic transducers; 5, casting tray; 6, moving mechanism for ultrasonic sources; and 7, holding furnace containing molten metal.

Experimental results

The procedure of nondendritic solidification is widely used in continuous casting of light alloy ingots with small, medium, and large cross-sections [5–11]. In this case, the ultrasonic processing of molten metal is performed in the liquid bath of an ingot. Figure 1 shows a diagram of continuous casting with ultrasonic treatment of aluminum alloy ingots up to 1200 mm in diameter. The ultrasonic treatment under conditions of cavitation when applied to continuous casting of large-sized commercial ingots of Al-Zn-Mg-Cu or Al-Cu-Mg alloys with 0.13–0.14% Zr leads to the formation of the nondendritic structure, the nondendritic grain size being less than 150 μm. Transition to nondendritic solidification results in enhanced ductility in large-scale ingots, which is very important for casting and subsequent hot forming operations.

The ultrasonic treatment of solidifying large-sized ingots of aluminum structural alloys gives an increase of 1.5–2.0 times in the ductility at 20°C and in the temperature range of forming (Fig. 2).

Nondendritic solidification and the improvement of ductility of the as-cast metal has the major advantage of allowing an increase in the size of the cross-section of ingots produced from high-strength Al-Zn-Mg-Cu-Zr alloys without cracking [6–9, 11]. As an example, the maximum size of round ingots made of 1973 (7050) grade alloy containing Zr was successfully increased from 830 mm to 960 mm. This result cannot be obtained by other techniques.

The second important advantage of nondendritic crystallization is a "hereditary" formation of a refined grain structure and, hence, improved ductility of mill products produced by hot deformation, virtually independent of the deformation process used.

The mechanical properties obtained for a wide variety of mill products (stampings, forgings, massive shapes, slabs, etc.) from high-strength Al-Zn-Mg-Cu-Zr alloys show that

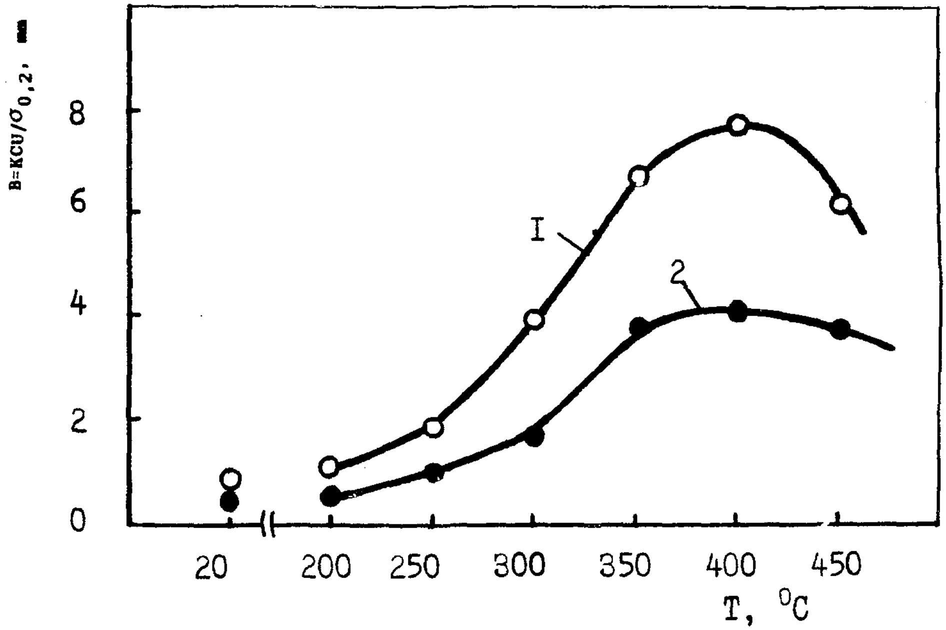

Figure 2. Effect of nondendritic structure on ductility in the hot-deformation temperature range (B = KCV/0.2% YS), where KCV is the impact toughness). Data for an ingot 830 mm in diameter from a 1973 grade alloy (Al-Zr-Mg-Cu-Zr) with 0.15% Zr: 1, casting with ultrasonic processing (nondendritic structure) and 2, casting without ultrasonic processing (dendritic structure).

Table 1. Effect of Ingot structure on height properties of small-size stamping and of large-sized extruded plates made of high-strength Al-Zn-Mg-Cu-Zr alloys.

Ingot Structure	Stampings of 1960 grade alloy (Al-8.5%Zn-2.5%Mg-2.3%Cu-0.15%Zr)		Extrusions of 1973 grade alloy (Al-6.0Zn-2.0%Mg-1.7%Cu-0.15%Zr)	
	UTS, MPa	El., %	UTS, MPa	El., %
Nondendritic	600	5.1	484	3.8
Dendritic	540	3.2	474	2.4

Note: average data of 6 to 25 measurements

the nondendritic structure of an ingot decreases the microstructure and property anisotropy over a section of deformed metal, and further increases the ductility in a height direction without strength loss (Table 1). This latter fact is very significant.

The positive effect of a nondendritic structure on the quality of wrought Al-Zn-Mg-Cu-Zr alloys was confirmed by studying particular structure-sensitive properties such as stress-corrosion resistance, creep and fatigue endurance, fracture toughness, fracture crack propagation rate, etc. [6–11]. When the concentration of Zr in Al-Zn-Mg-Cu alloys exceeds

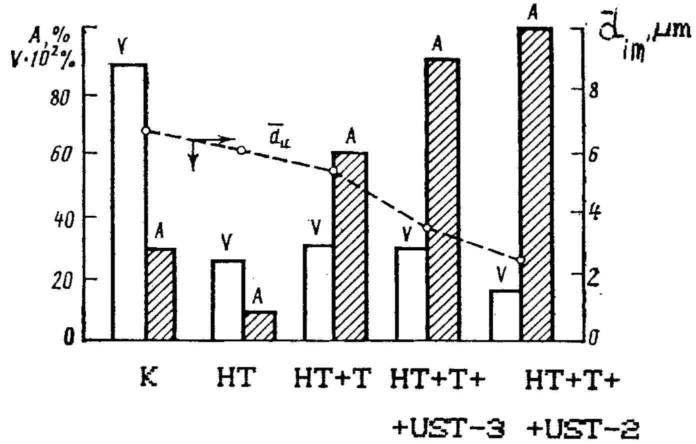

Figure 3. Effect of ultrasonic processing of molten metal in combination with other kinds of treatment on the structure of 1973 (0.3% Zr) ingots 145 mm in diameter (see explanations in text).

0.2%, the unavoidable primary solidification of Al_3Zr lowers the metal quality. The size and volume fraction of primary intermetallics depends critically on the molten metal preparation and solidification conditions.

To suppress the primary solidification of intermetallics, we used (a) the filtration of molten metal through multilayer small-mesh screen filters (F), which resulted in the capture of solid nonmetallic particles up to 1 μm in size and (b) long-time heat-treatment of molten metal (HT) to deactivate impurities.

The number of solidification nuclei can be increased by the following methods: (a) the introduction of nucleation modifiers, (b) ultrasonic processing of molten metal in a trough (UST-1), (c) undercooling of a molten metal stream in a water-cooled tray (T), (d) a combination of undercooling and ultrasonic processing (UST-3), and (e) ultrasonic processing of molten metal in the liquid bath (UST-3). These methods were applied separately and in different combinations and compared with results obtained without additional processing (K).

We studied continuous casting into a sliding solidifier of ingots 145–270 mm in diameter. The ingot structure was studied to estimate the average size ($\bar{d}_{im}$), morphology and volume fraction (v) of Al_3Zr primary intermetallics, proportion of nondendritic grains (A), and the Zr content in the matrix solid solution. The nondendritic grain fraction was evaluated in an optical microscope using the colored oxidation of specimens; the volume fraction of intermetallics was determined in a Quantimet-720 instrument; and the Zr concentration in solid solution was evaluated using a Supersonde GXA-733 setup.

The formation of coarse primary Al_3Zr intermetallics in an ingot can be prevented by means of long-term heat treatment of the molten metal in a mixer. Long-time (up to 3.0 h) overheating of molten metal to 1000°C [12] should deactivate impurities and affect the primary Al_3Zr intermetallic solidification. The results of long-term heat treatment in combination with cooling in the tray, ultrasonic processing in the trough, and ultrasonic processing in the bath during casting of 1973 alloy (0.3% Zr) ingots 145 mm in diameter are given in Fig. 3. The sequential influence of long-term heat treatment, stream undercooling, and ultrasonic processing of the molten metal decreases the intermetallic size and facilitates the nondendritic solidification.

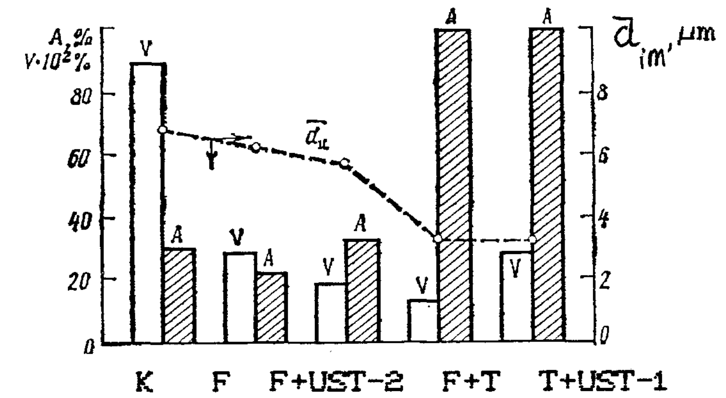

Figure 4. Effect of fine filtration (Usfirals-process) using multilayer screen filters (F) combined with other kinds of molten metal treatments on structure of 1973 (0.3% Zr) ingots, 270 mm in diameter (see explanations in text).

A more pronounced effect of molten metal preparation on the primary solidification can be observed in the structure of 270-mm diameter ingots processed using the fine filtration of molten metal in which the fine nonmetallic particles are effectively removed from the melt. These results are shown in Fig. 4.

Filtration through seven layers is so effective in purification of the melt that a nondendritic structure is difficult to obtain even using UST-2 and 0.25% Zr; the average intermetallic size is reduced and the volume fraction decreases from 0.855 to 0.199%. A five-layer filter used in combination with tray undercooling and UST-1 allows one to reduce the volume fraction and size of intermetallics as well as obtaining a completely nondendritic ingot structure.

Figure 5 shows that the average Al_3Zr intermetallic size is decreased to 3.25–3.0 μm and, corresponding volume fraction to 0.13%, accompanied by the formation of a nondendritic structure with a grain size of $\leq$50 μm.

Discussion

An analysis of the experimental results presented above allow us to postulate some general behavior characteristics.

Fine filtration of molten metal through multilayer screen filters is much more effective in removing solidification sites from the metal than the long-term overheating. The filtration through a seven-layer filter results in such an efficient purification of the melt that it is difficult to produce a nondendritic structure even when using the UST-2 mode. In contrast, the undercooling of a melt in the tray combined with the five-layer filtration assures a nondendritic structure.

The investigations have shown that the undercooling of molten metal in a tray (with and without UST-2) is an extremely effective method of nuclei formation (multiplication of solidification sites) and thus is a very efficient method for producing nondendritic solidification, refinement of intermetallics, and reduction of their volume fraction. The average Al_3Zr size is reduced to 3–4 μm with a volume fraction of only 0.15–0.28% as compared with the reference experiment in which the average size is 6 μm and the volume fraction of

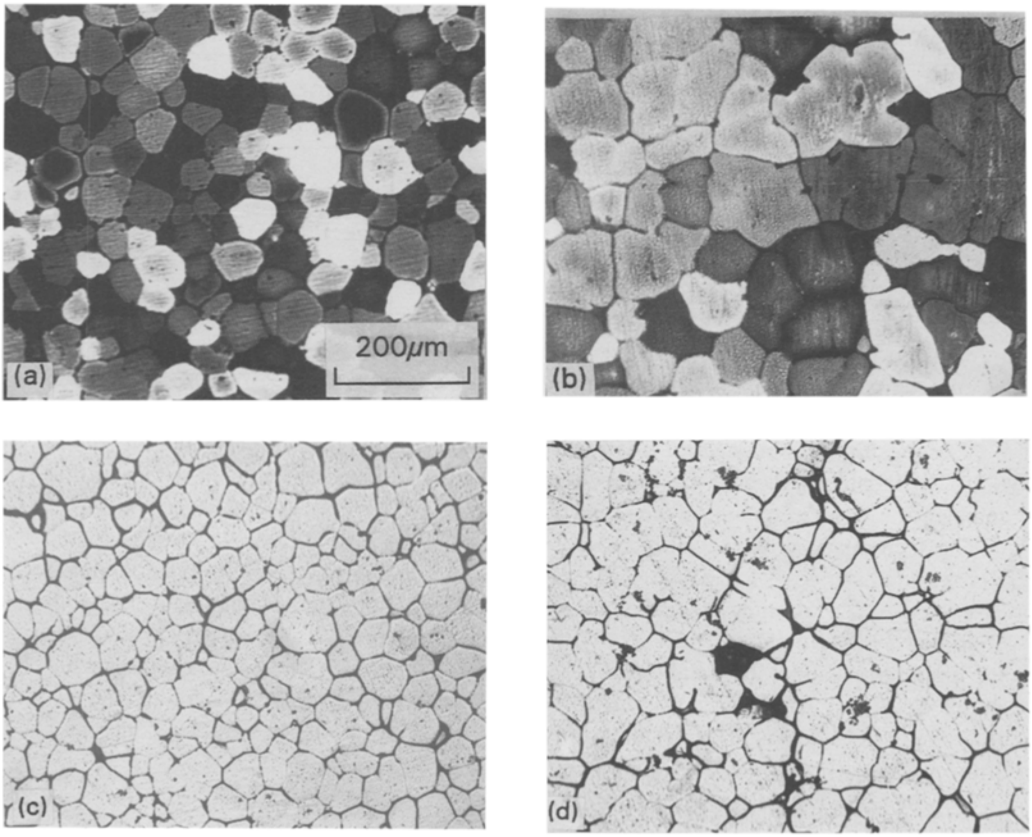

Figure 5. Grain structure of 1973 (0.3% Zr) ingots 270 mm in diameter (a, b ×100) and intermetallic particles (c, d ×100) in a standard process (b, d) and after the following treatment of the molten alloy: F + T + UST-2 (a, c).

primary intermetallics can reach 0.9%. Moreover, it must be noted that ultrasonic processing of molten metal also retards the growth of primary intermetallic crystals in the liquid bath due to additional overheating of the melt (by 20–25°C). This promotes the nondendritic solidification of ingots [5–6].

It can be assumed that such methods of casting and solidification (filtration, tray undercooling, ultrasonic processing) ensure a considerable reduction of the intermetallic Al_3Zr volume fraction and, hence, should increase the Zr concentration in the solid solution. This is confirmed by electron-probe investigations which have shown that a reduced volume fraction (v) and average size ($\bar{d}_{im}$) of the intermetallics typical of nondendritic solidification, accompany an increase in the Zr content in the solid solution (Table 2).

Figure 5c, d shows the distribution of primary intermetallic sizes in 1973 alloy ingots of 270 mm diameter. The ingots have both dendritic and nondendritic structures. The fine filtration, undercooling in the trough, and ultrasonic treatment when applied in combination can decrease the maximum intermetallic size to 10 μm, up to 80% of the Al_3Zr particles being less than 5 μm in size. In this case, Al_3Zr primary particles are of the same size as basic alloy element particles. A reference ingot produced without use of the methods

Table 2. Degree of nondendritic solidification (A) as effected by average size of Al_3Zr crystals ($\bar{d}_{im}$), volume fraction (v) and concentration of Zr in solid solution.

A, %	d_{im}, μm	v, %	Concentration of Zr in Solid Solution, %
30	6.80	0.855	0.067
80	3.65	0.290	0.073
100	3.25	0.129	0.110

discussed in the present work contains intermetallics with maximum size ($\bar{d}_{im}$) of 20–25 μm and the average size ($\bar{d}_{im}$) is 6.8 μm.

These results are obtained for small-scale (270 mm) ingots, from alloys containing up to 0.3% Zr. It can be expected that ingots to 300–400 mm in diameter will behave in a similar manner at lower Zr content, in the range 0.15–0.17%.

The "hereditary" influence of increased Zr concentration (up to 0.3%) has been evaluated for extruded strips 40 $\times$ 200 mm in cross-section produced from 1973 ingots containing dispersed Al_3Zr intermetallics, and compared with a similar strip obtained using the conventional 1973 alloy with 0.12% Zr. While tensile properties of extruded mill products in the longitudinal and transversal directions differ only by 10–15%, the low-cycle fatigue endurance (σ_{max} = 160 MPa) is increased from 110 to 200 kilocycles with a simultaneously improved stress-corrosion resistance (the stress corrosion resistance parameter increases from 150 to 200 MPa).

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