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Abstract

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# THE INFLUENCE OF THE COOLING RATE DURING CRYSTALLIZATION ON SEGREGATIONAL MICROINHOMOGENEITY AND THE COMPOSITION OF SOLID SOLUTIONS IN Al–Mg ALLOYS

*(Presented by Academician A. A. Bochvar on 11 February 1965)*

In works ([1], [2]) it was shown that, over a wide range of cooling rates (up to tens of degrees/sec), the composition of the axial regions of dendritic branches during crystallization of certain aluminum and copper alloys remains constant.

In the present work the aim was to investigate the influence of a further increase in the cooling rate on segregational microinhomogeneity and on the composition of solid solutions in Al–Mg alloys. Alloys of the following compositions were studied: 4.8; 9.6; 11.0; 13.0; 16.4; 20.5; 28.9; 33.0; 35.2 and 36.0 at.% Mg.

Different cooling rates were produced by crystallizing the alloys between two massive steel plates, by casting into a metallic chill mold. The highest cooling rates ($10^6$ degrees/sec and more) were attained in very thin films obtained by catapulting a molten droplet onto the inner wall of a rotating brass cylinder ([3], [4]). The maximum linear speed of rotation of a point on the cylinder wall was 75 m/sec. The minimum thickness of the films obtained was 0.015 mm.

The composition of dendritic cells (micrograins) was determined radiographically from the parameter of the crystal lattice of the solid solution. To convert the values of the parameter into composition, the analytical relation established from experimental data ([5]) was used:

$$a = 0.0044C + 4.041, \tag{1}$$

where $a$ is the crystal-lattice parameter, kX; $C$ is the composition, at.% Mg.

Determination of the composition of the axial regions of dendritic branches is facilitated by the fact that, in the investigated range of cooling rates, most of

Fig. 1

Figure 1: Fig. 1

the core of the micrograin acquires a constant composition. Toward the periphery of the dendritic cells the concentration of the dissolved element increases sharply. This leads to the fact that the form of the interference contour in X-ray examination has the appearance shown in Fig. 1, *a*. Region 1, with a clearly expressed maximum, corresponds to reflection from the core of the micrograin; blurred region 2 corresponds to the periphery of the micrograin. With such a form of the interference contour, the composition of the axial regions of the dendritic branches ($C_c$) can be calculated from the position of the maximum $L_c$ with a sufficient degree of accuracy ($\pm 0.75\%$ Mg). Strictly speaking, reflection from the central regions of the micrograins corresponds to the maximum of the dashed curve $L_c'$. Therefore the values of $C_c$ obtained in the work are somewhat overestimated. However, as analysis showed, this overestimation is small on the average and lies within the indicated measurement errors.

If the magnesium content in the alloy is comparatively high ($> 13\%$), the maximum composition of the peripheral regions ($C_p$) can also be determined.

*To the article by A. A. Borisov, S. M. Kogarko, and A. V. Lyubimov, p. 125*

**Fig. 1.** Development of instability on the surface of glycerin after passage of a detonation wave in a mixture of $2H_2 + O_2$ at a pressure of 1 atm. Time between frames: 95 $\mu$sec. 1—gas, 2—liquid. The detonation wave passed from left to right.

**Fig. 2.** Development of instability on a thin film of technical-grade vaseline (thickness on the order of 0.1 mm) after passage of a detonation wave in a mixture of $CH_4 + 2O_2$ at a pressure of 1 atm. Time between frames: 95 $\mu$sec. 1 —gas, 2—thin film of vaseline.

*To the article by I. S. Miroshnichenko, p. 137*

**Fig. 4.** X-ray diffraction pattern of an alloy crystallized at $v_{cool} > 10^6$ deg/sec. *a*—Al + 36.0%Mg, BRS camera, $d = 114$ mm, back-reflection method; Cu radiation.
*b*—pure aluminum.

As follows from Fig. 2, the composition of the axial regions of dendritic branches remains practically constant over a very wide range of cooling rates (from 10 to $10^6$ deg/sec). Even a certain decrease in the magnesium content is observed when the cooling rate becomes sufficiently high. The magnesium concentration in the central regions of the dendritic cells is close to the composition of the equilibrium solidus point $C_n^*$ and differs comparatively strongly from the composition of the initial liquid $C_0$. This indicates that the redistribution of components at the crystallization front has time, to a considerable extent, to occur even at cooling rates $\sim 10^6$ deg/sec.

Fig. 2

Figure 2: Fig. 2

**Fig. 1.** Shape of the interference contour in X-ray diffraction of alloys crystallized at different cooling rates ($v$):
$a$—$v_1$; $b, c$—$v_2$; $d$—$v_3$ ($v_1 \ll v_2 \ll v_3$)

**Fig. 2.** Effect of the cooling rate on the composition of the axial regions of dendritic branches ($C_c$):
$a$—Al + 9.6% Mg; $b$—Al + 13.0% Mg; $c$—Al + 16.4% Mg; $d$—Al + 20.5% Mg

In the peripheral regions, which solidify in liquid of enriched composition, the magnesium content rises sharply and reaches the limiting solubility at the eutectic temperature ($C_m$).

These regularities in the formation of the composition of the solid solution were obtained in alloys crystallized at cooling rates $\leq 10^6$ deg/sec. In very thin films crystallized on a rotating brass cylinder ($v_{cool} > 10^6$ deg/sec), fundamentally new effects are observed.

Against the background of the X-ray line considered earlier, very sharp reflections $L_0$ appear (Fig. 1, $b, c$). Their position corresponds to reflection from regions of the alloy having the same composition as the initial liquid phase ($C_0$). The curves in Fig. 1, $b$ and $c$, refer to alloys whose compositions are, respectively, lower and higher than $C_m$.

With increasing cooling rate, the maximum $L_c$, corresponding to reflection from the core of the micrograin, weakens, while the maximum $L_0$ increases. At sufficiently high cooling rates only one very sharp maximum $L_0$ is observed (Fig. 1, $d$). In this case microsegregation is absent.

It should be especially noted that no gradual shift of the maximum $L_c$ toward $L_0$ with increasing cooling rate was found. This

* $C_n = C_0 k$, where $k$ is the distribution coefficient; $C_0$ is the composition of the initial melt.

indicates that the transition from ordinary crystallization, when the magnitude of microsegregation remains practically constant, to crystallization without any traces of microsegregation takes place within a very narrow interval of change in the cooling rate (less than one order of magnitude). Moreover, in Al–Mg alloys this transitional crystallization interval lies in the region of very high cooling rates ($\sim 10^6$ deg/sec).

Figure 3 gives the values of the lattice parameters of the solid solutions $C_0$ and their composition, calculated from formula (1). All the experimental points lie close to a straight line drawn at an angle of 45° to the coordinate axes. It may therefore be considered that the composition of the solid solutions $C_0$ is equal to the composition of the initial liquid. This is also supported by the

Fig. 3. Lattice parameters and composition of solid solutions ($C_0$) in alloys of different composition

Figure 3: Fig. 3. Lattice parameters and composition of solid solutions ($C_0$) in alloys of different composition

absence, on the X-ray diffraction patterns, of lines of other phases besides the $\alpha$-solid solution, and by the sufficiently high ductility of the films obtained. The small deviations of the experimental points from the straight line lie within the errors in measuring the lattice parameter of the solid solution and the chemical composition of the initial alloy.

**Fig. 3.** Lattice parameters and composition of solid solutions ($C_0$) in alloys of different composition

The maximum lattice parameter of the solid solution obtained in the work is equal to 4.195 kX, which corresponds to a solution of 35 at.% Mg. The interference lines on the X-ray diffraction patterns of all highly supersaturated solid solutions are very sharp (Fig. 4, *a*, see insert facing p. 126). With increasing magnesium content the intensity of the lines decreases and the intensity of the diffuse background increases, which indicates an increase in the static distortions of the crystal lattice.

The attainment of high supersaturations of magnesium in solid aluminum is yet another experimental confirmation that in many cases the maximum solubility according to the equilibrium phase diagram does not characterize the limiting ability of the crystal lattice of a given component to accept atoms of another component (6). In particular, suppression of crystallization of a stable intermediate phase can lead to a considerable expansion of the solid-solution region (7).

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*Note: Figure translations are in progress. See original paper for figures.*