Structural and phase changes in metals and alloys crystallized in a gamma-ray flux

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This review presents data on the influence of γ-irradiation (10^{11}–10^{12} kW/cm^2) on the process of crystallization of metals and alloys. Studies are presented of the structure, phase composition, crystallization parameters, and redistribution of impurities along single crystals. We show that the relationship between the phase and structural components changes under irradiation, while an improvement of the structure of single crystals of metals and alloys, an enhancement of the efficiency of purifying them of impurities, more intensive mixing of alloys, and a decrease in the thickness of the diffusion layer at the melt-crystal phase boundary take place. Under the action of γ-irradiation the crystallization parameters are altered: the equilibrium temperatures of the process and the segregation coefficients of impurities. This alters the crystallization mechanism. Possible mechanisms of the observed phenomena are discussed.

INTRODUCTION

The phenomenon of crystallization of alloys and solutions is many-faceted and depends on various external factors.[1–4] The problem of obtaining crystals with a given structure and parameters is important in scientific and practical respects. In this regard the influence has been studied of various external agents—pressure, ultrasound, magnetic and electric fields[2,4]—on the structure of metals and alloys crystallizing from the melt. The task of these studies was to obtain structurally more perfect single crystals or fine-grained polycrystalline materials. A number of advances have been attained along this line (e.g., Ref. 4).

With the development of radiation physics, the study began of the action of nuclear radiation on the properties and structural and phase transitions in solids.[5–7] A substantial influence of irradiation on these processes was found. Hence one could suppose that the introduction under nuclear irradiation of a dosed amount of defects will lead to a change in the kinetics and mechanism of crystallization of solids sensitive to defects in alloys and solutions and to a change in the rate of diffusion of atoms.

The study of the action of nuclear irradiation on crystallization was first conducted by Soviet and foreign experimenters on nonmetallic objects, mainly on ionic crystals (Ref. 9 reviews these data).[8–10] It was shown that x-irradiation influences this process by altering the structure and morphology of the crystals. A change occurs in the number of centers, the rate of nucleation of crystals, and the time of the incubation period. A new, unusual phenomenon was found—it turned out that nuclear irradiation can lead not only to destruction of the material, but also to improving its structure. In ionic crystals formed in an x-ray beam, the structure and morphology are more perfect than when they are formed under usual conditions.[9,10] Here the effectiveness of the influence of irradiation on the structure of the crystals depends on their degree of purity.

1. STRUCTURE AND PHASE COMPOSITION OF ALLOYS CRYSTALLIZED IN A FLUX OF γ-QUANTA

An analysis of the results of study of the properties and structure of nonmetallic crystals crystallized under the action of nuclear irradiation obtained before 1968 and presented in Refs. 9 and 10 led the authors of Ref. 11 to conclude that one could expect also an influence of γ-irradiation on the crystallization of metals. This was confirmed by the results of studies that showed that γ-irradiation at room temperature exerts an influence on the phase and structural stability of metallic solid solutions.[5–7] However, these were mainly studies on the after effects of irradiation performed at room temperature. Of great interest were a few data (e.g., Refs. 12 and 13) that showed that γ-irradiation at elevated temperature exerts an especially strong influence on the phase and structural changes in alloys.

Taking these circumstances into account, the authors of Ref. 11 studied by x-ray and microscopic methods the structure and phase composition of metallic interstitial and substitution alloys crystallized from the melt in a flux of γ-quanta of intensity $J \approx 5 \times 10^{11}$ kW/cm^2. Crystallization of these materials was performed under ordinary conditions in the same thermal regime (without irradiation). The methodology of crystallization and structural study is described in Ref. 11. It was established that the relationship of the phase components and their structure were altered in the alloys under irradiation (Fig. 1).[9,11,14] Analysis of the integral intensity of the diffraction lines established in the high-speed steel P-18 an increase in the amount of a carbon-rich phase—the double carbide $(Fe_3W_3)C$, owing to removal under irradiation of carbon atoms from the matrix solution, austenite. Hence the microstructure of the steel was changed. In alloys crystallized under irradiation ("irradiated" alloys) one observes a directionality of the phase components. Thus, the ledeburite eutectic (austenite and tungsten carbide) is arranged in the form of narrow regions extended in one direction, which is not observed in crystallization under ordinary conditions ("unirradiated" alloys) (Fig. 1a).

Analogous phenomena have been found also in the crystallization of cast iron under γ-irradiation. It contains a considerably greater amount of free carbon in the form of graphite inclusions than in nonirradiated material (Fig. 1b). This results from the decay under irradiation of the cementite contained in the cast iron, as is implied by the microscopic and x-ray data. Thus, the integral intensity of the (002)

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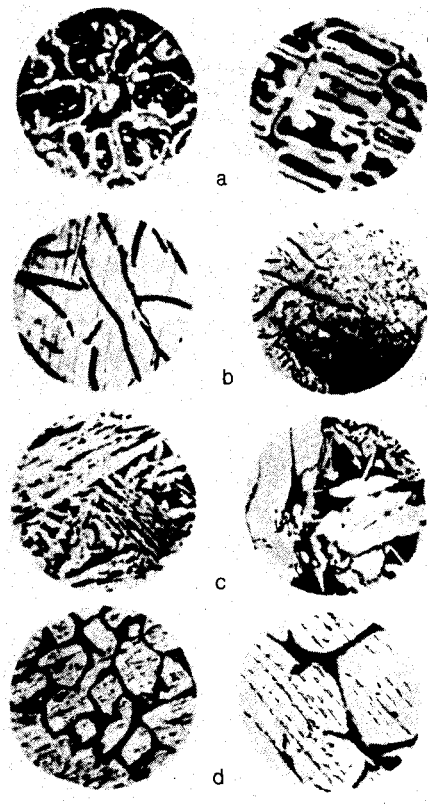

FIG. 1. Microstructure of unirradiated (left) and irradiated (right) specimens. a–High-speed steel R-18. b–Cast iron. c- – $\alpha+\beta$-brass (the white regions are the α-phase). d–α-brass. $\times 200$

diffraction line of graphite in irradiated specimens is twofold greater than in unirradiated ones.[11] Thus graphite is formed intensively when γ-irradiation acts on the crystallization of cast iron. This agrees with the data of Ref. 12, where complete graphitization of cementite was observed after irradiation with a dose of 5×10^{18} kW/cm^2 at temperatures 600–620 °C. The intensification of the process of graphite formation under γ-irradiation diminishes the amount of cementite in the irradiated cast iron. This is confirmed by the microscopical data. The microhardness (H_v) of the metallic basis of cast iron—pearlite (ferrite + cementite) is lower in the irradiated specimens than in the unirradiated ones (Table I).

In the crystallization of cast iron under γ-irradiation, a change also occurs in the dimensions and form of the graphite inclusions. They become more fine-grained and more uniformly distributed throughout the ingot, while rosette precipitates appear alongside the plate-like precipitates (Fig. 1b). As is known, cast iron that contains such graphite particles is characterized by improved mechanical properties.[15] Usually this is attained in practice by treating the cast iron by introducing modifiers—admixtures that facilitate the grain refining of the graphite inclusions.[15]

A change in the quantitative relationship of the phases in crystallization in a flux of γ-quanta has been found also in the substitution alloys of Cu and Zn–α-brass and $\alpha+\beta$-brass (L-68, 32 weight % Zn, and L-59, 41 weight % Zn,[16] respectively).[11,14] It was established that the the microstructures of the irradiated and unirradiated brass differ substantially (Fig. 1c). In unirradiated L-59 brass one observes the usual "basket" structure with narrow and relatively small-grained regions of the α-phase (Fig. 1c). A change in the form of the crystals of the α-phase occurs upon irradiation. They constitute coarsened regions occupying a large area. The hardness of the irradiated specimens of L-59 amounts to 44 units, and 49 units for unirradiated. This difference in hardness is due to the large amount in the irradiated specimens of the α-phase, less hard than the β-phase. This is also confirmed by the fact that in the irradiated L-59 alloy the intensity of the x-ray lines of the α-phase is greater than in the unirradiated one.[11,14] These data and the microscopic studies lead to the conclusion that the stability of the α-phase is elevated under irradiation, and the formation of the β-phase is retarded.

It was also established[11,14] that the action of irradiation in the crystallization of alloys leads to an appreciable increase in the dimensions of the phase constituents of the alloys (see Fig. 1). Thus, in the irradiation of α-brass, the dimensions of the grains were increased severalfold (Fig. 1d), this effect being enhanced upon increasing the irradiation dose.[14] In $\alpha+\beta$-brass the regions of the α-phase are made coarser. An estimate of the dimensions of the particles of the double carbide $(Fe_3W_3)C$ in R-18 steel from the half-width of the (511) x-ray lines yields a value of 250 Å in irradiated specimens and 180 Å in unirradiated.

Two hypotheses have been advanced to explain the effect of grain coarsening in crystallization under irradiation. One of them is the possible breakdown in the alloys under the action of ionizing radiation of certain crystallization centers, which leads to a decrease in their number, and hence to an increase in dimensions. In Ref. 14 the grain coarsening in alloys is explained by a possible increase upon irradiation in the rate of migration of grain boundaries from their center. Later this hypothesis was confirmed experimentally in Ref. 17, which established that the grain boundaries in nickel directly acted on by an electron beam of intensity $\sim10^{16}$ electrons/(cm$^2\cdot$s) at temperatures of 600 and 650 °C migrate from the center analogously to what is observed in recrystallization.

In studying the structure of alloys crystallized under γ-irradiation, another phenomenon has been discovered that is important for understanding the nature of the processes being discussed. It was established by x-ray diffraction that the half-width of the diffraction reflections obtained from the phase components of irradiated specimens is smaller than for unirradiated specimens (Table II).[11] As is known, this indicates a more perfect crystal structure of the phase components in the irradiated specimens. Thus, e.g., a calculation

TABLE I. H_v, kg-weight/mm^2.

Alloy	Stainless steel austenite	High-speed steel R-18, martensite	Cast iron pearlite
Without irradiation	230	1000	220
with irradiation	200	800	130

TABLE II. β, radians $\times 10^3$.

Alloy	Stainless steel austenite		High-speed steel			Cast iron ferrite
			Martensite		$(Fe_3W_3)C$	
hkl	(111)	(311)	(110)	(220)	(511)	(311)
Without irradiation	9,0	25,0	17,1	96,0	13,2	33,0
After irradiation	8,2	19,8	15,2	59,5	10,5	24,5

of the crystal-lattice distortions ($\Delta a/a$) and the dimensions of the mosaic blocks (L_m) by harmonic analysis of the shape of the (220) diffraction line of doped martensite showed that in the martensite of unirradiated steel the values are $\Delta a/a = 4\times 10^{-3}$ and $L_m = 200$ Å, while in irradiated steel they amount to 3×10^{-3} and 200 Å, respectively. The more perfect structure of the phase components in alloys crystallized under irradiation is also evidenced by their smaller microhardness (see Table I).

An analysis of the obtained data indicates that γ-irradiation exerts a substantial influence on the process of crystallization of metallic alloys. To understand the nature of this complex process, a number of phenomena have been studied in the crystallization of pure metals, and the role has been examined of impurities in the structural changes of alloys under γ-irradiation. These studies are presented in the following sections of this review.

2. STRUCTURE OF SINGLE CRYSTALS OF ALUMINUM CRYSTALLIZED IN A FLUX OF γ-QUANTA

The data of Sec. 1 showed that in the crystallization of alloys under γ-irradiation one can obtain single crystals of metals with more perfect structure. Since this problem is important in practice, the further attention of the authors of Ref. 11 was concentrated on studying this phenomenon. For this purpose they studied the substructure of single crystals of aluminum grown under γ-irradiation.[9,18-21]

Aluminum is a convenient object for study because of its relatively low melting point and well developed methods for studying its structure. Single crystals of aluminum of grade AVOOO (99.996% Al) were grown in an apparatus especially made for working in a γ-ray field[20,21] by a modified Bridgman method in high-purity graphite crucibles of grade MGOSCh. The dimensions of the single crystals that were obtained were $5\times 10\times 50$ mm^3 and $10\times 10\times 60$ mm^3. The thermograms, recorded at a remote station with an ÉPP 09 MZ potentiometer to record the change in temperature of the specimens with three thermocouples arranged at the middle and the two ends of the ingots being crystallized, showed that a strictly defined thermal regime was maintained in all experiments under irradiation and under ordinary conditions.

Four series of experiments were performed. In the first series the aluminum single crystals were grown completely in a γ-ray field. In the second series the crystallization was performed in the irradiation zone, but with the ^{60}Co sources absent. In the third series the nucleation and growth of the single crystals began in the γ-ray field. When the ingot had grown by about one-third of its length, the γ-ray field was removed, and further growth of the single crystals was conducted without irradiation. In the fourth series of experiments the growth of the monocrystals was begun without irradiation. When the single crystals had grown to one-third of their length, the γ-ray sources were introduced into the irradiation zone. After this, the irradiation continued to the end of growth of the single crystals. The intensity of irradiation in all the experiments was $(1\text{–}3)\times 10^{12}$ kW/cm^2. Thus information was obtained on the stage (nucleation or growth of the crystals) in which γ-irradiation acts most effectively.

The substructure of the single crystals was studied by x-ray (topographic) and metallographic methods.[9,18-21] The microhardness was measured in the PMT-3 instrument under a load of 10 g. The x-ray topographic study of the surface of the single crystals was performed in a divergent beam of white x-rays by a modified Schultz method.[18,21] Refinements were introduced into the method so as to obtain contrast topograms from a large surface of the single crystals. To do this a lead screen with apertures was placed in the path of the primary divergent x-ray beam. The first-order parameters of the substructure were calculated from the contrast topograms thus obtained[18,21]—the dimensions of the macroblocks (in the direction transverse to the growth of the crystals), and their angular disorientation, from the width of the dark and light bands (δ), which constitute the boundaries between the macroblocks.[18,21] From the crystallographic studies information was obtained on the second-order substructure—the dimensions of the microblocks, the distribution of their subboundaries, and the density of etch pits. The Laue method was used to determine the orientation of single crystals of aluminum, tin, and lead with respect to their growth direction when crystallized under irradiation and under ordinary conditions (90, 39, and 36 crystals, respectively). Also the variation of the first- and second-order substructures of the single crystals along the ingots was studied.

The calculations showed good agreement of the characteristics of the substructures of single crystals grown under identical conditions. Therefore we discuss the results averaged over these specimens. It was established that the substructure of aluminum single crystals grown under different conditions is different. In completely unirradiated single crystals the size of the macroblocks L_I amounts to about 2 mm. Their mean angles of disorientation φ are 17′ (Table III). As the unirradiated single crystals grow, the disorien-

TABLE III. Averaged characteristics of the structure of aluminum single crystals.

Treatment	n_d/cm^2	φ_{av}, min	φ_{max}, min	L_I, mm	L_{II}, mm
Unirradiated	$5\cdot 10^5$	17	30	2	0,45
Irradiated	$3\cdot 10^4$	4	10	4,5	1,1

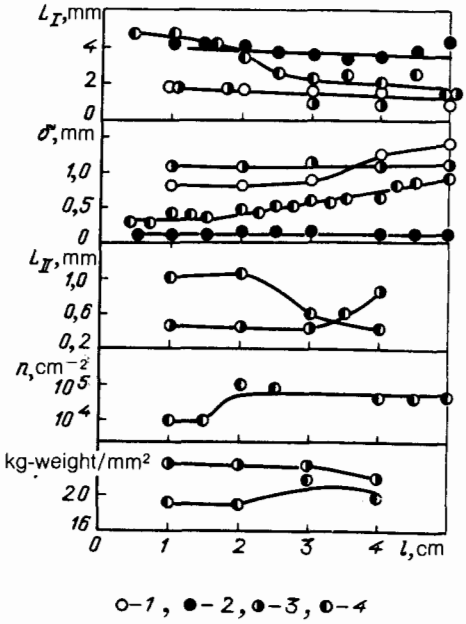

FIG. 2. Characteristics of the substructure of single crystals of aluminum grown under different irradiation conditions: without irradiation (*1*), under irradiation (*2*), with γ-irradiation introduced during the process of growth of the single crystals (*3*), and removed during the process (*4*).

tations of the macroblocks increase almost twofold, as can be seen from the variation of the quantity δ (Fig. 2). The density of etch pits (n) in unirradiated single crystals was $(2\text{–}8)\times 10^5/\text{cm}^2$. The dimensions of the microblocks (L_{II}) determined from the number of subboundaries in the direction transverse to the growth of the crystals lay in the range 0.4–0.5 mm (see Table III). These quantitative characteristics are close to those obtained for single crystals of aluminum grown under ordinary conditions by other authors (e.g., Ref. 22).

In completely irradiated single crystals one observes coarser macroblocks (4–5 mm) and microblocks (0.9–1.2 mm) and considerably smaller disorientation angles (4–10′) than in unirradiated specimens. It was also established that the density of etch pits is almost ten times smaller in irradiated than in unirradiated specimens (see Table III and Fig. 2). These quantitative characteristics indicate the formation in the γ-ray field of a more perfect substructure of aluminum single crystals than under ordinary conditions.

If aluminum single crystals nucleated in a γ-ray field continue to grow in its presence, then their substructure is maintained throughout the ingot (Fig. 2). A different pattern is observed when single crystals are formed in a γ-ray field and then continue to grow under ordinary conditions. In this case the substructure formed at the beginning of the ingot changes: the disorientation of the macroblocks (defined by the quantity δ) increases, and their dimensions decrease, while the density of etch pits increases about tenfold (see Fig. 2). It was found in observation under the microscope that in the initial part of single crystals formed in a γ-ray field the number of subboundaries is smaller than in the unirradiated part (Fig. 3). The pattern of distribution of the subboundaries implies that the dimensions of the microblocks decrease from 1 to 0.5 mm after removal of the γ-ray source from the irradiation zone (see Fig. 2). Hence the second-order substructure is impaired. The characteristics of the substructure become close to those characteristic of single crystals nucleated and grown under ordinary conditions. The observed substructural changes are accompanied by an increase in the microhardness by 2 kg-weight/mm^2 (see Fig. 2), which is equal to the difference in microhardness between irradiated and unirradiated single crystals.[20,21]

The introduction of the γ-ray field during the process of growth of single crystals that had been formed under ordinary conditions does not lead to substantial changes in the first-order substructure. Only certain defects of the second-order substructure are removed. Thus, in the irradiated part of the single crystals the number of subboundaries in the direction transverse to the growth of the crystals diminishes, while the dimensions of the microblocks increase from 0.4 to 0.8 mm (see Fig. 2).

Thus it was established from the obtained data that a more perfect substructure is formed in aluminum single crystals crystallized in a flux of γ-ray quanta than under ordinary crystallization conditions. As is implied by Table III, in irradiated specimens the dimensions of the macro- and microblocks are twofold larger, while their disorientation angles are 3–4 times smaller, and the density of dislocations (n_d) is about 10 times smaller than in unirradiated specimens (see Table III).

Analysis of the obtained results indicates that γ-irradiation acts primarily on the nucleation of the crystals. There-

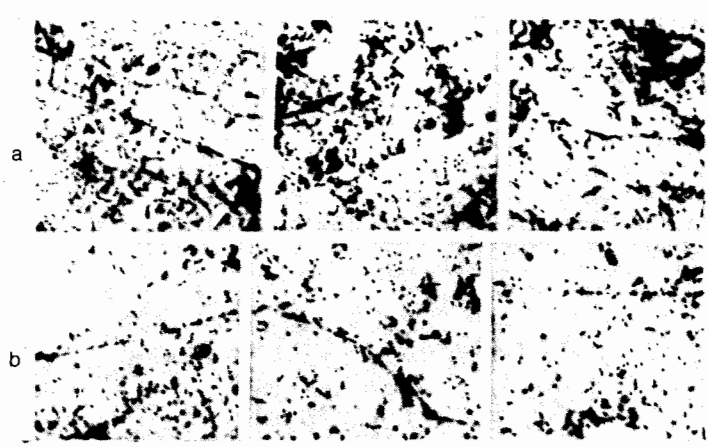

FIG. 3. Distribution of subboundaries in unirradiated (a) and irradiated (b) parts of a single crystal of aluminum.

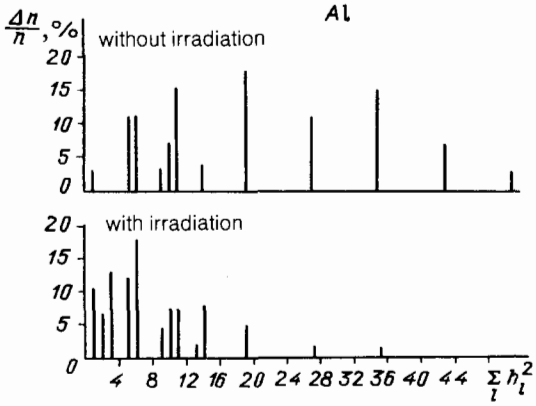

FIG. 4. Dependence of the relative number of single crystals of aluminum ($\Delta n/n$) on the sum of squares of the indices (hkl) in the direction of growth.

fore no influences of irradiation on the structure of aluminum single crystals grown from seed specimens has been observed.[21] The conclusion on the prevailing role of γ-irradiation in the nucleation of crystals is confirmed by its influence on their spatial orientation. It was established that in a flux of γ-quanta single crystals of metals mainly grow in the close-packed crystallographic directions,[21] while under ordinary conditions all directions of growth are equally probable (Fig. 4). These data indicate the possibility of decreasing the surface energy at the phase boundary between melt and crystal.

The observed influence of γ-irradiation on the formation of the structure of metallic single crystals is a consequence of its action on the different stages and parameters of the crystallization of metals. The hypothesis has been advanced[9,20,21] that γ-irradiation can lead to dissociation of clusters owing to acceleration of the diffusion of atoms[23] and absorption of the radiation energy. As is known, such aggregates of atoms having short-range order are formed in metallic melts near the crystallization temperature T_0.[24–26] The existence of clusters in metallic melts is confirmed by the literature data presented in Ref. 25 on the anomalous increase in viscosity in the stage of precrystallization of metals. The conclusion was drawn,[9,20,21] upon taking account of the hypothesis of dissociation of clusters in a flux of γ-quanta, that a more perfect substructure of single crystals of metals is formed under these conditions because of the attachment to the moving crystallization front of individual atoms (atomwise attachment), rather than of whole groups of them (clusters), which would violate the correct structure of the crystal. Subsequently the hypothesis of the dissociation of clusters was confirmed experimentally in Ref. 26, which showed that, even under the action of x-irradiation, the dimensions of the ordered regions (clusters) in liquid metals decreases. The possibility of enhancement under γ-irradiation of the probability of atomwise attachment in the formation of crystals is confirmed by the results of Ref. 27, where the process of epitaxy of layers of germanium and gallium arsenide on a substrate in a flux of γ-quanta was studied. Irradiation led to formation of layers with a greater homogeneity of structure, which is useful in practice.

The data on the influence of γ- and x-irradiation on the crystallization of metals.[9,18–21] and ionic crystals[8–10] agree with the results of the influence of electron irradiation on the crystallization of amorphous Fe-C films[28] and the recrystallization of nickel.[29] It was shown[28] that amorphous Fe-C films crystallized under the action of an electron beam to form carbide grains of dimensions 2×10^{-5} cm uniformly distributed in the field of the matrix. The crystallization was shown to occur only at the site of entry of the beam, while neighboring regions remained amorphous. A very fast reaction of the rate of growth of the carbide grains to altered beam intensity was noted. These data confirm the fact that radiation acts specifically on the nucleation of crystals.

The recrystallization has been studied[29] of deformed nickel directly under electron irradiation of intensity $\approx10^{18}$ el/(cm^2·s) at elevated temperature (500 °C). It was established that recrystallization occurred only in the irradiated volume, in which the density of dislocations proved to be smaller than in the surrounding, unirradiated volume (10^{10} and 10^{11}/cm^2, respectively). The anisotropic character of the growth of grains recrystallized under irradiation was noted,[29] namely, preferentially in the closest-packed {100} direction, which agrees with the data of Refs. 18, 20, and 21 (Fig. 4). The authors explain the results[29] by migration of grain boundaries under irradiation, as was found also in Ref. 17. However, this phenomenon was associated[29] not with radiation defects themselves, as in Ref. 17, but with the additional energy accumulated as a result of the appearance of the defects and the heightened rate of diffusion of atoms upon irradiation.

One can note many common features upon comparing the results of study of the influence of nuclear irradiation on the formation of metallic and nonmetallic crystals. The results obtained on metals have confirmed, and in a number of cases have supplemented and extended the existing information on the role of irradiation in processes of crystallization of solids. Effects such as the improvement of the substructure of crystals, their growth in definite crystallographic directions, and the enlargement of the dimensions of crystals are desirable in practice. As the presented data have shown, considerably larger irradiation intensities are required to manifest in metals the effects that have been observed in ionic crystals ($\sim10^2$ R/s (Refs. 18–21) and ~10 R/s (Refs. 8–10), respectively).

3. THE INFLUENCE OF γ-IRRADIATION ON THE DISTRIBUTION OF IMPURITIES IN SINGLE CRYSTALS OF ALUMINUM AND ON THE SEGREGATION COEFFICIENT

3.1. Study of Al-Cu alloys having a segregation coefficient less than unity

Analysis of the obtained results[9,20,21] has led to the conclusion that impurities with a segregation coefficient $K<1$ can be more effectively expelled under γ-irradiation by the moving crystallization front toward the end of growing single crystals or to the grain boundaries,[11] which facilitates improvement of the substructure.

The elucidation of the question of the influence of γ-irradiation on the distribution of impurities in single crystals furnishes additional information on the mechanism of improvement of the substructure of crystals nucleated under radiation conditions. To elucidate this, the substructure and impurity distribution were studied[9,21,30] in aluminum single crystals doped with copper (0.14 and 0.18 weight percent Cu). These alloys are homogeneous α-solid solutions of Cu

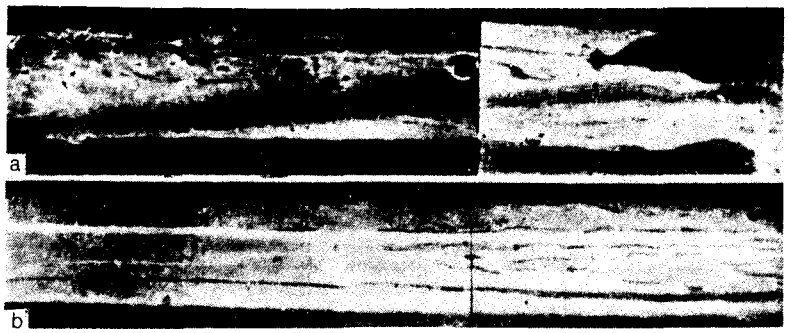

FIG. 5. Topograms of Al-Cu single crystals–unirradiated (a) and irradiated (b).

in Al.[31] Specimens of dimensions $10\times10\times60$ mm^3 were crystallized in a flux of γ-ray quanta of intensity $(1.5–5)\times10^{12}$ kW/cm^2, and without irradiation under analogous thermal conditions, according to the method described in Sec. 2. It was shown that an improvement of the substructure also occurs in these single crystals upon crystallization under irradiation, while this effect is manifested more strongly than in pure aluminum. Thus, in the unirradiated Al-Cu ingots extra crystals lying along the edges exist in addition to the main single crystal. The irradiated ingots constitute a unitary single crystal.

Topograms of the specimens were obtained by the method described in Sec. 2. It was established that the irradiated Al-Cu single crystals have a more perfect first-order substructure than do the unirradiated ones. It was established that the disorientation angles of the macroblocks in the irradiated single crystals are fourfold smaller than in the unirradiated ones ($\varphi = 30'$ and $120'$, respectively). It was also noted that the macroblocks in the irradiated single crystals lie along the direction of growth. In the unirradiated specimens a rotation of the macroblocks was noted with respect to the direction of growth of the single crystal (Fig. 5).

As is known, one of the properties sensitive to the concentration of copper in aluminum is the microhardness H_v.[31] The microhardness of a pure single crystal of aluminum is 28.5 kg-weight/mm^2, while that for alloys with 0.14 weight percent Cu is 35.5, and with 0.18 weight percent Cu is 38 kg-weight/mm^2. The values were used to determine the concentration of Cu in Al from the microhardness along the ingots (Figs. 6 and 7). The distribution of copper along the single crystals was also determined from the change in their density ρ.[9,21,30] The value of ρ of individual parts of the cut-apart ingots was determined by hydrostatic weighing with an accuracy of 0.02–0.03%.[21] We see from Figs. 6 and 7 that the concentration of copper (C_g) proved to be smaller in the main fraction of the volume of the irradiated specimens than in the unirradiated ones. Moreover, it was shown that the concentration profile varied with increasing intensity of irradiation (see Figs. 6 and 7). These data imply that under γ-irradiation copper, having a segregation coefficient less than unity, is more effectively driven toward the end of the single crystals.

The following relationship:[32]

$$C_g = K_{\text{eff}}\cdot C_0(1-g)^{K_{\text{eff}}-1}, \tag{1}$$

where C_0 is the concentration of copper in the original specimens, was used to calculate the effective segregation coefficients (K_{eff}) from the data of the concentration profiles. Figure 8 shows the dependence of K_{eff} on the irradiation intensity. As J is increased up to 5×10^{12} kW/cm^2, the value of K_{eff} approaches the equilibrium value (0.17).[32] This indicates that the crystallization conditions approach equilibrium under irradiation. Usually this is not easily realized in practice owing to the low rate of remixing of the impurity in the melt.[33]

The observed distribution of the Cu impurity in unirradiated Al-Cu specimens corresponds to the condition of normal crystallization, in which partial remixing in the liquid phase occurs,[33] which is described by the dashed curves drawn through the experimental points (see Fig. 6). As the intensity of γ-irradiation is increased, a transition occurs from partial to complete remixing of the impurity in the

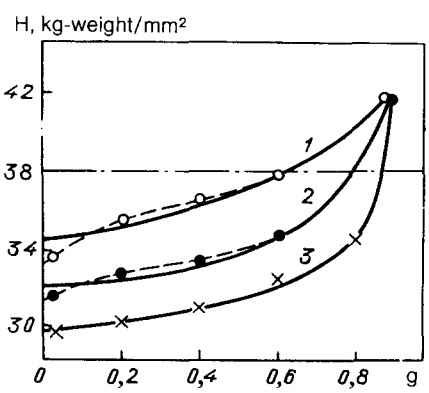

FIG. 6. Variation of the microhardness of Al-Cu single crystals along the ingots: without irradiation (*1*), with $J=2\times10^{12}$ kW/cm^2 (*2*), and 5×10^{12} kW/cm^2 (*3*). The dashed curves correspond to the experimental data, and the solid curves are averaged; g is the fractional length of the ingot.

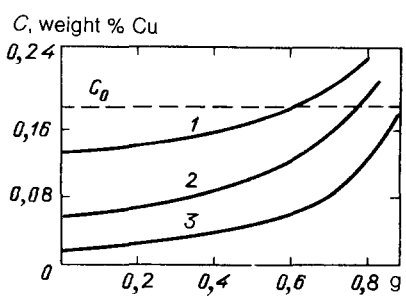

FIG. 7. Concentration profiles of the Cu distribution along Al-Cu single crystals: without irradiation (*1*), with $J=2.4\times10^{12}$ kW/cm^2 (*2*), and 5×10^{12} kW/cm^2 (*3*).

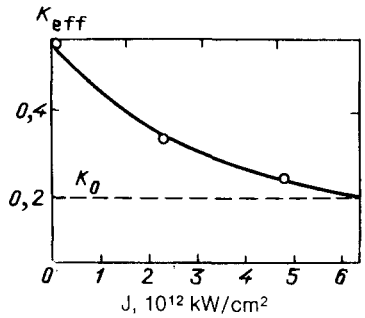

FIG. 8. Dependence of the effective segregation coefficient of Cu K_{eff} on J.

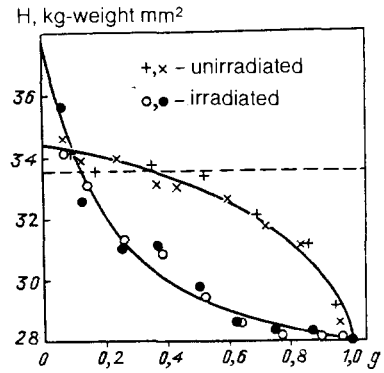

FIG. 9. Variation of the microhardness of Al-Cr single crystals along the ingot.

melt. This implies a result important in practice—the effective purification of aluminum from copper that one observes upon irradiation (5×10^{12} kW/cm^2) during a single crystallization is attained under ordinary conditions only after 10–12 cycles of recrystallization.[33]

The value of ζ/D was calculated by using the relationship associating the effective segregation coefficient K_{eff} with the other crystallization parameters:[32]

$$K_{eff} = \left[K_0/(1-K_0)\exp(-\zeta/DV)\right] + K_0. \qquad (2)$$

Here ζ is the thickness of the diffusion layer at the liquid-crystal phase boundary, D is the diffusion coefficient of the impurities in the melt, and V is the rate of growth of the crystals. Starting with values of K_{eff} equal to 0.6 for unirradiated single crystals, and 0.2 for irradiated, at $J=5\times10^{12}$ kW/cm^2 and taking into account the fact that $K_0=0.17$, while $V=10^{-3}$ cm/s, it was estimated that $(\zeta/D)_{unirr}=10^3$ s/cm, while $(\zeta/D)_{irr}=10^2$ s/cm. Thus the value of (ζ/D) is decreased by an order of magnitude upon irradiation. This phenomenon indicates two possibilities—increasing the rate of mixing of the atoms in the melt and/or decreasing the thickness of the diffusion layer ζ. Both these phenomena should facilitate the improvement of the structure of crystals formed in a γ-ray field. Actually the enhanced rate of mixing of the atoms upon irradiation confirms the assumption[9,20,21] of enhanced probability of atomwise attachment to the moving crystallization front owing to the dissociation of clusters,[34] which is the reason for the improved perfection of their structure in the flux of γ-ray quanta.[9,19] The latter depends also on the thickness of the diffusion layer at the crystal-melt phase boundary.[35] As is known, the smaller it is, the more perfect is the structure of the crystals.[35] It does not seem possible to estimate the change upon irradiation of each parameter D and ζ separately on the basis of the existing experiments. We can only conclude that the parameter D increases by no more than an order of magnitude upon irradiation, which agrees with the quantitative estimates obtained in Refs. 36 and 37 of the change in mobility of atoms upon γ-irradiation of the order of $\approx 2\times10^{12}$ kW/cm^2.

3.2. Study of Al-Cr alloys having a segregation coefficient greater than unity

It was of interest to elucidate the question of the possibility of an influence of γ-irradiation on the equilibrium segregation coefficient. The influence of nuclear radiation on this parameter had not heretofore been studied. As is known,[32] a value $K_0>1$ depends weakly on the external conditions of crystallization, i.e., essentially we have $K_{eff}=K_0$. In this case it is easier to find an influence of γ-irradiation on K_0 if it really occurs. Hence the Al-Cr system was studied, having $K_0=1.4$.[32] The initial concentration of chromium in Al was 0.1 weight percent. The crystallization of Al-Cr ingots of dimensions $5\times5\times80$ mm^3 was performed by the method described above in graphite crucibles in a flux of γ-ray quanta of intensity 3.5×10^{12} kW/cm^2 and under the same thermal conditions without irradiation. The distribution of chromium along the ingots after crystallization was determined by measuring the microhardness (Fig. 9). As is known, within the solid-solution range the microhardness of alloys of aluminum with impurities is proportional to the concentration of the dopant component.[31] Upon using the microhardness data (Fig. 9) of the Al-Cr alloy in the original state and that of a pure aluminum single crystal (33.2 and 28.0 kg-weight/mm^2, respectively) and taking into account the linear dependence of H_v on C,[31] concentration profiles of the distribution of chromium along the length of the ingot were drawn (Fig. 10). We see from them that, in the initial part of the single crystals grown under ordinary conditions, C_g is greater than the original C_0. With increasing g one observes a decrease in the concentration of chromium atoms C_g, slow at first and then sharper. Such a concentration profile (curve *1* in Fig. 10) agrees with the literature data for $K>1$.[32]

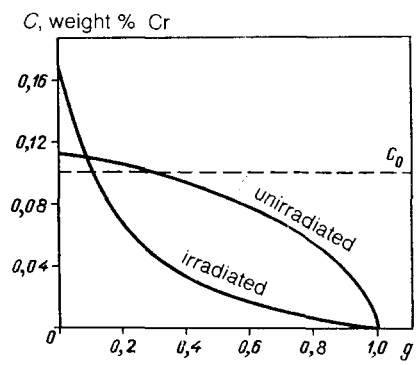

FIG. 10. Concentration profile of the Cr distribution along the ingots.

The character of the variation of C_g with g for single crystals grown under irradiation differs substantially from the ordinary case. In the initial region of the ingot one observes a sharp decline in the value of C_g. Even for $g = 0.15$ the content of chromium in the aluminum becomes less than the original value. With increasing g the value of C_g smoothly approaches zero at the end of the ingot. Thus $\approx 85\%$ of the volume of the single crystal contains less chromium than in the original state. The areas bounded by the curves and the coordinate axes give the overall amount of chromium in the ingot before crystallization (M_0), and after crystallization (M_g). Since under ordinary conditions $M_g/M_0 = 0.91$, i.e., is close to unity, we can apply the usual relationship (1) to calculate K. For an Al-Cr single crystal grown under ordinary conditions we have $K = 1.5$, i.e., close to the known value 1.4.[32] This shows that the use of microhardness data to determine the concentration profile is warranted. The ratio M_g/M_0 after crystallization under γ-irradiation is 0.4. Consequently one cannot use Eq. (1) to calculate the segregation coefficient K_γ.The considerable decrease in the amount of chromium, even in the initial regions of the ingots crystallized under irradiation, indicates that it has been lost from the specimen. This can be caused by removal of chromium atoms onto the active substrate, which apparently is the graphite crucible, which scavenges this admixture owing to the strong interaction of chromium and carbon atoms.[15,16] This phenomenon leads to a more effective purification of the aluminum from chromium under irradiation conditions.[21,38] This process can be described by diffusion from a body of finite dimensions having restrictive boundaries. To estimate the values of the parameter K_γ under irradiation, Fick's diffusion equation was solved with account taken of the residual concentration of chromium atoms in the melt at any instant of time $t > 0$.[21,38] Here two processes were considered—redistribution, upon crystallization from the melt in the flux of γ-ray quanta, of chromium atoms along the ingot, and their removal by the scavenging substrate. The diffusion coefficients D_γ of chromium in liquid aluminum and the segregation coefficients in the γ-ray field were obtained for different values of g. The mean value of D_γ, as was shown in Refs. 21 and 38, is 0.5×10^{-5} cm^2/s, which is close to the value of the diffusion constant of impurities in liquid metals $\approx (0.5\text{–}2) \times 10^{-5}$ cm^2/s. This indicates the correctness of both the assumptions made in calculating D_γ and K_γ and of the calculation itself. The value of K_γ corresponding to the equilibrium segregation coefficient of chromium in aluminum in a flux of γ-ray quanta amounts to 2 ± 0.2 on the average. Thus, under irradiation the equilibrium segregation coefficient K_0 in Al-Cr is increased by a factor of 1.5 as compared with K_0 under ordinary conditions.

As is known, the parameter K_0 is associated with the other parameters of crystals by the relationships[30,32]

$$K_0 \sim \exp(E_N - E_j)/kT, \quad K_0 = C_S/C_L \,. \tag{3}$$

Here k is the Boltzmann constant, E_N and E_j are the binding energies of the main and of the impurity atoms, and C_S and C_L are the limiting solubilities in the solid and liquid phases, respectively. The alteration of K_0 in a flux of γ-ray quanta indicates an influence of irradiation on the binding forces of the atoms in the lattice (E_b) and on the limiting solubility of the impurity. This agrees with the conclusion drawn in Ref. 37 based on experimental data that the parameter E_b can be altered under irradiation. As early as 1971, the hypothesis was advanced[39] of a change in the limiting solubility of impurities under nuclear irradiation. The relationship (6) and a number of theoretical and experimental studies (e.g., Ref. 40) have subsequently confirmed this hypothesis.

4. THE INFLUENCE OF γ-IRRADIATION ON THE EQUILIBRIUM CRYSTALLIZATION TEMPERATURE OF METALS

The data of Sec. 3 imply that the rate of mixing of atoms is increased in a flux of γ-ray quanta, which leads to dissociation of clusters. That means, the structure of the melt is altered to favor disorder. Therefore, according to Ref. 2, one could assume that upon γ-irradiation the temperature T_0 of the melt-crystal transition is lowered. This phenomenon had been observed earlier in the self-irradiation of the salts Na_2SO_4 and Na_2WO_4.[41] It was found that their melting points are decreased by 4–8 °C, depending on the degree of radioactivity of the substances.

To elucidate the question of whether an analogous phenomenon occurs in metals, the influence was studied of γ-irradiation on the solid-liquid phase transition temperature T_0 in lithium, aluminum, and tin. To do this, they were melted (and crystallized) in a flux of γ-ray quanta.[9,20,21,25,39]

The tin and aluminum were melted in graphite crucibles, and the lithium in a hermetically sealed container made of heat-resistant steel with a wall thickness of 0.1 mm. The crystallization and melting of each metal were performed repeatedly, alternating these processes under irradiation and in its absence. Also the dependence of the melting point of aluminum on the intensity of γ-irradiation was studied. To do this, the number of ^{60}Co sources in the irradiation zone was varied. The maximum intensity of the γ-ray field was 3.2×10^{12} kW/cm^2. The temperature of the specimens was measured with a differential platinum-rhodium thermocouple (Pt–Pt + 10% Rh) with a diameter of conductors of 0.1 mm. The temperature readings in crystallization and melting of the specimens were recorded remotely with a low-resistance dc potentiometer every half-minute during the entire process of melting (or crystallization). The method allowed measuring the temperature of the specimens with an accuracy of 0.05 °C. We see from the thermograms (time course of the temperature, Fig. 11) that one observes good reproducibility of the results of measuring the temperature under identical conditions, independently of the order of performing the experiments.

It was established that a change in the temperature T_0 occurs upon irradiating lithium and aluminum, this effect depending on the intensity of irradiation.[25] For tin a tendency was noted toward decrease in T_0 under irradiation. However, ΔT_0 lay within the limits of error of the measurements and amounted to 0.05 °C.[25] The dependence of the value of ΔT_m for aluminum on the intensity of γ-irradiation is practically linear (Fig. 12). The deviation of certain points from a straight line is explained by the nonidentical intensities of the individual sources.

The effects found in different metals were compared quantitatively. Here account was taken of the fact that a single atom of each of them absorbs a different radiation energy owing to the different densities of the substances be-

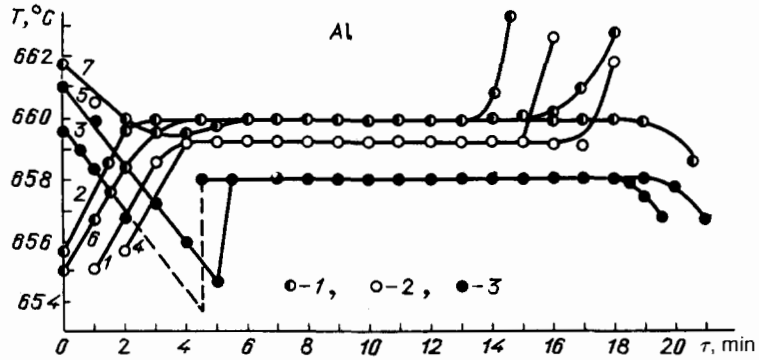

FIG. 11. Thermograms of aluminum: melting and crystallization without irradiation (*1*), melting under irradiation (*3*), and crystallization under irradiation (*2*) (the numbers denote the order of performance of the experiments).

ing studied. Therefore the variation of ΔT_0 per unit interval of absorbed energy was estimated, i.e., the quantity $\Delta T_{\rm eff}$:

$$\Delta T_{\rm eff} = \Delta T/\bar{\varepsilon}, \tag{4}$$

Here $\bar{\varepsilon}$ is the mean energy of γ-ray quanta absorbed by a single atom of the substance per unit time:[43]

$$\bar{\varepsilon} = J_0\varepsilon_\gamma(1 - \exp(-\mu d))/N_0 v. \tag{5}$$

Here ε_γ is the energy of the γ-ray quanta, J_0 is the number of γ-ray quanta incident per unit area per second, v is the irradiated volume, μ is the linear absorption coefficient, and N_0 is the number of atoms per unit volume. Under the same experimental conditions, if we take v and d to be the same in different specimens and take account of (4) as well as the values of N_0 and μ ($N_{0,\rm Li} = 0.46\times 10^{23}$, $N_{0,\rm Al} = 0.6\times 10^{23}$, $N_{0,\rm Sn} = 0.5\times 10^{23}$, $\mu_{\rm Li} = 0.03/{\rm cm}$, $\mu_{\rm Al} = 0.146/{\rm cm}$, $\mu_{\rm Sn} = 0.32/{\rm cm}$), we obtain[21,25]

$$\bar{\varepsilon}_{\rm Al}/\bar{\varepsilon}_{\rm Li} \approx 3, \quad \bar{\varepsilon}_{\rm Sn}/\bar{\varepsilon}_{\rm Li} \approx 9, \tag{6}$$

$$\Delta T_{\rm eff\,Li}/\Delta T_{\rm eff\,Al} \approx 3, \tag{7}$$

$$\Delta T_{\rm eff\,Li}/\Delta T_{\rm eff\,Sn} \approx 50. \tag{8}$$

Thus the experimental results show that the quantity $\Delta T_{\rm eff}$ declines with increasing atomic weight of the metal ($A_{\rm at}$). The effectiveness of the influence of γ-irradiation on the parameter T_0 depends on the parameters that characterize the metallic melt near the crystallization (or melting) temperature. The fractions of clusters (Φ) in the volume of the melt, their energies of formation ($E_{\rm f}$), and the number of atoms per cluster (f) in aluminum and lithium were determined[25] on the basis of the data of Ref. 24. These quantities respectively amount to 10%, 8 eV, and 100 atoms for aluminum, and 1.5%, 0.7 eV, and 20 atoms for lithium.[25] Comparison of these parameters for these elements shows that the probability of dissociation of clusters upon irradiation in lithium should be larger than in aluminum. This is one of the reasons for the stronger effect of depressing T_0 in lithium as compared with aluminum.

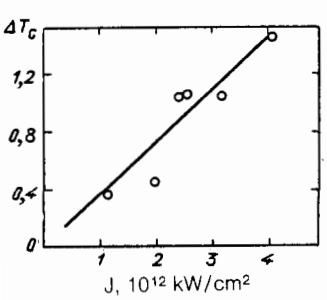

FIG. 12. Dependence of the depression of the melting point of aluminum ΔT_c on the intensity of irradiation J.

The hysteresis of the melting and crystallization temperatures observed in lithium and aluminum[21,25,39] confirms the hypothesis of Secs. 2 and 3 that γ-irradiation exerts an influence on the structure of the liquid metal at the precrystallization stage. Apparently a melt subjected to continuous irradiation during crystallization proves to be a liquid of a different type from the melt formed upon melting of the crystals. This has the result that $\Delta T_{\rm cr} > \Delta T_{\rm m}$. We should note that the influence of γ-irradiation on the structure of the liquid metal has no aftereffects. The melting temperature of the metal crystallized under irradiation is determined only by the presence or absence of irradiation at the instant of melting, and does not depend on the previous treatment (see Fig. 11).

The effect of decrease of the crystal-melt transition temperature upon γ-irradiation is correlated with the phenomenon of improvement of the substructure of single crystals of metals under these conditions. Actually, the depression of the crystallization temperature of metals under irradiation indicates, as we see from the model proposed by Jackson,[3] a decrease in the atomic roughness at the crystal-melt phase boundary. Here the following relationship holds between the change in free energy and the number of filled nodes at the boundary:[3]

$$\Delta F/N_0 kT_0 = \alpha x(1 - x) + x\ln x + (1 - x)\ln(1 - x), \tag{9}$$

Here $\alpha = Q\psi/RT_0$, $x = N_{\rm a}/N_0$, $N_{\rm a}$ is the number of occupied nodes, and N_0 is the total number of nodes at the surface of the crystal-melt surface, Q is the latent heat of melting, and ψ is a factor that depends on the structure of the metals and is equal to 2 for them.[3] For metals at the equilibrium melting temperature, the minimum free energy corresponds to surfaces at which half the nodes are occupied (such surfaces are considered rough).[3]

The relationship (9) implies that a change in the roughness at the phase boundary occurs upon changing T_0. The estimate obtained from (9)[20,21] showed that a decrease in T_0 by 1 °C increases the number of nodes occupied at the surface ($\approx 0.5\times 10^{16}/{\rm cm}^2$) by about $10^{13}/{\rm cm}^2$, which decreases its roughness. The degree of roughness is associated with the perfection of the substructure of the single crystals. The smoother the surface is at the liquid-solid phase boundary,

the more perfect the substructure of the growing crystal will be.[3] This is confirmed by experiment.[43] It was established that salol, when crystallized under x-irradiation (which improves the structure of this substance), actually forms a smoother phase-boundary surface than usual.

The model of Jackson also implies that, with decreasing crystallization temperature, the probability increases that the liquid-crystal phase boundary in aluminum will be close-packed planes. This is experimentally confirmed by the fact that the growth of metal crystals in a flux of γ-ray quanta occurs preferentially in the close-packed directions (Fig. 4).[9,20,21]

One cannot explain the effect of lowering the parameter T_0 in metals under the action of γ-irradiation by the formation of point defects, analogously to Ref. 44. In the situations discussed in the review of Ref. 44 according to Ref. 45, a decrease in the free energy, and this implies that of T_0, can occur owing to formation of a relatively large number of direct displacements of atoms from the nodes ($N_d \sim 10^{-3}$/atom). Upon γ-irradiation of intensity $\approx 2\times 10^{12}$ kW/cm^2, the magnitude of N_d amounts to something of the order of 10^{-10}/atom over the time of performing the experiment (~ 60 min). This cannot appreciably influence the change in the free energy upon melting (or crystallization); this means that it cannot alter the value of T_0. The conclusion that the effect of decreasing the parameter T_0 does not involve point defects is confirmed by the experimental data. Actually we have $N_d \sim \sigma_d$ (σ_d is the transverse scattering cross section of the process of displacement of atoms from nodes).[5-7] Since σ_d is proportional to Z, the atomic number of the metal, we have $N_d \sim A_{at}$. This means that the quantity ΔT_{eff} must be proportional to A_{at}. However, the relationships (7) and (8), which were derived from the experimental data, imply that ΔT_{eff} does not increase with increasing atomic weight of the material, but conversely declines. Thus the reason for the depression of the parameter T_0 upon γ-irradiation is not the formation of point defects, but other mechanisms. In γ-irradiation a large part of the absorbed energy is transferred to the electronic subsystem.[5-7] Here, as was shown in Ref. 46, the contribution of free electrons to the effect of lowering the equilibrium temperatures in a flux of γ-ray quanta is very small. Apparently the ionization of the inner electron shells upon γ-irradiation makes an appreciable contribution to the change in the parameter T_0. The lifetime of such an ionized state amounts to no less than 10^{-13} s,[47] which is comparable with the period of the thermal vibrations of atoms in the lattice. At present there are no theoretical estimates of the contribution of ionization of the inner electron shells to the change in various properties of metal crystals upon γ-irradiation. However, the experimental data on the lowering of the activation energy of phase transitions and on the increase in mobility of atoms[37] imply that one must not neglect this contribution.

CONCLUSION

A change occurs during crystallization in a flux of γ-ray quanta in such parameters as the equilibrium transition temperatures, the mobility of atoms at the melt-crystal phase boundary, and the segregation coefficients of impurities. These phenomena lead to the complex changes that are observed in the crystallization of binary and multicomponent alloys in a flux of γ-ray quanta.

We can conclude from analyzing the presented data that the observed phenomena—improvement in a flux of γ-ray quanta of the structure of metallic single crystals, enhanced purification from impurities, and decrease in the equilibrium temperature T_0—are caused primarily by the dissociation of clusters. This phenomenon can occur by the mechanism of Ref. 48, where it was shown that in grazing collisions of electrons (Compton events, in γ-irradiation) an energy 0.1–1 eV/atom is imparted to the atoms, which suffices to displace them to adjacent unoccupied positions. Here the scattering cross section is greater[48] by a factor of 10^3–10^4 than in direct collisions of electrons with atoms. Therefore the number of atoms displaced by the mechanism of Ref. 48 is considerably larger than N_d. The probability of such an inelastic process increases in favorable situations—when the lattice contains many vacant sites and/or when the binding energy of the atoms in clusters is 10^{-1}–10^{-2} eV/atom. Hence the energy transferred in grazing collisions of Compton electrons with the atoms suffices to displace them from the surface of the cluster (which contains, according to Ref. 25, no more than 100 atoms) and/or to disorder them by atomic rearrangements.[37] Reference 49 studied the realization of these processes in precritical nuclei, even in the case of irradiation that does not form cascades.

As is known,[25,37,50,51] γ-irradiation substantially affects the phase and structural stability of solid solutions. The action of irradiation is especially marked near the equilibrium temperatures of phase transitions in processes characterized by phase boundaries. The dissipation of the energy absorbed in γ-irradiation by the electronic subsystem occurs, according to Refs. 52 and 53, mainly at the boundaries of the phase and structural components and at various structural inhomogeneities such as, e.g., clusters in melts. This enhances the probability of their dissociation. Hence even less intense x-rays break up these aggregates.[26]

A comparison of the action of ionizing radiation with the influence of other external agents on the process of crystallization leads to the following conclusion. Ionizing radiation is an effective factor of action on the formation of metallic crystals. By facilitating the increased probability of atomwise attachment to the growing face of the crystal, it leads to results useful in practice—increased perfection and dimensions of crystals, more effective purification of them from impurities, and elimination of random defects at the crystal-melt phase boundary.[34,38] Ionizing radiation leads to improvement of the characteristics of the substructures of single crystals of metals without introducing additional defects (as ultrasound does) or contamination (as in mechanical mixing of the melt). The method of improving single crystals of metals by using ionizing radiation can be widely employed in practice.[34,38]

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