
**EFFECT OF ELECTRON AND γ -RADIATIONS
ON THE STRUCTURE OF Fe–Si–B AMORPHOUS ALLOYS****V.YU. POVARCHUK, V.B. NEIMASH, A.M. KRAITCHINSKII,
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By X-ray diffraction and thermal magnetic methods, we have studied the effect of electron ($E = 1$ MeV) and γ -radiations ($E = 1.17$ and 1.33 MeV) on structural changes in amorphous alloys of the Fe–Si–B system. The dependences of the height of the first maximum of the structural factor $i(s_1)$, the Curie temperature T_C , and the crystallization temperature T_X on the dose are investigated. It is experimentally shown that the mechanisms of the effect of electron and γ -radiations on the structure of amorphous metallic alloys of the Fe–Si–B system are identical. It is established that the doping of these alloys with Ni and Mo atoms leads to a decrease of the sensitivity of their characteristics to radiation. The sign and value of the radiation-induced changes of the Curie temperature and the crystallization temperature of undoped alloys significantly depend on the ratio of the concentrations of Si and B in their composition. We have proposed two mechanisms of the effect of radiation on the structure of amorphous metallic alloys and a qualitative model of the effect of radiation-stimulated diffusion of Si and B on T_C and T_X of amorphous alloys of the Fe–Si–B system.

1. Introduction

Amorphous metallic alloys (AMAs) possess unique physical properties which define their intense introduction into various branches of science and technique. The spectrum of their use can spread also to the case where alloys undergo the action of radiation. However, the efficient use of such materials under conditions of ionizing radiation and the control over their properties with the help of radiation treatment become possible if the mechanisms of the effect of radiation on the structure of AMAs are established. Thus, ionizing radiation can serve

as a tool in studying the physics of disordered systems.

AMAs are complicated scientific objects for physical modeling, which is testified by the absence of a single structural model of such systems. In addition, the difficulties of the creation of scientific models of the effect of radiation on the structure, thermal stability, and the processes of crystallization of AMAs are related to the scarcity and ambiguity of the available data in the literature.

The irradiation of AMAs with electrons causes the disordering of their structure [1–4]. It is worth noting that the results of the effect of this kind of radiation on the processes of crystallization and the thermal stability of AMAs are ambiguous. It is considered that radiation induces a deceleration [3, 4] of the crystallization of AMAs, which is related to the fracture of clusters with short-range order (SO) inherent to nuclei of the crystal phase. However, it was shown in [5] that the electron radiation ($E = 0.4$ and 1 MeV) can induce either acceleration or deceleration of the crystallization of AMAs. The direction of the process is determined by the form of a combination of a pair “metal (Fe, Ni) – nonmetal (P, C, B, Si)”.

The data [1, 6–14] which concern the effect of γ -radiation on the structure and the processes of crystallization in AMAs are also ambiguous. The analysis of the results in [6–9] testifies that γ radiation ($E = 1.2$ MeV, $F = 10^9$ rad) causes the disordering of a structure of AMAs, but the results in [10] testify to the ordering. As for the data on the effect of this kind of

radiation on the process of crystallization of AMAs, the situation is analogous. Works [8, 11] present the results of studies of the effect of γ -radiation on the crystal phase in alloys, whose amorphous matrix contains a small amount ($<5\%$) of crystals. The authors of work [8] testify to the radiation-stimulated crystallization of alloys $(\text{FeCr})_{85}\text{B}_{15}$. But work [11] demonstrates the results which indicate that, under the action of γ -radiation, there occurs the fracture of crystals in alloy $(\text{Fe}_{90}\text{Cr}_{10})_{85}\text{B}_{15}$.

The authors of works [1, 10–14] agree only in that the mechanisms of structural changes in AMAs are related to the migration of atoms of light elements – B, C, Si.

2. Materials, Methods of Their Fabrication, and Results of Investigations

The object of studies is the structure of alloys of the Metglas type: MG-1 – $\text{Fe}_{80}\text{Si}_6\text{B}_{14}$, MG-3 – $\text{Fe}_{78.5}\text{Si}_6\text{B}_{14}\text{Ni}_1\text{Mo}_{0.5}$, MG-5 – $\text{Fe}_{82}\text{Si}_2\text{B}_{16}$, and MG-8 – $\text{Fe}_{75.5}\text{Si}_2\text{B}_{16}\text{Ni}_{3.5}\text{Mo}_3$. Amorphous strips of these alloys were produced by the method of spinning of a melt in air on an installation of the open type. Their sizes were as follows: the width was 10 mm, and the thickness was 23–26 μm . In the fabrication of alloys, we took high-purity components: Fe – 99.96 mass. %, Si – 99.999 mass. %, B – 99.9 mass. %, Ni \geq 99.9 mass. %, and Mo \geq 99.8 mass. %. The alloys were produced in an induction furnace in the inert atmosphere of Ar. The chemical composition of alloys was determined by the method of X-ray fluorescence analysis.

The structural state of the specimens of amorphous strips was investigated on a diffractometer DRON-3 in monochromatic MoK_α -emission. A monochromator (a LiF single crystal) was positioned in the primary beam. We calculated the structural factors $i(s)$ ($s = 4\pi \sin \theta / \lambda$, where θ is a half of the scattering angle, and the wavelength $\lambda = 0.71 \text{ \AA}$) in the interval of the diffraction vector from 0.77 to 12.5 \AA^{-1} with a scanning step equal to $6'$ in the region of the main maximum and to 0.5° for the other scattering angles. The survey duration was chosen so that the statistical error of calculations on the “tail” of an X-ray diffraction pattern be at most 1 %. The errors of the determination of the heights of maxima of the structural factor, $i(s_1)$ and $i(s_2)$, were ± 0.05 , and the errors of positions of its maxima, s_1 and s_2 , were $\pm 0.01 \text{ \AA}^{-1}$.

The Curie temperature T_C and the crystallization temperature T_X of amorphous alloys were determined from the temperature dependences of their magnetization $J(T)$ which were measured on a vibrating

magnetometer LDJ MODEL 9000–9500. The rates of heating of the specimens under study were constant and equal to 10 and 20 $^\circ\text{C}/\text{min}$, and the magnetic fields acting on the specimens were 1.3 kA/m and 0.56 MA/m. While approaching the Curie temperature, the temperature dependences of the magnetization of MG-alloys in the saturation field (0.56 MA/m) smoothly decreased to zero. In fields far from the state of magnetic saturation of specimens (1.3 kA/m), we observed a sharp drop of the magnetization under approaching T_C , which allows us to increase the measurement accuracy for this characteristic. The growth of the magnetization of such materials at higher temperatures is related to the formation of crystals $\alpha\text{-Fe}(\text{Si})$ in the amorphous matrices of alloys (the primary crystallization). The primary crystallization is running in two stages. At the temperature of the beginning of primary crystallization, T_{X1} , we observed the separation of crystals of a solid solution of Si in bcc-iron. At the higher temperature T_{X2} , the stage of intense primary crystallization begins. The stages are distinguished by the change in the slope of the plot $J(T)$. The measurements were executed on three control specimens and three ones irradiated by a certain dose. The dispersion of values of T_C and T_X for specimens made of each of the alloys did not exceed 3 $^\circ\text{C}$.

3. Experimental Results and Their Discussion

The structure of AMA is characterized by SO. In order to determine the parameters of SO in AMAs, one uses the X-ray structural factor $i(s)$. It represents the interference pattern of the scattering of coherent X-ray radiation by a specimen. In Table 1, we give the results of studies of the effect of electron radiation ($F = 2 \times 10^{17} \text{ cm}^{-2}$) on the basic parameters of the structural factor of MG-alloys: the height of the first maximum $i(s_1)$ and the positions of the first and second maxima s_1 and s_2 of amorphous alloys.

As seen from Table 1, the irradiation induces a significant change in only the height of the first

Table 1. Effect of electron radiation ($\Phi = 2 \times 10^{17} \text{ electron/cm}^2$) on the parameters of the structural factor of amorphous alloys

Alloy	Specimen	$i(s_1)$	$S_1, \text{\AA}^{-1}$	$S_2, \text{\AA}^{-1}$
MG-1	Control	3.65	3.11	5.24
	Irradiated	3.39	3.11	5.26
MG-3	Control	3.79	3.10	5.20
	Irradiated	3.82	3.10	5.22
MG-5	Control	3.49	3.09	5.20
	Irradiated	3.67	3.11	5.23
MG-8	Control	3.74	3.10	5.25
	Irradiated	3.80	3.11	5.24

maximum of the structural factor $i(s_1)$. It is the most structure-sensitive parameter $i(s_1)$ of the amorphous alloys under study. It is seen that the height of the first maximum of the structural factor of doped alloys MG-3 and MG8 under the action of radiation varies essentially less than that of undoped alloys MG-1 and MG-5. In addition, of interest is the fact of the opposite effect of radiation on $i(s_1)$ of the base alloys MG-1 and MG-5, despite a small difference in their chemical composition. The chemical compositions of these alloys differ significantly from each other by the concentration of Si, whereas the concentrations of Fe and B in them are approximately the same.

To clarify the possible reasons for such a difference, we studied the dependence of the height of the first maximum of the structural factor $i(s_1)$ of amorphous alloy MG-1 on the dose of electron radiation. The results of studies are presented in Fig. 1, *a*. As seen, the dependence of $i(s_1)$ on the radiation dose is nonmonotonous. The irradiation of amorphous alloy MG-1 with the dose which does not exceed approximately $0.2 \times 10^{17} \text{ cm}^{-2}$ leads to an increase in the height of the first maximum of the structural factor. In the interval of doses from 0.2×10^{17} to 0.8×10^{17} electron/cm², the height of the first maximum of the structural factor of alloy MG-1 is practically invariable, and it decreases at higher radiation doses. On the whole dose dependence, the height of the first maximum of the structural factor does not exceed the values (4.04 units), at which we cannot reveal the domains with crystal structure in the alloy. The amorphous strips, for which $i(s_1)$ does not exceed 4 units, are called usually "X-ray-amorphous" [15, 16]. Thus, the irradiation by electrons with an energy of 1 MeV causes no crystallization processes in specimens of amorphous alloy MG-1.

Analogous studies of a structure were carried out for the amorphous alloys irradiated with γ -quanta. In Fig. 1, *b*, we give the dose dependences of the height of the first maximum of the structural factor of alloys MG-1, MG-3, and MG-8. First, it is seen that $i(s_1)$ of the alloys doped with Ni and Mo is less sensitive than that of undoped ones for this kind of radiation. Second, the dependences of the height of the first maximum of the structural factor of alloy MG-1 under electron and γ -radiations has a similar form. This can indicate the identity of the mechanisms of a change in SO under the action of these kinds of radiation. With regard for the fact that the changes in SO in metallic systems under the action of γ -radiation are caused mainly by the action of secondary electrons [17], the result seems to be regular. In our case, the greatest contribution is given by

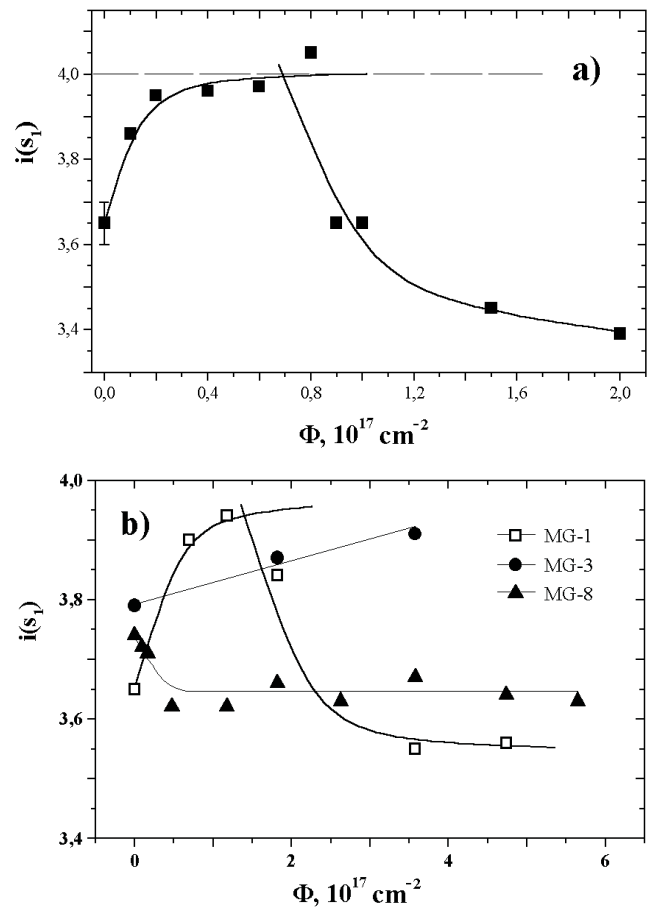


Fig. 1. Dependence of the height of the first maximum of the structural factor of alloy MG-1 *a*) on the dose of electron of radiation and alloys MG-1, MG-3, and MG-8 *b*) on the dose of γ -radiation

Compton electrons. Therefore, the difference (in quantitative indicators at certain doses of radiation) of the effects of electron and γ -radiations on the structure of MG alloys is caused by 1) difference in the energy parameters of high-energy electrons (those of the electron beam and secondary electrons, respectively); 2) different homogeneities (due to different values of the absorption coefficients for electrons and γ -quanta, respectively) of their radiation treatment.

In the general case, the observed changes in the height of the first maximum of the structural factor are the evidence of radiation-stimulated changes in SO in MG alloys. The nonmonotonicity of the dependence of $i(s_1)$ on F_e for undoped alloys testifies to the realization of several mechanisms of structural changes in a material. A decrease in the height of the first maximum

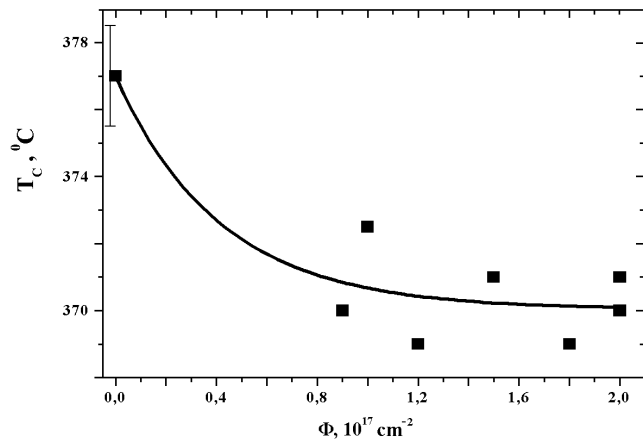


Fig. 2. Dependence of the Curie temperature ($H_m = 0.56 \text{ A/m}$, $v = 20 \text{ }^{\circ}\text{C/min}$) of alloy MG-1 on the dose of electron radiation

of the structural factor in AMA under the action of radiation is the well-known result [6–9, 11]. Such an effect is related to the disordering of an atomic structure. Therefore, a decrease in $i(s_1)$ at doses of radiation from 0.8×10^{17} to 2×10^{17} electron/ cm^2 can be associated with a decrease in the number and/or size of clusters which are dominant by a type of SO. In other words, under the interaction of high-energy electrons with the atomic structure of an amorphous alloy, there occurs the fracture of chemically ordered atomic formations with the type of SO which corresponds to $\alpha\text{-Fe}(\text{Si})$, Fe_3B . The increase in the height of the first maximum of the structural factor of alloy MG-1 at low doses of radiation (Fig. 1) turns out to be the unexpected result. We obtained a distinctive “effect of small doses” which indicates the radiation-stimulated ordering of the structure. The difference of radiation-stimulated structural changes in AMAs in different intervals of radiation doses can be caused by different mechanisms of changes of SO. In many-component amorphous alloys including MG-alloys, one should distinguish the compositional and topological SOs [16]. The compositional (chemical) SO describes an ordered distribution of atoms of different sorts. It is obvious that this mechanism of a change in SO more significantly affects the height of the first maximum of the structural factor of alloy MG-1 at great radiation doses. The topological (configurational, geometric) SO describes a spatial arrangement of atoms without regard for their chemical nature. An increase in $i(s_1)$ in amorphous alloy MG-1 at low radiation doses can be related to the topological ordering, at which there occurs a decrease of fluctuations of the elastic interatomic interaction [18] leading to a decrease in the dispersion

of interatomic distances. An analogous increase in $i(s_1)$ AMA is caused by a low-temperature thermal treatment of MG-alloys, at which one observes the relaxation of the amorphous alloy structure [15, 19]. The structural changes induced by these types of treatment are, of course, not identical. However, the mechanisms of a change in SO in these systems can be similar. For amorphous alloys obtained by the method of spinning, a characteristic feature is the presence of high internal stresses [20] and the free volume in the structure [21, 22]. On the structural relaxation, there occur a partial elimination of internal stresses and a decrease in the free volume [21, 22]. The atomic structure of alloys becomes more homogeneous [20]. That is, the increase of the height of the first maximum of the structural factor of an alloy can be caused by radiation-stimulated relaxational atomic displacements. The invariability of the height of the first maximum of the structural factor of an alloy in the interval of radiation doses from 0.2×10^{17} to 0.8×10^{17} electron/ cm^2 can apparently testify to the weak action of both mechanisms of a change in SO: radiation-stimulated relaxation and disordering. In this case, the radiation-induced relaxation stops to significantly affect the height of the first maximum of the structural factor, whereas the radiation-induced disordering begins to act. Thus, it is clear that each of the mechanisms of the action of radiation on the structure of AMA can be efficient in different intervals of radiation doses, by depending on both the conditions of its fabrication and its chemical composition.

The same mechanisms of the action of electron radiation on a structure can be inherent to other alloys of the given system. The difference of the significant effects of this kind of radiation on the structural factor of MG-alloys (Table 1) can be related to the different radiation resistance of different atomic formations [7, 23], whose number depends on their chemical composition. The absence of the significant effect of radiation on the parameters of the structural factor of alloys doped with Ni and Mo (Table 1, Fig. 1, *b*) is obviously caused by a high radiation resistance of clusters which are composed of atoms of these doping components and atoms of the most mobile components such as Si and, first of all, B.

On the mechanisms of the effect of radiation on both the structure of AMAs and the process of their crystallization, we can judge on the basis of the data on changes of their Curie temperature T_C and the crystallization temperature T_X . The Curie temperature is defined by the nearest environment of “magnetic” atoms [24]. Changes of the crystallization temperature due to the action of various external factors are related

to a change in the number of clusters with a short-range order inherent to nuclei of the crystal phase [1, 2, 6, 25].

In Fig. 2, we display the dose dependence of the Curie temperature $T_C(F_e)$ in the interval of doses from 0.9×10^{17} to 2×10^{17} electron/cm². The dependence $T_C(F_e)$ for alloy MG-1 decreases monotonously with the tendency to approach a stationary level at high radiation doses. The Curie temperatures of nonirradiated alloy MG-1 and the alloy irradiated with electrons by a dose of 2×10^{17} electron/cm² are, respectively, 377 and 370 °C (Fig. 2). A similar form of the dependence of the Curie temperature on the dose of proton radiation for amorphous alloys of the Ni-Fe-B system was obtained in [26]. Figure 3 shows the effect of electron radiation at a dose of 2×10^{17} electron/cm² on the temperature dependences of the magnetization of alloys MG-1, MG-5, and MG-8. On this basis, we determined the temperature Curie T_C and the temperatures of the initial (T_{X1}) and intense (T_{X2}) stages of primary crystallization of amorphous alloys. The results of studies are given in Table 2. It is seen that a shift of the temperatures T_C , T_{X1} , and T_{X2} of irradiated specimens of alloys relative to those of control specimens depends significantly on the chemical composition. We note again that the doping of alloys of the Fe-Si-B system with Ni and Mo increases the radiation stability of their physical characteristics. As seen from Table 2, no changes of the Curie temperature and the temperatures of the beginning of the different stages of primary crystallization under the action of electron radiation are revealed for alloys MG-3 and MG-8. The effect of electron radiation at a dose of 2×10^{17} electron/cm² on T_C for alloys MG-1 and MG-5 (Table 2), as well as on $i(s_1)$ (Table 1), turns out opposite. We observe the decrease in T_C by 7 °C for irradiated alloy MG-1 and the increase by 18 °C for MG-5.

The effects of radiation on the temperature of the initial stage of crystallization of MG-alloys are

Table 2. Effect of electron radiation ($\Phi = 2 \times 10^{17}$ electron/cm²) on the temperatures of phase transitions in amorphous alloys ($v_h = 10$ °C/min)

Alloy	Specimen	T_c , °C	T_{X1} , °C	T_{X2} , °C
MG-1	Control	386	410	492
	Irradiated	379	405	495
MG-3	Control	369	423	495
	Irradiated	367	423	494
MG-5	Control	340	370	450
	Irradiated	358	370	465
MG-8	Control	252	375	461
	Irradiated	251	375	460

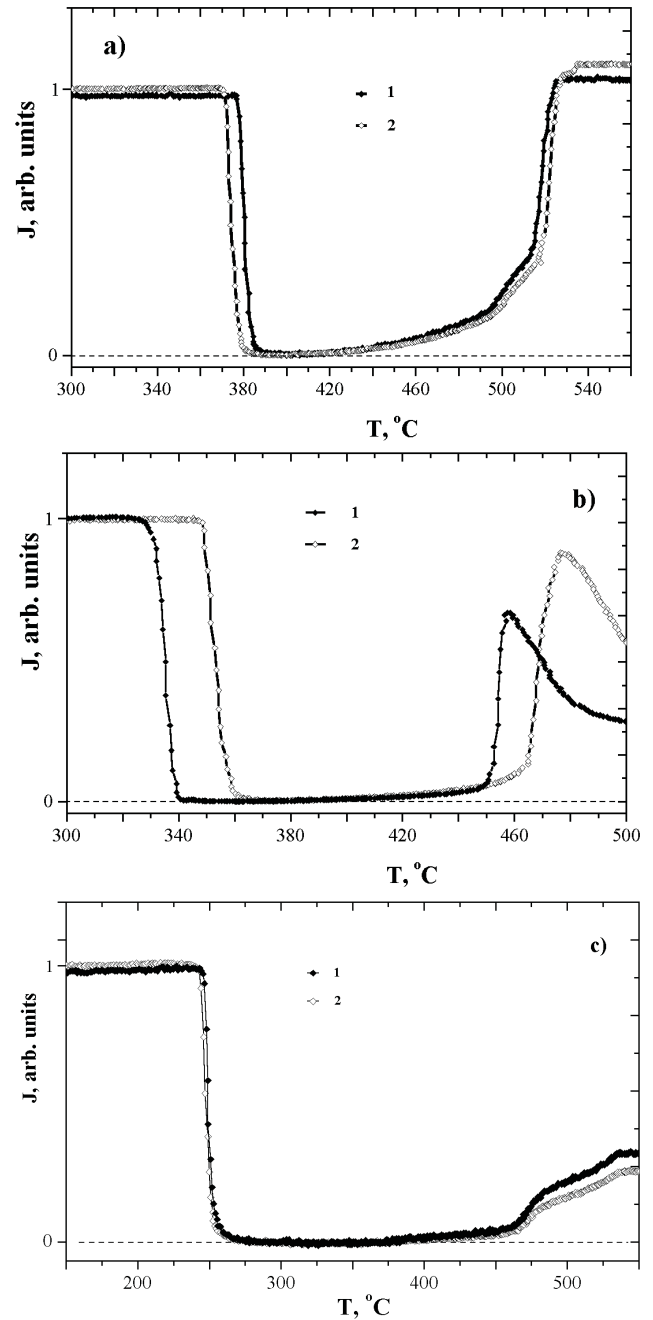


Fig. 3. Temperature dependences ($H_m = 1.3$ A/m, $v = 10$ °C/min) of the magnetization of control alloys 1 and 2 alloys MG-1 a), MG-5 b), and MG-8 c) irradiated by electrons (2×10^{17} electron/cm²)

manifested less significantly. For alloy MG-1, we observe a decrease in T_{X1} by 5 °C, whereas T_{X1} for alloy MG-5 does not vary. The temperature of the intense stage of crystallization, T_{X2} , grows considerably (by 15 °C) only for alloy MG-5.

Despite a certain similarity of the data on the effect of radiation on the Curie temperature, crystallization temperature, and the parameters of the structural factor of undoped alloys, we cannot interpret the effect of electron radiation on changes of T_C and T_X in terms of “radiation-induced relaxation” and “disordering”. The reason consists in the absence of a correlation between the dose dependences of the height of the first maximum of the structural factor and the Curie temperature (Figs. 1 and 2) and in the absence of reliable data on how such mechanisms of a change in SO affect these quantities. Therefore, it is expedient to relate the difference of the effects of radiation on values of the Curie and crystallization temperatures of MG-alloys to the difference of their chemical compositions.

A magnetism of AMAs of the Metglas type is defined by the direct exchange interaction between d -electrons of transition elements [27]. The Curie temperatures of amorphous, as well as crystalline, ferromagnetic materials are defined by SO. As distinct from crystals, amorphous alloys are characterized by several types of SO, each of which corresponds to a certain value of the exchange integral. The quantity T_C for ferromagnetic materials is a measure of the exchange interaction between atoms. Therefore, the changes in this characteristic are related to both a change in the number of the nearest “magnetic” atoms and the dependence of the exchange integral on the distance between them. The opposite characters of changes of the Curie temperatures of alloys MG-1 and MG-5 under the action of radiation can be related, first of all, to the different concentrations of Si which is one of the most mobile components of these materials. In alloy MG-5, the concentration of Si is low as compared with the concentration of B. Therefore, the change in T_C under the action of radiation on this material can be certainly related to the radiation-stimulated diffusion of atoms of B. The opposite change of this characteristic in MG-1 under the action of radiation can be related to the diffusion of Si atoms. It is probable that the mechanisms of diffusion of atoms of these elements are different. The increase in the concentration of B to about 30 % in amorphous alloys Fe-B induces the growth the Curie temperature. It is considered that B diffuses by the interstitial mechanism [24, 27]. Therefore, such changes of T_C in alloy MG-5 according to the Bethe–Slater curve can be associated with increase in the exchange interaction due to an increase in distances between Fe atoms [24, 28, 29]. Si atoms play the role of substitution elements. Therefore, as the concentration of Si in alloys of the Fe–Si–B system increases, the decrease in T_C

under the action of electron radiation can be a reason for the decrease in the number of Fe-Fe coordinations [24].

The development of the crystallization processes in MG-alloys on the initial stage of primary crystallization is running mainly by the mechanism of creation of nuclei of crystals. On the second intense stage of primary crystallization of these materials, the growth of crystals dominates [30, 31]. It is expedient to begin the analysis of the effect of radiation-stimulated diffusion of atoms of Si and B on the mechanisms of crystallization in MG-alloys from MG-5. The electron irradiation has no effect on T_{X1} (Table 2) for this material. This means that the radiation-induced diffusion of B renders no significant effect on the process of origination of crystals. A significant increase in T_{X2} can testify that the diffusion of atoms of this element to nuclei of the crystal phase decelerates the process of growth of crystals. The decrease in T_{X1} of alloy MG-1 can indicate that the radiation-induced diffusion of Si stimulate the creation of nuclei. The high solubility of this element in α -Fe can be a reason for a less effect of radiation on T_{X2} of alloy MG-1. Therefore, it is probable that the radiation-induced diffusion of Si has no significant effect on the growth of crystals.

The invariability of T_c , T_{X1} , and T_{X2} for alloys MG-3 and MG-8 under the action of electron radiation can be related to the smallness of the diffusion coefficient of B atoms which belong to clusters containing also Mo and/or Ni.

4. Conclusions

1. We have revealed the effect of “small doses” under the action of electron and γ -radiations on amorphous metallic alloys. In this connection, we have proposed a mechanism of radiation-stimulated relaxation in addition to the well-known mechanism of radiation-induced disordering.

2. It is established that the changes of the structural factor, Curie temperature, and crystallization temperature of alloys of the Fe–Si–B system under the action of radiation depend essentially on the concentrations of Si and B in their composition. It is established that the difference of the effects of radiation on the structure and the process of crystallization in these alloys is related to the difference of the mechanisms of radiation-stimulated diffusion of atoms of Si and B.

3. We have shown that the doping of alloys of the Fe–Si–B system with Ni and Mo leads to a decrease in the sensitivity of the structural factor, Curie

temperature, and crystallization temperature to the action of radiation.

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ВПЛИВ ЕЛЕКТРОННОГО І ГАММА-ОПРОМІНЕННЯ НА СТРУКТУРУ АМОРФНИХ СПЛАВІВ Fe-Si-B

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Резюме

Методами рентгеноструктурного аналізу та термомагнітометрії вивчено вплив електронного ($E = 1$ MeV) і γ -опромінення ($E = 1, 17$ та $E = 1, 33$ MeV) на структурні зміни в аморфних сплавах системи Fe-Si-B. Досліджено дозові залежності висоти першого максимуму структурного фактора $i(s_1)$ та температури Кюрі T_C сплавів системи Fe-Si-B, а також вплив електронного опромінення на їх температури Кюрі та температури кристалізації T_X . Експериментально показано, що механізми впливу електронного і гамма-опромінення на структуру аморфних металевих сплавів системи Fe-Si-B однакові. Встановлено, що легування сплавів Fe-Si-B атомами Ni та Mo приводить до зменшення радіаційної чутливості їхніх характеристик. Знак та величина ініційованих опроміненням змін температури Кюрі та температури кристалізації нелегованих сплавів суттєво залежить від співвідношення в їхньому складі концентрацій Si і B. Запропоновано два механізми впливу опромінення на структуру аморфних металевих сплавів та якісну модель впливу радіаційно-стимульованої дифузії Si та B на T_C і T_X аморфних сплавів системи Fe-Si-B.