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PERSPECTIVE TRENDS OF CREATION AND APPLICATION: HIGH-ENTROPY GLASSES

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Abstract. One of the main tasks of modern physical materials science to develop and study high-entropy alloys of the latest generation is formulated. A brief review of recent publications on promising areas of creation and application of high-entropy alloys is given. A set of high performance characteristics is identified for high-entropy alloys for use in modern science-intensive industries: wear resistance, strength and impact strength, chemical, radiation and corrosion resistance, low density, super plasticity and superconductivity, high and low thermal conductivity, diffusion resistance, low temperature coefficient of resistance, environmental friendliness, etc. The areas of promising applications of high-entropy alloys in nuclear reactors, aerospace engines, gas and oil pipelines, offshore structures, computers and electronic devices are indicated. It is noted that many high-entropy alloys can be used in dual-use products. As an example, a proposal for the creation of thin-film high-resistive materials with a low temperature coefficient of resistance by the spinning method is considered. A tape made of the high-entropy Cantor's alloy of a non-equiatomic composition has been obtained and its properties have been studied. An assumption about the further development of high-entropy alloys has been made and substantiated.

Keywords: spinning, tape, Cantor alloy, structure, properties, deformation curve

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Оригинальная статья

ПЕРСПЕКТИВНЫЕ НАПРАВЛЕНИЯ РАЗРАБОТКИ И ПРИМЕНЕНИЯ: ВЫСОКОЭНТРОПИЙНЫЕ СПЛАВЫ

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Аннотация. Сформулирована одна из основных задач современного физического материаловедения по разработке и изучению высокоэнтропийных сплавов последнего поколения. Приведен краткий обзор последних публикаций по перспективным направлениям создания и применения высокоэнтропийных сплавов. Определен набор высоких эксплуатационных характеристик высокоэнтропийных сплавов для использования в современных наукоемких отраслях промышленности: износостойкость, прочность и ударная вязкость, химическая, радиационная и коррозионная стойкость, низкая плотность, сверхпластичность и сверхтекучесть, высокая и низкая теплопроводность, диффузионное сопротивление, низкотемпературный коэффициент сопротивления, экологичность и др. Указаны области перспективного применения высокоэнтропийных сплавов в ядерных реакторах, аэрокосмических двигателях, газо- и нефтепроводах, морских конструкциях, компьютерах и электронных устройствах. Отмечено, что многие высокоэнтропийные сплавы могут быть использованы в продукции двойного назначения. В качестве примера рассмотрено предложение по созданию тонкопленочных высокорезистивных материалов с низким температурным коэффициентом сопротивления методом спиннинга. Получена лента из высокоэнтропийного сплава Кантора неэквиатомного состава и исследованы ее свойства. Высказано и обосновано предположение о дальнейшем развитии высокоэнтропийных сплавов.

Ключевые слова: спиннинг, лента, сплав Кантора, структура, свойства, кривая деформации

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Introduction

One of the fundamental and practice-oriented problems of modern material science is a development of physical bases of creation of novel metallic materials and technologies of their production with a complex of the required physical – mechanical and operational characteristics. At the end of the XX century the first scientific papers on creation and integrated study of new so-called high-entropy alloys (HEAs) including up to 5 – 6 principle elements, each being in a large concentration (for instance, from 5 to 35 %) have appeared [1]. Along with characteristics typical of metallic alloys the materials possess the unique and unusual properties common to, for instance, metal ceramics.

Multicomponent alloys may be interesting in that they can be a basis for other compositions which will possess better properties than other alloys, especially those based on a single element. Metallic glasses (MG) based on high-entropy alloys (HEAs) may be isolated into a separate group due to the fact that the materials (MG, HEAs) have remarkable properties of both constituents – the high hardness and elasticity of MG and the high tensile ductility of HEAs [1].Therefore, a formation, crystallization and kinetics of the materials is a subject of a steady study nowadays.

Bulk amorphous alloys, so-called metallic glasses [2; 3] may be considered to be precursors of HEAs. The materials may also contain a large number of components in comparable concentrations, and on crystallization from a melt these form a single phase. But the phase is amorphous and metastable, existing only due to the fact that on rapid cooling atoms have no time to form a crystal structure and ‘solidify’ in a disordered state when their mobility decreases due to a temperature drop.

Due to the difference in atom sizes of various metals a crystal lattice of HEAs turns out to be severely distorted therefore a structure of such phases may be considered as an intermediate one between the stable crystal phase with a relatively little equilibrium concentration of defects, including impurity atoms and metastable metallic glasses wherein a long-range order is absent. In consequence of structural peculiarities HEAs are characterized by the small coefficients of diffusion, corrosion resistance, increased plasticity at low temperatures and other particular properties that may be very useful for many perspective materials and technologies [2].

For the last two decades about 10 000 scientific articles (according to databases Scopus and Web of science), numerous reviews [4 – 12], monographs [13 – 16] have been published on the topic of high-entropy alloys (HEAs).

The aim of the present research is to analyze the recent publications and to estimate the most perspective trends of creation and application of tape glasses from HEAs in different science – intensive branches of industry.

Results and Discussion

As it has already been noted in [12 – 14] one of the most attractive features of HEAs is the high strength and plasticity at lower and even cryogenic temperatures. It is also reported about the high fracture toughness of HEAs at low temperature, for instance, 232 MPa/m^{½} at 77 K. A set of the properties makes HEAs very perspective materials for application in conditions of the Arctic regions [2, 17].

The studies of magnetic properties carried out for HEAs based on ferromagnetic metals show a promising development of magnetosoft materials on their base [2] and also, the magnetic properties may be controlled by alloying, variation in stoichiometry and annealing [18 – 21]. The paramagnetic HEAs CuCrFeTiNi with a small fraction of a ferromagnetic phase has been obtained. The HEAs based on the rare-earth elements and the metal of iron group, for instance, GdTbDyAl*M* (*M* = Fe, Co, Ni), wherein a magnetocaloric effect [14] was discovered, are of a considerable interest.

The achievements of HEAs application mentioned above give no grounds for saying about the immediate prospects for replacing the traditional alloys in any branches of industry. For this it is necessary to develop the new compositions of HEAs, to study their properties in connection with the specific conditions of the practical application. The trends in this field revealed for the recent two decades indicate that the leading development of this material class will be only intensified. For this time being, there goes an accumulation and conceptualization of a large volume of information [15; 16]. A development of HEAs compositions and their study should be accompanied by a creation of a new technologies and processes of production of unique alloys.

In addition to numerous techniques in [15; 16] these should provide a heat and mechanical treatment, high-speed crystallization, superplastic molding by spraying, equichannel angle extrusion and other types of severe plastic deformation, friction stir welding as well as combination of these [13]. In this connection it is necessary to take into account a technological production effectiveness of the processes, an accessibility of raw materials and possibility of recycling. From the commercial and engineering points of view the main considerations should be the cost criteria of finished products [22] besides a high level of properties.

By the present time the area of material properties requiring higher values has already been stabilized. These are first of all: the wear resistance, strength and impact toughness, resistance to temperatures, chemical, corrosion, radiation resistance, low density, superplasticity and superelasticity, magnetic properties, superconductivity and low temperature coefficient of resistance, high and low heat conductivity, resistance to diffusion, environmental friendliness, functional and biological properties, etc. It is apparent, this enumeration is far from being complete. A rapid development of science-intensive branches stimulates a development of principally new materials on the basis of miniaturization, multifunctionality, environmental friendliness.

Among the perspective applications already now requiring the novel and improved materials are [23]: materials for surfacing, materials for nuclear constructions, materials for avia and spacecraft engines, chemical pipelines, sea vessels and off-shore structures, high-frequency communication materials and computers, materials to store hydrogen, thermoelectric materials and superconductors, light-weight transport materials, electrical and magnetic materials for precision electric and electronic devices. The series should include the materials of dual purpose.

For a detailed consideration of the perspective trends it is necessary to have at least the experience of research in the corresponding trend.

In our research generalized in monographs [15; 16]:

1. The mechanical properties of the Co – Cr – Fe – Ni – Mn HEAs under compression and tension, obtained at different regimes of a deposition complex, are determined. It enabled us to reveal a regime providing the best combination of strength and plasticity. In compression tests the conventional yield strength amounted to 279 MPa, the fracture strength 1689 MPa, the relative deformation 54 %, and in tension tests – 279 MPa, > 500MPa and > 75 %, respectively. The wear parameter amounted to $2.9\cdot10^{-4}$ mm^3/(N·m), the friction factor 0.62. A fracture surface has a tough pit character.

2. The electron beam processing (EBP) of the Co – Cr – Fe – Ni – Mn HEAs with the energy density of E_s = 10 j/cm^2 is accompanied by a primary recrystallization with a formation of grains 1.5 – 3 μm in size. At E_s = (15 – 30) j/cm^2 a process of the collective recrystallization is developed and is accompanied by a growth of grain medium size from 35 to 120 μm. The high-speed crystallization cells being formed in a bulk of grains increase from 310 nm at E_s = 15 j/cm^2 to 800 nm at E_s = 30 j/cm^2. The scalar dislocation density decreases nonmonotonously to 130 μm with a greater distance from the irradiation surface. A type of dislocation substructure varies from a nondisoriented cellular to chaotic through nondisoriented cellular-net-like one. The bend extinction contours are absent.

3. The EBP is accompanied by a precipitation of the nanodimensional (1 – 3 nm) particles of round shape on dislocations. The increase in E_s under tension tests results in a decrease in the plasticity by more than 2 times, the hardness by 1.3 times, the microhardness by 1.6 times relative to the initial state. The material regions whose failure occurred with a formation of a the band (lamellar) structure are detected. The area of the lamellar-structure fracture increases from 25 % at E_s = 10 J/cm^2 to 65 % at E_s = 30 J/cm^2, which may be one of the reasons for

Fig. 1. A roll foil 70 μm thick from the $Al_5Cr_{12}Fe_{35}Mn_{28}Ni_{20}$ alloy obtained by arc melting and cold rolling (the elongation in rolling is 4.257 % depending on a blank with the initial thickness of ~ 3 mm and hardness of 147 HV [13; 23])
Рис. 1. Рулонная фольга толщиной 70 мкм из сплава $Al_5Cr_{12}Fe_{35}Mn_{28}Ni_{20}$, полученная дуговой плавкой и холодной прокаткой (удлинение при прокатке составляет 4,257 % для заготовки с исходной толщиной примерно 3 мм и твердостью 147 HV [13; 23])

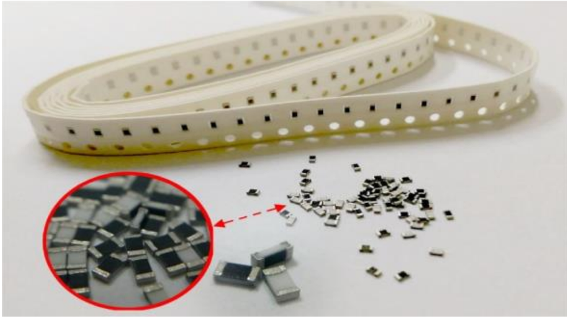

Fig. 2.Thin-film resistors fabricated from a HEA thin tape [24]
Рис. 2. Тонкопленочные резисторы, изготовленные из тонкой ленты ВЭС [24]

reduction of strength and plasticity of a material in the irradiated state.

As the examples of HEAs application in the form of thin bands, foils and tapes the Al – Cr – Fe – Mn – Ni and Ni – Cr – Si – Fl – Ta alloys (Fig. 1) [13; 23] may be given.

Fig. 1 shows a foil 70 μm thick subjected to cold rolling with the elongation of 4.257 % without any cracking on edges [13; 23]. The curve of strengthening in processing shows that its hardness approaches the saturation of about 360 HV under a large deformation. In this state a foil still may be folded without a crack formation. This indicates that the ability of the alloy to bending is excellent. This readily rolled alloy may be used potentially as flexible substrates for solar cells and displays.

In many branches of electronic industry one of the principal components of electronic circuits is thin-film high-ohm Ni – Cr resistors (Fig. 2) to which the decisive requirements is the low-temperature resistance coefficient.

In the paper [24] the Ni – Cr – Si – Al – Ta HEA thin film resistors obtained by spraying on the Al_2O_3 substrate at a room temperature have been studied in order to produce thin films with a high specific resistance and a low temperature resistance coefficient. Different procedures of annealing in air were performed at various temperatures from 250 °C to 500 °C. The authors have discovered that the $Ni_{23.5}Cr_{14.6}Si_{23.6}Al_{16.8}Ta_{21.5}$ sprayed films after annealing in air at 300 °C showed the least temperature resistance coefficient, 10 parts per million/°C, and the higher specific resistance, 2200 Ohm/cm.

Metallic glasses based on HEAs may become an alternative for production and application of thin tapes and foils [25] in different areas. The studies [26 – 30] which appeared in recent time indicate the principal possibility of producing the metallic glasses from HEAs. However, these are bulk metallic glasses [31 – 35].

The information on amorphous tapes from HEAs is practically absent [25]. The metallic glasses in the form of tapes are usually produced by a spinning method known as early as 60's of the last century.

A melt is placed into a quartz nozzle which is heated by means of inductor. Under the action of a small excess pressure a melt is fed to a fast-revolving copper drum-cooler through a narrow opening in a nozzle with the result that a thin jet of a melt turns into a solid state in the form of a tape from 30 to 100 μm thick. The drum is cooling with a running water of temperature 8 °C. A specific character of the melt consists in a 'abrupt quenching' of a melt at a rate of 10^6 deg/sec [2].

The process fits splendidly for production of long thin tapes practically of any composition of alloys. A speed of drum revolution can be controlled to obtain a maximum effectiveness of the tape production process.

For the Cantor's CoCrFeNiMn and CoCrFeNiAl HEAs of nonequatomic compositions corresponding to earlier ones studied in [2; 15; 16] the tape ~ 80 μm thick were produced. A satisfactory state of tapes has been obtained only for the Cantor HEA alloyed with aluminum at present time (Fig. 3). Technical parameters of HEA casting (spinning) are: the temperature of melt – 1480 °C; the linear speed of a quenching drum – 32.6 m/sec; the value of extrusion (excess) pressure – 0.3 – 0.35 atm; the size of nozzle – 0.7 × 20 mm; the gap between a nozzle and a drum – 0.27 mm.

The parameters of tape of Cantor's HEA alloyed with aluminum, of nonequiatomic composition has been studied by the methods of modern physical material science.

By the methods of micro-X-ray spectral analysis it has been established that the alloy under study has the following composition (at. %): Al – 9.6; Cr – 12.2; Fe – 39.6; Co – 12.5; Ni – 24.3; Mn – 2.0.

That is it can be referred to the HEA of a nonequiatomic composition. The micro-X-ray spectral

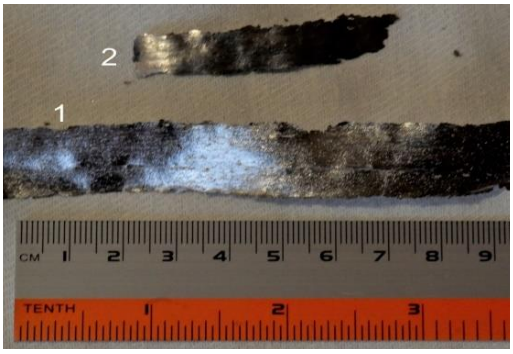

Fig. 3. The CoCrFeNiMn HEA tape after quenching (1) and after rolling (2)
Рис. 3. Лента ВЭС CoCrFeNiMn после закалки (1) и после прокатки (2)

analysis of the alloy performed by the methods 'by points' has detected a slight variation in a tape composition when passing from one area of scanning to another (Fig. 4; table). When analyzing the results presented in table it can be noted that the most liquating elements in the tape under study are aluminum and manganese i.e. chemical elements with a comparatively (relative to other elements of alloy) low temperature of melting.

The tape under study is a two-phase material. By the method of X-ray structural analysis (a diffractometer XDD-6000, CuKα – radiation) we have revealed a solid solution based on iron (bcc crystal lattice) with the parameter *a* = 0.28681 nm and a compound Co0.8Cr0.8Fe0.8Mn0.8Ni0.8 (fcc crystal lattice) with the parameter *a* = 0.35859 nm. A relative content of phases is 82 mass % and 18 mass %, respectively.

The microhardness of the HEA tape determined on device PMT 3 under indenter load of 0.5 is 367 HV.

Conclusion

A shot review of published scientific papers of recent years on perspective areas of creation and application of high-entropy alloys in different science-intensive branches of industry has been carried out. A complex of high operational properties required for application in aerospace engines, nuclear reactors, seagoing vessels, gas –and oil pipelines, electronic devices and computers demanded by industry has been analyzed.

The attention is paid to the problems of creating the thin-film high-resistive materials with the low temperature coefficient of resistance.

The HEA-based metallic glasses are proposed to be used for the purposes. The tape ~ 80 μm thick from the Cantor's HEA of nonequiatomic composition has been produced by the method of spinning and its properties have been studied.

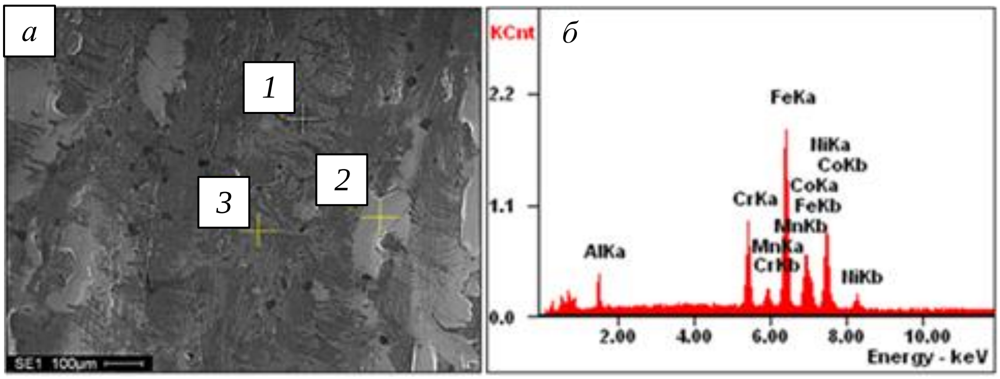

Fig. 4. Electron microscopic image of the HEA tape (*a*); *б* – energy spectra corresponding to the area designated by digit 2 in *a*
Рис. 4. Электронно-микроскопическое изображение ВЭС-ленты (*а*); *б* – энергетические спектры, соответствующие области, обозначенной цифрой *2* на поз. *а*

Results of the micro-X-ray spectral analysis performed by the method «by points» (areas of the analysis *1, 2, 3* are shown in Fig. 4, *a*)
Результаты микрорентгеноспектрального анализа, выполненного методом «по точкам» (области анализа *1, 2, 3* показаны на рис. 4, *а*)

Element	area *1*		area *2*		area *3*	
	w.%	at. %	w.%	at. %	w.%	at. %
Al(Kα)	06.26	12.28	04.99	09.92	05.29	10.47
Cr(Kα)	11.46	11.67	11.42	11.77	11.74	12.06
Mn(Kα)	01.67	01.61	01.90	01.86	02.01	01.96
Fe(Kα)	40.09	37.98	40.73	39.10	40.72	38.95
Co(Kα)	12.95	11.62	13.80	12.56	13.54	12.27
Ni(Kα)	27.56	24.83	27.15	24.80	26.70	24.29

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