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COMPARISON OF CORROSION RESISTANCE OF BULK AND TAPE AMORPHOUS $\text{Fe}_{60}\text{Co}_7\text{Cr}_4\text{V}_2\text{W}_1\text{Mo}_1\text{Nb}_1\text{B}_{20}\text{C}_2\text{Si}_2$ ALLOY IN DIFFERENT AGGRESSIVE ENVIRONMENTS

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Bulk Metallic Glasses (BMG or Bulk Amorphous Alloys) are a new class of metallic alloys with a unique amorphous atomic structure. Bulk amorphous alloys exhibiting attractive properties for their widespread applications. Even after significant progresses in their research, practical applications of bulk amorphous alloys seem to be still challenging. Amorphous alloys have some excellent physical and chemical properties, among them good mechanical and magnetic properties, high corrosion resistance in aggressive environments. In this paper, authors compared corrosion resistance of bulk and tape amorphous Fe-based alloy in 0.5 M aqueous solutions of NaCl, KOH and HCl. It was found that hydrochloric acid is the least aggressive environment for both samples of the amorphous alloy. Tape samples of Fe-based alloy have better corrosion resistance compared with bulk samples with the same elemental composition.

Keywords: bulk and tape amorphous alloys, corrosion resistance, aggressive environment, voltammetry, potentiometry

1. Introduction

Solid-state materials with the major bonding types including ionic, covalent, van der Waals, hydrogen, and metallic can be made by various ways into amorphous solid forms. Metallic amorphous alloys are comparatively newcomers to the amorphous materials group. The formation of the first metallic glass of $\text{Au}_{75}\text{Si}_{25}$ was reported by Duwez in 60-s [1]. They developed the rapid quenching techniques for chilling metallic liquids at very high rates of 10^5 – 10^6 K/s. The significance of Duwez's work was that their method permits large quantities of an alloy to be made into glassy state comparing to other methods, for instance, vapor condensation. Formation, structure and property investigations of metallic glasses have attracted increasing attention because of their fundamental scientific importance and engineering application potential [2–5]. The techniques of melt quenching have been extensively developed and elaborated for the purpose of producing a wide variety of metallic glasses.

If one arbitrarily defines the millimetre scale as "bulk", the first bulk metallic glass (BMG) was the ternary Pd–Cu–Si alloy prepared by Chen in 1974 [6]. They used simple suction-casting methods to form millimetre-diameter rods of Pd–Cu–Si metallic glass at a significantly lower cooling rate of 10^3 K/s. In 1982, Turnbull and co-workers [7-8] successfully prepared the well-known Pd–Ni–P BMG by using boron oxide fluxing method to purify the melt and to eliminate heterogeneous nucleation.

In the 1980s, a variety of solid-state amorphization techniques, which are based on completely different mechanism from rapid quenching, such as mechanical alloying, diffusion induced amorphization in multilayers, ion beam mixing, hydrogen absorption, and inverse melting, had been developed [3]. A variety of metallic glasses in the form of thin films, or powders can be obtained by interdiffusion and interfacial reaction at temperatures well below the glass transition temperatures. In the late 1980s, Inoue et al. in Tohoku University of Japan succeeded in finding new multicomponent alloy systems consisting mainly of common metallic elements with lower critical cooling rates [9, 10].

In addition to the importance to basic sciences, BMGs have some excellent physical and chemical properties which are promising for applications.

Since 1995, a series of Fe- and Co-based BMGs with ferromagnetism at room temperature has been developed by copper mold casting or water quenching [11–14]. The formation and properties of multicomponent Fe-based BMGs have attracted increasing attention because of the fundamental interests in their properties and industrial application potential. Compared with the Fe- and Co-based metallic glasses ribbons, the BMGs have the following advantages in soft magnetic properties [15]: 1) high electrical resistivity of 200–250 mOcm at room temperature; 2) lower coercive force of 0.2–4 A/m; 3) higher initial permeability; 4) controllable arrangement of domain wall structure achieved by control of casting and/or cooling processes; 5) better high-frequency permeability; (6) good micro-forming ability in supercooled liquid region.

Bulk metallic glasses have different resistance to dissolution due to its elemental composition and in environments with different pH. For example, Zr-based BMGs exhibit excellent corrosion resistance in 30%, 66% HNO₃ solutions; 9.1%, 17.2% NaCl solution and 20%, 40% NaOH solution for the function of passivating, which indicates its high corrosion resistance ability at acid, alkali and salt circumstances. Zr₆₅Al_{7.5}Ni₁₀Cu_{17.5} BMG sample is seriously corroded 5 mol/l HCl solution. However, by adding appropriate Nb element, its resistance to pitting corrosion can be greatly improved [16]. Fe-based BMGs shows lower corrosion current density values and displays higher pitting potential values than steels in the salt balanced solution [17], bulk amorphous Ni–5Cr–5Ta–3Mo–16P–4B alloy spontaneously passive in 6 and 12 M HCl [18]. Such corrosion properties of the BMGs can be used in their application.

Applications. With the unique and unconventional characteristics, BMG materials are adopted for application in various fields. One of the great advantages of BMGs is the ease of formation of complicated shapes. Up to now, BMGs have already been used as die materials (Pd–Cu–Ni–P BMG), sporting equipment (ZrTiCuNiBe and ZrTiNiCu BMGs) and electrode materials (PdCuSiP BMG). The development of Fe-based BMGs has reached the final stage for application as soft magnetic materials for common mode choke coils. Success in this area will result in the increasing importance [19].

Optimization of nanoscale microstructure is using in amorphous alloys for increasing their physico-chemical characteristics [20, 21]. Absence of grain boundaries and

composition homogeneity in nanoscale should ensure high chemical resistance and increase application competitive ability.

2. Experimental details

In the form of a tape amorphous alloy $\text{Fe}_{60}\text{Co}_7\text{Cr}_4\text{V}_2\text{W}_1\text{Mo}_1\text{Nb}_1\text{B}_{20}\text{C}_2\text{Si}_2$ (HB-4) was obtained by melt-spinning method (10^6 K/s) on a copper cooling rotating drum. For such amorphous metallic ribbons identify contact (c) and external (e) sides, which are characterized by different physico-chemical properties. Bulk samples of AMA were prepared in quartz or ceramic crucibles in air atmosphere in the Institute for metal physics of NAS of Ukraine, Kyiv.

Changes of electrochemical parameters of AMA tape were estimated by chronopotentiometric methods and cyclic voltammetric methods in potentiodynamic mode with automatic scanning in time (rate = 20 mV/s) fixed potential of metal surface with electrode scheme: AMA-electrode – Ag/AgCl/KCl_{sat} and auxiliary electrode – Pt-plate. The measurements were carried out on Potentiostat type EP 20A.

Microscopic investigation of the elemental composition of AMA surface and microphotographs of the surface were obtained using electronic microscope-microanalyzer REMMA – 102-02 02 with electrons beam (electrons energy – 0.2-40 kV).

3. Results and discussion

The amorphous state of the similar AMA-electrodes was confirmed by x-ray method on diffractometer DRON-3.0M (CuK_α radiation, 2θ range 10° – 140° , step 0.04° , 1 s) in [22].

We investigated corrosion resistance of the bulk AMA $\text{Fe}_{60}\text{Co}_7\text{Cr}_4\text{V}_2\text{W}_1\text{Mo}_1\text{Nb}_1\text{B}_{20}\text{C}_2\text{Si}_2$ (HB-4). It is known that presence of chromium in alloys significantly improves passivity [23]. Chromium forms a dense defect-free films of hydrate oxohydroxide ($\text{CrO}_x(\text{OH})_{3-2x}\cdot n\text{H}_2\text{O}$), which become a diffusion barriers for aggressive ions to the metal surface. Niobium stabilizes nanostructure of alloys, additions of vanadium and tungsten increases microhardness. However, widespread using of optimized HB-4 requires experimental confirmation of its corrosion resistance.

Analysis of measured by chronopotentiometric method in 0.5 M NaCl aqueous solution electrochemical characteristics of tape and bulk samples of HB-4 showed that potential values of both sides of tape is more positive than values of free potential of bulk sample. Tape of HB-4 has better corrosion resistance compared to bulk alloy with similar elemental composition (Table 1).

Table 1

Results of the chronopotentiometric investigation of HB-4 in different solutions						
Environment	Sample	Side	$-E_0$, V	$-E_p$, V	ΔE , V	$v \cdot 10^4$, V/s
0.5 M NaCl	Tape	c	0.39	0.40	0.01	3.33
		e	0.27	0.38	0.11	6.33
	Bulk		0.37	0.46	0.09	5.21
0.5 M KOH	Tape	c	0.32	0.27	0.05	8.00
		e	0.40	0.41	0.01	6.67
	Bulk		0.78	0.82	0.04	6.67
0.5 M HCl	Tape	c	0.20	0.23	0.03	1.67
		e	0.48	0.40	0.08	1.67
	Bulk		0.42	0.50	0.08	5.00

A similar regularity was observed in alkaline medium (Table 1). In acidic environment contact side of the HB-4 tape is also more stable, and potential values of external surface of the tape and bulk sample are similar (Table 1).

Obtaining features of tape and bulk samples cause difference in elemental composition of surfaces of AMA with different shape, which determines electrochemical activity in aqueous corrosive media [24].

In the case of tape samples by potential scanning can be recorded a sequence of individual stages of dissolution and passivation of the HB-4 surface. Composition heterogeneity of the bulk sample surface causes parallel passing of different stages simultaneously. Such heterogeneity is caused by slower hardening of the melt in the obtaining process of bulk alloy compared to tape sample and higher probability of clustering and phase regrouping in the bulk samples. This causes separation of mechanisms of metallic components oxidation and ions emission in solution.

Determined by energydispersive analysis content of metallic components of HB-4 with different shape presented in Table 2. On the bulk sample surface can be seen the dark phase (Fig. 1), enriched by Fe and Co compared to charge composition. Light phase contrariwise enriched by Cr, Y, W, Mo i Nb.

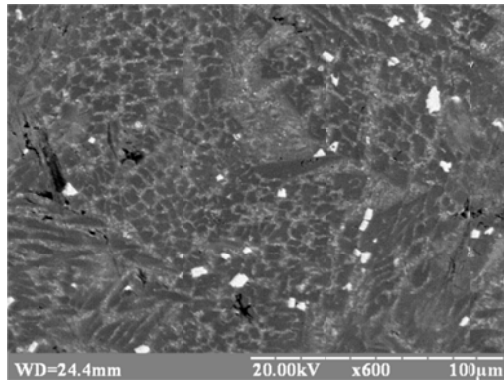


Fig. 1. Microphoto of the bulk AMA $\text{Fe}_{60}\text{Co}_7\text{Cr}_4\text{V}_2\text{W}_1\text{Mo}_1\text{Nb}_1\text{B}_{20}\text{C}_2\text{Si}_2$.

Table 2

Metallic components content on the surface of samples of HB-4 (mass %)

Sample	Phase	Element						
		Fe	Co	Cr	V	W	Mo	Nb
Charge composition	–	60.00	7.00	4.00	2.00	1.00	1.00	1.00
Tape	–	72.26	8.97	5.17	0.01	0.18	2.38	0.00
Bulk	Dark	76.27	12.22	4.84	0.96	1.99	3.28	0.42
	Light	33.10	4.71	12.73	7.68	15.36	18.08	8.30

According to cyclic voltammetry in conditions of forced overlapping of external potential within (-1.5 ± 0.5) V it can be argued that the electrochemical properties of the alloy HB-4 in the form of tape and bulk sample in 0.5 M NaCl aqueous solution are close (Fig. 2, Table 3). However, the form of cyclic voltammograms is significantly different, it shows different mechanisms of samples interaction in aggressive media regardless from the cycle number (Fig. 2).

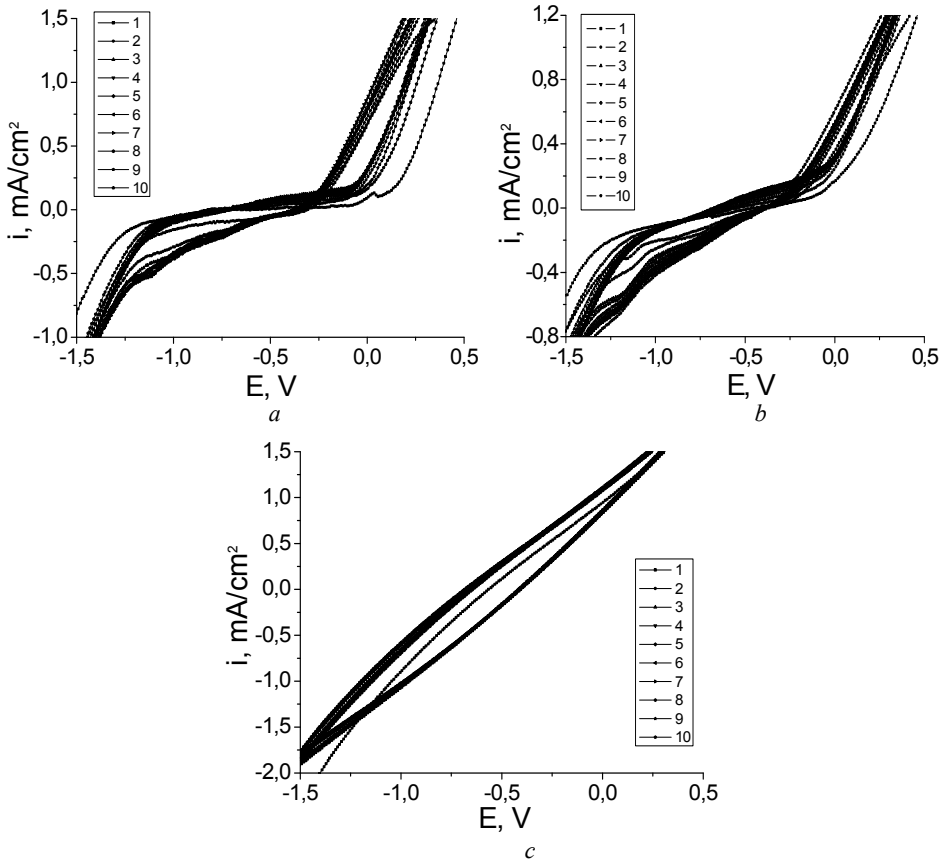


Fig. 2. Cyclic voltammograms of contact (a), external (b) surfaces of tape and bulk sample (c) of AMA HB-4 in 0.5 M aqueous solution of NaCl.

In contact with the aggressive environment of 0.5 M sodium chloride and multiple cyclic potential scanning, corrosion resistance of tape and bulk sample reduced and after the 5th cycle stabilized (Fig. 3). The corrosion currents of bulk alloy increased already during the 2nd cycle and practically stabilized after the 5th cycle.

Table 3

Electrochemical characteristics of AMA HB-4 in 0.5 M aqueous solution of NaCl

Cycle	Tape AMA				Bulk AMA	
	Contact side		External side		E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²
	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²		
1	-0.65	0.02	-0.45	0.12	-0.55	0.26
2	-0.70	0.01	-0.54	0.02	-0.64	5.32
3	-0.74	0.07	-0.59	0.11	-0.65	3.31
4	-0.74	0.7	-0.62	0.04	-0.66	6.32
5	-0.77	0.21	-0.65	0.10	-0.66	5.39
6	-0.72	0.24	-0.66	0.08	-0.67	5.72
7	-0.72	0.03	-0.66	0.13	-0.67	7.86
8	-0.70	0.15	-0.67	0.08	-0.69	6.75
9	-0.74	0.21	-0.66	0.27	-0.69	5.73
10	-0.72	0.06	-0.67	0.02	-0.69	1.29

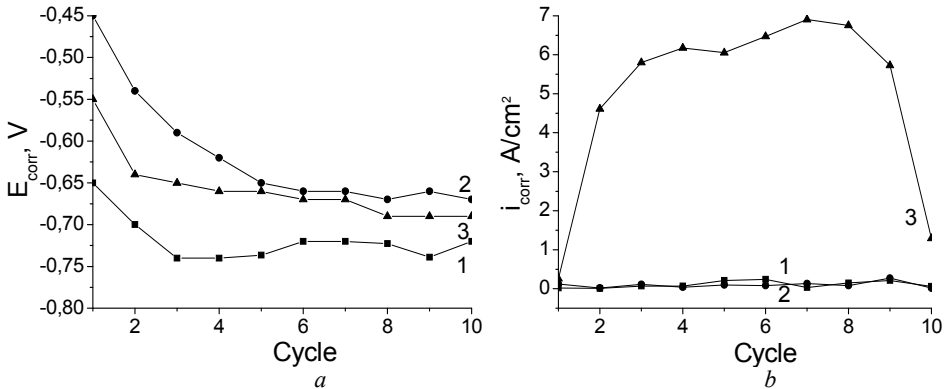


Fig. 3. Change of corrosion potentials (a) and corrosion currents density (b) of AMA HB-4 with different shape (1 – contact, 2 – external surface of tape, 3 – bulk sample) in 0.5 M solution of NaCl.

Comparison of the values of corrosion currents and corrosion potentials of AMA HB-4 with different shape in 0.5 M aqueous solution of potassium hydroxide presented in Table 1 shows that the tape sample is more stable in this aggressive environment than a bulk sample with the same elemental composition. These was confirmed by the lower values of corrosion currents and more positive corrosion potentials of the tape sample (Table 4, Fig. 4). As a result of prolonged contact with aggressive environment corrosion potentials move in the cathodic side, and the corrosion currents increase slightly, this confirms the slight acceleration of oxidative dissolution of investigated alloy in this aggressive environment.

Similar investigations were carried out in 0.5 M aqueous solution of HCl. The results are presented in Table 5 and on Fig. 5. After analyzing the experimental data of cyclic

voltammetry it can be argued that in this environment the contact side of AMA HB-4 shows higher corrosion resistance than external side and bulk sample (Table 5, Fig. 5).

It was found that hydrochloric acid is the least aggressive environment for samples of AMA with such elemental composition, corrosion potentials moves in anodic side and corrosion currents are lower compared to the characteristics of the electrochemical dissolution in 0.5 M solutions of NaCl i KOH.

Table 4

Cycle	Electrochemical characteristics of AMA HB-4 in 0.5 M aqueous KOH solution					
	Tape AMA				Bulk AMA	
	Contact side		External side		E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²
	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²
1	-0.72	0.15	-0.69	0.05	-0.92	2.82
2	-0.75	0.34	-0.72	0.14	-0.96	1.20
3	-0.74	0.19	-0.71	0.08	-0.96	0.50
4	-0.75	0.11	-0.72	0.10	-0.97	2.9
5	-0.75	0.28	-0.74	0.06	-0.97	2.84
6	-0.76	0.09	-0.75	0.03	-0.98	2.62
7	-0.73	0.32	-0.78	0.41	-0.98	0.80
8	-0.72	0.21	-0.81	0.80	-0.98	0.30
9	-0.72	0.16	-0.82	0.50	-0.98	0.52
10	-0.72	0.17	-0.83	0.59	-0.98	2.72

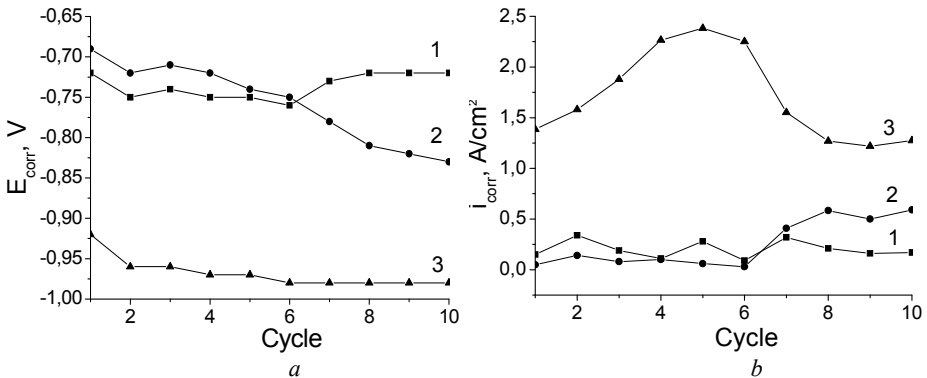


Fig. 4. Change of corrosion potentials (a) and corrosion current density (b) of AMA HB-4 with different shape (1 – contact, 2 – external surface of tape, 3 – bulk sample) in 0.5 M solution of KOH.

Table 5

Cycle	Tape AMA				Bulk AMA	
	Contact side		External side		E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²
	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²	E_{corr} , V	$i_{\text{corr}} \cdot 10^5$, A/cm ²		
1	-0.25	0.09	-0.45	0.12	-0.37	0.50
2	-0.28	0.08	-0.54	0.02	-0.39	0.45
3	-0.29	0.35	-0.59	0.11	-0.41	1.60
4	-0.29	0.29	-0.62	0.04	-0.41	0.55
5	-0.29	0.12	-0.65	0.10	-0.41	0.57
6	-0.29	0.21	-0.66	0.08	-0.41	1.35
7	-0.29	0.03	-0.66	0.13	-0.41	1.27
8	-0.29	0.17	-0.67	0.08	-0.41	0.75
9	-0.29	0.11	-0.66	0.27	-0.41	0.28
10	-0.29	0.55	-0.67	0.02	-0.41	0.27

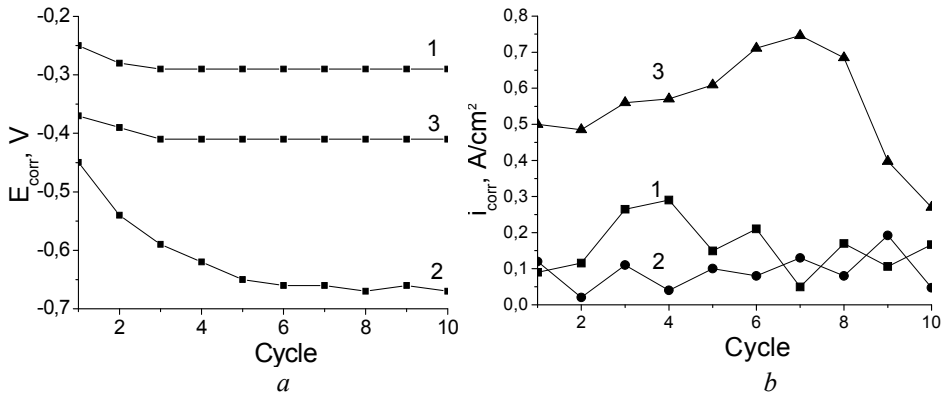


Fig. 5. Change of corrosion potentials (a) and corrosion current density (b) of AMA HB-4 with different shape (1 – contact, 2 – external surface of tape, 3 – bulk sample) in 0.5 M solution of HCl.

4. Conclusions

- Tape samples, especially the contact side, of AMA $\text{Fe}_{60}\text{Co}_7\text{Cr}_4\text{V}_2\text{W}_1\text{Mo}_1\text{Nb}_1\text{B}_{20}\text{C}_2\text{Si}_2$ are more corrosion resistant compared with bulk samples with the same elemental composition;
- On the surface of bulk alloys during hardening at lower cooling rates than in the process of obtaining tape samples appeared phases with different composition (dark and light) enriched with Fe and Co, or other metallic components;
- Selective dissolution of intermetallic phases in aggressive solutions leads to the formation of clusters with low reactivity, which during the further dissolution process increases. This assists formation of new phases due to the intra-cluster and inter-cluster diffusion initiated by vacancies formed of released ions;

- New phases can be formed from reactive atoms, this leads to passivation of the bulk samples surface after active dissolution.

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РЕЗЮМЕ

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ПОРІВНЯННЯ КОРОЗІЙНОЇ СТІЙКОСТІ ОБ'ЄМНОГО ТА СТРІЧКОВОГО АМОΡФНОГО $Fe_{60}Co_7Cr_4V_2W_1Mo_1Nb_1B_{20}C_2Si_2$ СПЛАВУ У РІЗНИХ АГРЕСИВНИХ СЕРЕДОВИЩАХ

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Об'ємні аморфні сплави представляють собою новий клас металевих сплавів з унікальною аморфною атомною структурою. Об'ємні аморфні сплави володіють корисними властивостями, які служать для їх широкого застосування. Навіть після значного прогресу в їх дослідженнях, практичне застосування об'ємних аморфних сплавів залишається, як і раніше складним завданням. Аморфні сплави мають унікальні фізико-хімічні властивості, серед них хороші механічні та магнітні властивості, а також високу корозійну стійкість в агресивних середовищах. У даній роботі автори порівнювали корозійну стійкість стрічкових і об'ємних зразків аморфного Fe-вмісного сплаву в 0,5 М водних розчинах NaCl, KOH і HCl. Було встановлено, що соляна кислота є найменш агресивним середовищем для обох зразків аморфного сплаву. Стрічкові зразки сплаву на основі Fe мають кращу корозійну стійкість в порівнянні з об'ємними зразками з таким же елементним складом.

Ключові слова: об'ємні та стрічкові аморфні сплави, корозійна стійкість, агресивне середовище, вольтамперометрія, потенціометрія

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