INFLUENCE OF γ-IRRADIATION ON INITIAL MAGNETIC PERMEABILITY OF AMORPHOUS AND NANOCRYSTALLINE Fe–Si–B-BASED ALLOYS

V.YU. POVARCHUK,[1] V.K. NOSENKO,[2] A.M. KRAITCHINSKII,[1] V.B. NEIMASH,[1] M.M. KRAS'KO,[1] V.V. MASLOV[2]

[1]Institute of Physics, Nat. Acad. of Sci. of Ukraine
(46, Prosp. Nauky, Kyiv 03028, Ukrain; e-mail: pvyu@i.ua)

[2]G.V. Kurdyumov Institute for Metal Physics, Nat. Acad. of Sci. of Ukraine
(36, Academician Vernadskyi Ave., Kyiv 03142, Ukraine)

PACS 61.82.Bg, 75.70.-i

By determining the inductance factor, the dependence of the initial magnetic permeability μ_i of amorphous and nanocrystalline Fe–Si–B-based alloys on the γ-irradiation dose has been studied. The doping of amorphous Fe–Si–B alloys with nickel and molybdenum is found to enhance the radiation sensitivity of μ_i. The initial magnetic permeability of nanocrystalline magnetic alloys is determined to be less sensitive to the action of γ-radiation than that of doped amorphous ones. A hypothesis is put forward that the influence of radiation on the initial magnetic permeability is associated with the creation of non-magnetic inclusions in the structure of amorphous alloys and in the amorphous matrix of nanocrystalline alloys.

1. Introduction

Amorphous and nanocrystalline alloys on the basis of the Fe–Si–B system are characterized by high values of saturation induction and magnetic permeability and by low magnetic reversal losses. Therefore, they are widely applied in the manufacture of inductive elements for the electrotechnical equipment: power transformers, small-sized transformers operating at high frequencies, throttles, *etc.* From the scientific viewpoint, the researches of the radiation influence on structurally sensitive characteristics of amorphous and nanocrystalline alloys can be useful for establishing the mechanisms of radiation-induced structural changes in such systems. An application purpose of our research was to study the expediency of using amorphous and nanocrystalline ferromagnets on the basis of the Fe–Si–B system under ionizing radiation conditions, as well as a possibility to control the properties of those substances taking advantage of their radiation treatment.

Available literary data did not enable us to estimate unambiguously the influence of γ-radiation on magnetic properties of amorphous soft magnetic alloys. The authors of work [1] reported that the irradiation to the dose $\Phi = 10^9$ rad stimulates a growth of the initial magnetic permeability μ_i and the residual induction B_r, as well as a reduction of H_c for magnetic circuits fabricated from amorphous soft magnetic alloys $Fe_{85-x}Co_xB_{15}$ ($x = 12 \div 25\%$). On the contrary, the results of work [2] testified to a deterioration of magnetic properties of amorphous FeNiMoSiB alloys subjected to this kind of radiation treatment. The irradiation of initial (nonannealed) amorphous alloy $Co_{83.5}Fe_{5.5}Si_{8.5}B_{2.5}$ to the dose $\Phi = 10^7$ rad stimulated the growth of both B_r/B_s and H_c [3]. For today, there are no data concerning the influence of γ-radiation on magnetic properties of nanocrystalline alloys on the basis of Fe–Si–B.

Ambiguous data on the influence of γ-radiation on magnetic characteristics of amorphous alloys made it expedient to study their dose dependences in the range as wide as possible. Evidences in favor of this purpose are also given by a nonmonotonous character of the dose dependence revealed by the first maximum height in the structural factor [4].

2. Experimental Materials, Methods of Their Fabrication, and Research Techniques

In this work, we studied the influence of γ-radiation (^{60}Co was a radiation source, $E_\gamma = 1.17$ and 1.33 MeV, the flow density $\phi_\gamma \approx 10^{11}\gamma_{\rm quant}/({\rm cm}^2{\rm s})$) applied in doses up to 5.7×10^{18} cm^{-2} = 3.4×10^9 rad on the initial magnetic permeability of amorphous alloys of the Metglas type (MG-alloys) and nanocrystalline alloys of the Finemet type (FM-alloys). The chemical compositions of MG-alloys are given in Table 1 and those of FM-alloys in Table 2. From Table 1, one can see that the MG-alloys can be divided into two groups according to the concentration of nonmetallic components in them: Fe-Si_6B_{14} (alloys MG1 to MG-3) and Fe–Si_2B_{16} (alloys MG-5 to MG-8). All alloys, but MG-1 and MG-5, were doped with Ni and Mo atoms. Doping MG-alloys with those atoms enhances their thermal stability and improves their magnetic properties [5].

For the preparation of initial alloys, components of high purity were used: Fe – 99.96 wt.%, Si – 99.999 wt.%, B – 99.9 wt.%, Ni $\geq$ 99.9 wt.%, Mo $\geq$ 99.8 wt.%, Cu $\geq$ 99.9 wt.%, Nb $\geq$ 99.9 wt.%, and Co $\geq$ 99.9 wt.%. Alloy MG-3T was produced from components of technical purity: Fe $\geq$ 98.6 wt.%, Mo $\geq$ 99 wt.%, ferroboron FB-17 (B $\approx$ 20.5 wt.%, Al – 1.2%, Si – 0.4%), and ferronickel FN-5 (Ni – 7.8 wt.%, Co – 0.36%, Cu – 0.11%, Al – 0.1%, Si – 0.03%).

Alloys were produced in an induction furnace, in an inert Ar atmosphere. The chemical composition of alloys was determined using the method of X-ray fluorescence analysis. Amorphous ribbons 10 mm in width and 24–26 μm in thickness were fabricated using the method of melt spinning in air on an open-type installation.

To study the magnetic properties of amorphous and nanocrystalline alloys, the initial (nonannealed) amorphous ribbon was used for the fabrication of magnetic circuits (circular cores) with the help of a special equipment for winding with the lowest tension. The geometrical parameters of twisted circular cores in the specimens were as follows: an internal diameter of 12 mm, an external diameter of 20 mm, and a height of 10 mm. Magnetic circuits made of MG-alloys were annealed at a temperature of 420 °C for 15 min in the carbon dioxide atmosphere. Cores fabricated from FM-6, FM-10, and FM-11 alloys were annealed in the carbon dioxide atmosphere at the temperature T = 520 °C for 30 min, and those from alloy FM-2T at T = 535 °C for 60 min. To measure the initial magnetic permeability ($H_m = 0.2$ A/m, $f = 10$ and 100 kHz), we applied the method of inductance factor determination [6]. The determination accuracy for μ_i was 5%.

T a b l e 1. Chemical composition (at.%) of amorphous alloys of the Metglas type

No	Alloy	Fe	Si	B	Ni	Mo
1	MG-1	80	6	14		
2	MG-2	76.2	6	14	3.8	
3	MG-3T	78.5	6	14	1	0.5
4	MG-5	82	2	16		
5	MG-6	78	2	16	1	3
6	MG-7	77.5	2	16	3.5	1
7	MG-8	75.5	2	16	3.5	3

3. Experimental Results and Their Discussion

The thermal treatment of MG-alloys gave rise to an increase of their initial magnetic permeabilities μ_i by several times and a reduction of the corresponding coercive forces H_c; the hysteresis loop became narrower, and the number of Barkhausen jumps diminished. Those phenomena are connected with a reduction of the magnetic anisotropy owing to the elimination of internal stresses [7]. These alloys find a wide application in the industry. The magnetic characteristic, which is the most sensitive to their structure, is the initial magnetic permeability μ_i. In this work, we report results of our researches concerning the influence of radiation on this parameter obtained for magnetic circuits annealed in the optimum regime.

From Fig. 1, one can see that radiation did not affect the initial magnetic permeability of undoped alloys MG-1 and MG-5. In MG-alloys doped with nickel and molybdenum, a reduction of μ_i was observed, as the exposure dose of γ-radiation grew. The largest relative variations of μ_i were observed for alloys MG-6 and MG-8 (by 28 and 23%, respectively), in which the Ni concentration amounted to 1 and 3.5%, respectively, and the Mo concentration was 3%. For alloy MG-7, in which the Ni concentration was 3.5% and the Mo concentration was 1%, the relative variation of μ_i was about 20%. An insignificant reduction of the initial magnetic permeability was observed for alloy MG-2, which contained 3.8% Ni and did not contain Mo. Hence, the reduction of the initial magnetic permeability in the examined MG-alloys under the action of γ-radiation can be associated, first of all, with the presence of Mo atoms in their chemical compositions.

The dynamics of variations in the initial magnetic permeability of alloy MG-3T fabricated from components of technical purity under the influence of irradiation is somewhat different (Fig. 1,*a*). It is evident that the impurities that enter into its composition play a substantial role.

T a b l e 2. Chemical composition (at.%) of amorphous alloys of the Finemet type

No	Alloy	Fe	Si	B	Cu	Nb	Co
1	FM-2T	73	15.8	7.2	1	3	
2	FM-6	73.6	15.8	7.2	1	2.4	
3	FM-10	71.25	16.4	7.7	1	2.1	1.55
4	FM-11	70.05	16.4	9	1	2	1.55

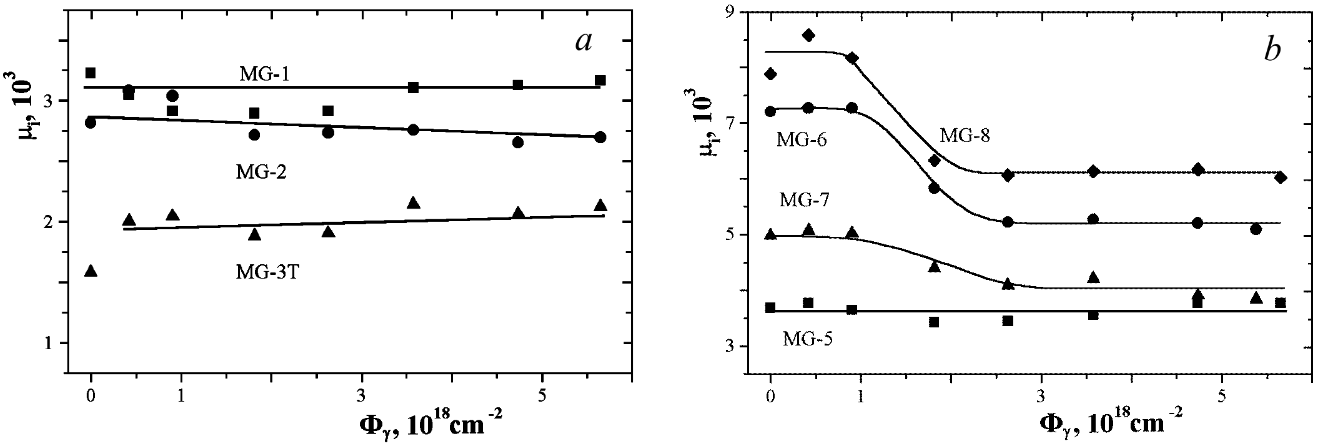

Fig. 1. Dose dependences of the initial magnetic permeability ($f = 10$ kHz) of magnetic circuits fabricated from MG-alloys

In work [4], we showed that the doping of amorphous alloys Fe–Si–B with nickel and molybdenum reduces the sensitivity of their structural factors $i(s)$ to radiation. This fact testifies that, at least in doped MG-alloys, the influence of radiation on structure-sensitive magnetic characteristics is not connected with variations in the short-range integral parameters. In view of the dependence of radiation-induced changes in the initial magnetic permeability of MG-alloys on the concentration of doping components, we may suppose that irradiation brings about the formation of clusters, which contain, first of all, molybdenum atoms and atoms of the most mobile alloy components, B and/or Si. The appearance of such nonmagnetic inclusions can lead to a decrease in the mobility of domain walls [8]. A possibility of their formation was confirmed in [9, 10]. The regions enriched with molybdenum are formed in the near-surface layer of amorphous metal alloys (AMAs). The authors of works [11–17] considered that the surface of boron-containing AMAs is also enriched with boron atoms, especially the free side of a ribbon, which did not contact with the disk surface at manufacturing an alloy [11, 12]. The enhanced concentration of atoms of this element near the AMA surface was observed not only in the initial amorphous ribbons, but also in the thermally treated ones [13–17]. A thermal treatment additionally increases the boron concentration in the near-surface layers (about 50 Å in thickness) of ribbons [13]. Therefore, the formation of clusters containing molybdenum and boron atoms under the influence of radiation should be expected to take place in the near-surface layers of a ribbon. The invariance of the initial magnetic permeability for Fe–Si–B alloys at irradiation doses above 2.5×10^{18} $\gamma_{\rm quant}/{\rm cm}^2$ can be explained by a saturation of chemical bonds between Mo atoms and atoms of nonmetallic elements.

Additional researches are needed to establish the basic mechanisms of influence of γ-radiation on the structure and the magnetic properties of undoped alloys belonging to the Fe–Si–B system.

In Fig. 2, the dependences of the initial magnetic permeability of nanocrystalline alloys on the exposure dose of γ-radiation are depicted. By comparing the data in Figs. 1 and 2, we arrived at a conclusion that μ_i's for FM-alloys are less sensitive to the radiation action, than the magnetic permeabilities of MG-alloys doped with molybdenum and nickel. The maximum variations of the initial magnetic permeability stimulated by γ-irradiation were 12% for alloy FM-2T, 8% for alloy FM-6tn ("tn" means a thinner ribbon of 24 μm), and 12% for alloy FM-6tc ("tc" means a thicker ribbon of 38 μm) (see Fig. 2,*a*). For the magnetic circuits fabricated from FM-10 and FM-11 alloys containing Co, a reduction of μ_i did not exceed 5 and 7%, respectively (Fig. 2,*b*).

Nanocrystalline alloys of the Finemet type are biphase systems, in which both the crystalline and amorphous phases are ferromagnetic at room temperature. The volume fraction of the crystalline phase in FM-alloys amounts to about 70%, and the composition of nanograins is estimated as Fe–Si (18–23 at.%) [18–20]. Other elements that enter into FM-alloys are practically insoluble in α-Fe(Si), being located therefore in the amorphous matrix. In general, variations of the initial magnetic permeability of FM-alloys stimulated by γ-radiation can be associated with atomic reconstructions in the amorphous matrix of alloys, as well as in the crystal lattice and at the crystal boundaries. However, the notable variations of the initial magnetic permeability

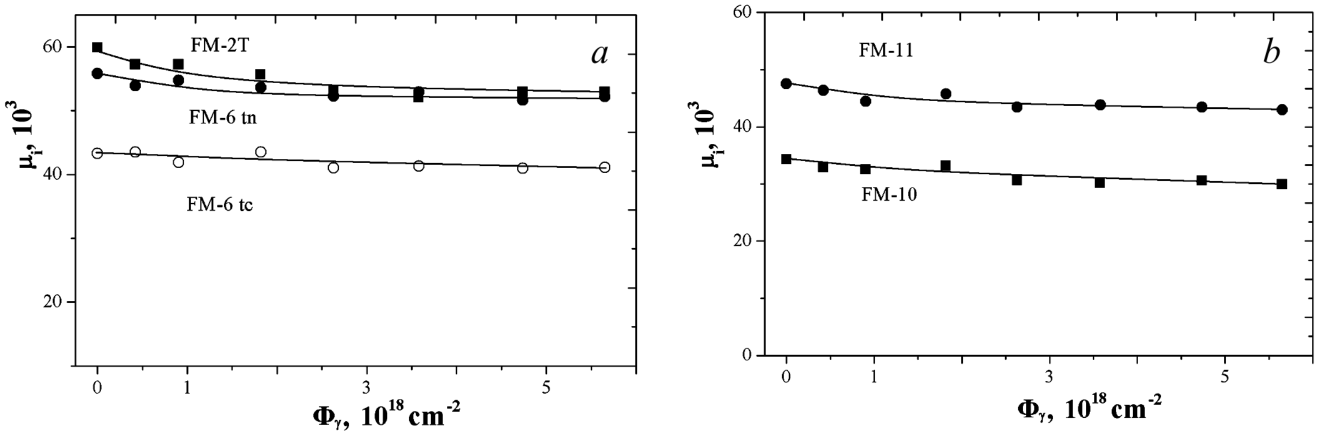

Fig. 2. Dose dependences of the initial magnetic permeability (f = 10 kHz) of magnetic circuits fabricated from FM-alloyis

of crystalline materials can be revealed at much higher doses of γ-radiation. This fact is confirmed by our researches dealing with the influence of the given kind of radiation on magnetic characteristics of the transformer steel. Since the most mobile element in the alloy, boron, is contained in the amorphous matrix, the radiation-induced variations of the initial magnetic permeability can be associated with its diffusion to crystal boundaries or with its participation in the formation of non-magnetic clusters in the amorphous matrix.

In addition, a reduction of the initial magnetic permeability under irradiation can occur owing to the growth of crystallite dimensions. From literature data [18, 21], it is known that the increase of a crystal size from 10 to 40 nm in alloys of the Finemet type worsens the magnetic properties of the latter. A substantial, by several orders of magnitudes, reduction of μ_i and an increase of H_c take place. However, with regard for the exposure dose and the fact that FM-alloys are rather stable quasiequilibrium systems (under their thermal treatment, the crystalline phase is segregated within a few minutes, i.e. the fraction of crystalline phase is not changed during almost the whole annealing process), this mechanism is hardly probable.

As is seen from Figs. 1 and 2, the substantial changes in μ_i and the saturation of the dependences $\mu_i(\Phi_\gamma)$ for MG-alloys doped with nickel and molybdenum, as well as for FM-alloys, occur within the same interval of γ-radiation doses. This fact may mean that the variations of the initial magnetic permeability stimulated by the radiation treatment connected with structural modifications in the amorphous matrix of FM-alloys. Moreover, a similarity of the dose dependences for initial magnetic permeabilities can also mean that the mechanisms of radiation influence on a structure of the amorphous matrix in FM-alloys and on a structure of MG-alloys are qualitatively identical. The role probably played by molybdenum in MG-alloys can be characteristic of niobium atoms in nanocrystalline ones. The formation of non-magnetic inclusions in FM-alloys under the action of ionizing radiation should be expected to take place at crystallite boundaries and/or in the near-surface layers of the ribbon.

4. Conclusions

In the present work,

1) the doping of amorphous alloys Fe–Si–B with nickel and molybdenum is found to enhance the sensitivity of their initial magnetic permeabilities to radiation action; the sensitivity of μ_i grows with the Mo concentration and depends a little on the Ni concentration;

2) the initial magnetic permeabilities of nanocrystalline alloys on the basis of Fe–Si–B are found to be less sensitive to the action of γ-radiation than those for doped amorphous alloys;

3) a hypothesis is advanced that the influence of radiation on the initial magnetic permeability is associated with the formation of non-magnetic inclusions in the structure of amorphous alloys and in the amorphous matrix of nanocrystalline ones.

1. V.V. Polotnyuk, T.V Efimov, A.M. Shalaev, and V.M. Shkapa, Metallofiz. **13**, No. 7, 97 (1991).
2. V.V. Maslov, V.K. Nosenko, V.B. Neimash, and V.Yu. Povarchuk, Metallofiz. Noveish. Tekhnol. **24**, 747 (2002).

3. A.M. Shalaev, V.V. Polotnyuk, M.P. Krulikovskaya, S.V. Zolkina, V.V. Kotov, and L.A. Olikhovskaya, Metallofiz. **6**, No. 4, 80 (1984).
4. V.Yu. Povarchuk, V.B. Neimash, A.M. Kraichinskii, V.K. Nosenko, V.V. Maslov, and G.M. Zelinska, Ukr. Fiz. Zh. **53**, 972 (2008).
5. V.V. Maslov, V.K. Nosenko, A.P. Brovko, G.M. Zelinskaya, and V.A. Mashira, Metallofiz. Noveish. Tekhnol. **24**, XXX (2002).
6. *Cores for inductance coils and transformers used long-distance communication facilities. Part 2. A guide on formulating specifications – GOST 29005-91 (MEK 367-1-74)* (Committee for Standardization and Metrology of the USSR, Moscow, 1991) (in Russian).
7. K. Suzuki, H. Fujimori, and K. Hashimoto, *Amorphous Metals* (Butterworths, London, 1983).
8. S. Chikazumi, *Physics of Ferromagnetism* (Oxford Univ. Press, New York, 2009).
9. L. Bednarska, M. Kovbuz, O. Bilyk, and M. Gorecha, in *Proceedings of the International Conference-Exhibition on the Problems of Corrosion and Anticorrosion Protection of Materials "Corrosion-98", Lviv, 1998,* p. 259.(in Ukrainian).
10. M. Naka, K. Hashimoto, and T. Masumoto, J. Non-Cryst. Solids **31**, 355 (1979).
11. A.Z. Nagy, B. Vasvari, P. Duwez *et al.*, Phys. Status Solidi A **61**, 689 (1980).
12. J. Nagy, T. Tarnoczi, M. Hosso, and F. Pavlyak, in *Proceedings of the Conference on Metallic Glasses: Science and Technology, Budapest, 1980* (Budapest, 1981), p. 223.
13. J.L. Walter, F. Bacon, and F.E. Luborsky, Mater. Sci. Eng. **24**, 239 (1976).
14. Th. Berghaus, H. Neddermeyer, W. Radlik, and V. Rogge, Physica Scripta **4**, 194 (1983).
15. T.I. Bratus, M.A. Vasilyev, and V.T. Cherepin, Metallofizika **5**, No. 1 (1983).
16. A.V. Romanova, V.V. Nemoshkalenko, G.M. Zelinskaya *et al.*, Metallofiz. **5**, No. 1, 71 (1983).
17. G.M.Zelinskaya, A.V. Romanova, V.V. Nemoshkalenko *et al.*, Ukr. Fiz. Zh. **28**, 1393 (1983).
18. I.B. Kekalo, *Nanocrystalline Soft Magnetic Materials* (MISiS Tekhn. Univ., Moscow, 2000) (in Russian).
19. I. Mat'ko, P. Duhaj, P. Švec, and D. Janičkovič, Mater. Sci. Eng. A **179–180**, 557 (1994).
20. E.N. Vlasova, N.B. Dyakonova, I.V. Lyasotskii, B.V. Molotilov, and D.L. Dyakonov, Metally No. 2, 55 (2001).
21. R.A. Andrievskii and A.M. Glezer, Fiz. Met. Metalloved. **88**, 50 (1999).

Received 29.03.11.
Translated from Ukrainian by O.I. Voitenko

ВПЛИВ γ-ОПРОМІНЕННЯ НА ПОЧАТКОВУ МАГНІТНУ ПРОНИКНІСТЬ АМОРФНИХ І НАНОКРИСТАЛІЧНИХ СПЛАВІВ НА ОСНОВІ СИСТЕМИ Fe–Si–B

В.Ю. Поварчук, В.К. Носенко, А.М. Крайчинський, В.Б. Неймаш, М.М. Красько, В.В. Маслов

Р е з ю м е

Методом визначення фактора індуктивності досліджено залежності початкової магнітної проникності μ_i аморфних і нанокристалічних сплавів на основі системи Fe–Si–B від дози γ-опромінення. Виявлено, що легування аморфних сплавів системи Fe–Si–B молібденом і нікелем приводить до збільшення радіаційної чутливості їхніх μ_i. Початкова магнітна проникність нанокристалічних сплавів на основі Fe–Si–B менш чутлива до дії γ-опромінення, ніж легованих аморфних. Висловлено припущення, що вплив радіації на початкову магнітну проникність зумовлений утворенням немагнітних включень у структурі аморфних і в аморфній матриці нанокристалічних сплавів.