

Application of High-Entropy Alloys

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Abstract—From the accumulated data on the structure, properties, stability, methods for obtaining high-entropy alloys (HEAs), created at the beginning of the 21st century, a whole range of useful properties were discovered, which allows for promising usages in various industries. A brief review of the literature from the last five years on the analysis of using HEAs in specific knowledge-intensive industries is carried out. In biomedicine, protective coatings made of HEAs (TiZrNbHfTa)N and (TiZrNbHfTa)O possess biocompatibility, a high level of mechanical properties, high wear and corrosion resistance in physiological environments, and excellent adhesion. (MoTa)_xNbTiZr products have successfully passed clinical trials when implanted in living muscle tissue. The developed HEAs based on rare earth elements and metals of the Fe group of the YbTbDyAlMe type (*Me* = Fe, Co, Ni) have a magnetocaloric effect, have the Curie point close to room temperature, and can be used in modern refrigeration devices. By changing the stoichiometric composition of the CoCrFeNiTi HES, alloying them and conducting heat treatment, it is possible to obtain soft magnetic materials. The application fields of HEAs as catalysts for ammonia oxidation (PtPdRhRuCe), decomposition of ammonia (RuRhCoNiIr), oxidation of aromatic alcohols (Co_{0.2}Ni_{0.2}Cu_{0.2}Mg_{0.2}Zn_{0.2}), electrocatalysts for the evolution of hydrogen (Ni₂₀Fe₂₀Mo₁₀Cr₁₅CuTi), redox reactions (AlCuNiPtMn and AlNiCuPtPdAu), and methanol/ethanol oxidation are considered. HEAs can be used as electrodes-anodes and cathodes for Li-ion and Na-ion batteries. Synthesized nanoporous AlCoCrFeNi HES have a high bulk density (up to 700 F/cm³) and cyclic stability (>3000 cycles) and are used in supercapacitors. High-entropy oxides of the (MgNiCoCuZn)_{0.95}Li_{0.05}O type with high dielectric properties in a wide frequency range can be used in electronic converters. Examples of HEAs application as coatings for parts of watercrafts operating in sea water, dissimilar welded joints—parts of nuclear reactors—are given. Prospects for expanding the areas of HEAs application are noted.

Keywords: high-entropy alloys, application, biomedicine, energy, catalysts, magnetocaloric effect

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INTRODUCTION

High-entropy alloys (HEAs) as a class of new materials appeared at the beginning of the 21st century. They contain up to five to six elements at a concentration of 5–35% [1, 2]. By changing the elemental composition, it is possible to form a high level of such useful properties as high-temperature strength, superparamagnetism, corrosion resistance, and high hardness (along with plasticity and many other properties) [3–6]. If we consider that there are about 80 metals in nature, more than 107 combinations of 5-element HEAs can be made. Then, the set of useful properties of such HEAs is very large. Unique characteristics will

significantly expand the scope of HEAs [7]. This is, first of all, the use of HEAs for the production of cutting tools, dies, magnetron sputtering targets, diffusion barriers in microelectronics, parts of nuclear power equipment, cryogenic technology, aerospace industry, etc. [8–17]. However, the results presented in these works are, at most, five years old. In the last five years, there has been an even more intensive accumulation of information about the structure, stability, methods for obtaining, and the prospects for the practical application of HEAs. It is still too early to talk about the real global practical use of HEAs, but an analysis of the latest literature data indicates a positive

trend in the possible use of HEAs in various science intensive industries.

In this work, a brief review of recent domestic and foreign studies of the possibilities of practical HEAs application is carried out.

BIOMEDICINE

Biomedicine is one of the promising fields of HEA application for nitride- and carbide-based coatings [7]. If the main requirements for HEAs are biocompatibility and high mechanical properties, then protective coatings must additionally have high chemical stability, wear and corrosion resistance in physiological environments, and strong adhesion to a deposited surface. (TiZrNbHfTa)N and (TiZrNbHfTa)O coatings [18] have similar properties, which do not cause cytotoxic reactions on osteoblasts. Studies of medium-entropy TiZrNbMo alloys with titanium content up to 65% made it possible to successfully test them [19].

A comprehensive study of the (MoTa)_xNbTiZr alloy has shown that products made from it have excellent plastic, strength and anti-corrosion properties. In the carried-out vivo tests (tests inside a living organism) of products from this alloy, they were implanted in muscle tissue for four weeks. A marked passive behavior in the phosphate buffered saline and a mild, non-toxic response of muscle tissue were revealed [20].

HEAS WITH SPECIAL MAGNETIC PROPERTIES

Varying alloying, stoichiometric composition (ratio of Co/Cr, Fe/Cr, Ni/Cr) and heat treatment (annealing for 2–10 h at 200 and 700°C) of HEAs (Co₃₅Cr₅Fe₂₀Ni₂₀Ti₂₀, Co₂₀Cr₅Fe₂₀Ni₃₅Ti₂₀) allows the development of soft magnetic-based materials [21, 22]. In this case, HEAs with an fcc lattice have a high saturation magnetization, in contrast to alloys with a bcc lattice [7], which is due to a higher atomic packing density and a high content of ferromagnetic elements (Fe, Co, Ni).

Of special interest are HEAs based on rare earth elements and metals of the iron group, such as YbTbDyAlMe (Me = Fe, Co, Ni) with a magnetocaloric effect [23], which manifests itself in a reversible change in the temperature of the magnetic material with a change in the magnetic field. Magnetic cooling is based on this effect. For HEAs of transition metals of the Mn_xCr_{0.3}Fe_{0.5}Co_{0.2}Ni_{0.5}Al_{0.3} (20.8 < x < 1.1) types with a magnetocaloric effect, the Curie temperature approaches room temperature, which makes them extremely efficient in modern refrigeration units [24].

HEAS IN THE POWER INDUSTRY

In works from Chinese researchers [25], theoretical and experimental results on the structure, properties, methods of obtaining HEAs with an emphasis on

energy are analyzed. The results are summarized in Table 1. Let us take a look at some of the highlights.

Catalysis

Decomposition of ammonia. Compared with traditional Co–Mo and Ru catalysts Co_xMo_yFe₁₀Ni₁₀Cu₁₀ (x + y = 70) and RuRhCoNiIr catalysts have more than tenfold efficiency. Such outstanding catalytic performance and high stability are due to the synergistic effect of ultrafine particle size, uniform dispersion, multi-element composition and fcc structure.

Oxidation of aromatic alcohols. A mesoporous HEA Co_{0.2}Ni_{0.2}Cu_{0.2}Mg_{0.2}Zn_{0.2}O provides ultra-high catalytic activity of aerobic oxidation of benzyl alcohol with 98% conversion, achieved in 2 h.

H₂ evolution reaction (HER). Compared to two-phase catalysts, HEAs have increased corrosion resistance. In addition to those listed in Table 1, it should be noted the highly efficient highly entropic oxide (FeMgCoNi)O_x (x ≈ 1.2) with a complex structure from a mixture of a simple cubic lattice and spinel.

O₂ evolution reaction (OER). Nanoporous catalysts made of HEAs AlNiCoFeMe (Me = Mo, Nb, Cr), developed from NiFe or NiCoFe nanoalloys, have high electrochemical stability.

In recent years, there has been an intensive study of the possibilities of creating electrocatalysts from double and ternary alloys without the use of noble metals. However, the further use of such electrocatalysts is limited due to their poor corrosion resistance. The authors of [26] obtained a highly efficient porous HEA CoCrFeNiMo by a fundamentally new microwave sintering method: the excess potential reaches 220 mV with a current density of 10 mA/cm². This is due to the ability of the porous structure to provide electronic transfer. High entropy oxide films (FeCrCoNiAl_{0.1})O_x obtained by magnetron sputtering provided an excess potential of 381 mV and electrolytic stability for 120 h in an alkaline solution at a current density of 10 mA/cm² [27].

Oxidation-reduction reaction (ORR). Nanoporous platinum-based catalysts (AlCuNiPtMn and AlNiCuPtPdAu) have high temperature stability (up to 600°C) and redox activity up to ten times higher than the characteristics of Pt/C catalysts.

Methanol/ethanol oxidation reaction. Synthesized HEAs (Ir_{0.19}Os_{0.22}Re_{0.21}Rh_{0.20}Ru_{0.19} and Ir_{0.2}6Os_{0.05}Pt_{0.31}Rh_{0.23}Ru_{0.15}) have exceptional activity and demonstrate high thermal stability at 1500 K.

Energy Storage

Electrode materials for lithium and sodium ion batteries. For lithium-ion batteries, materials based on high-entropy oxides can be used as anodes and cathodes. A cell of the anode (Co_{0.2}Cu_{0.2}Mg_{0.2}Ni_{0.2}Zn_{0.2})O and LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂ cathode provided an initial

Table 1. Scopes of HEA application in power engineering [25]

Application	Compound	Structure
Oxidation of ammonia	PtPdRhRuCe, PtCoNiFeCuAu, PtPdCoNiCuAu, PtPdCoNiFeCuAuSn	FCC
Decomposition of ammonia	$\text{Co}_x\text{Mo}_y\text{Fe}_{10}\text{Ni}_{10}\text{Cu}_{10}$ ($x + y = 70$)	FCC
H_2 evolution reaction (HER)	CoFeLaNiPt	FCC
	PtAuPdRhRu	FCC
	IrPdPtRhRu	FCC
	AlMoCuPdAu	FCC
	FeCoPdIrPt	FCC
	$\text{Cr}_{15}\text{Fe}_{20}\text{Co}_{35}\text{Ni}_{20}\text{Mo}_{10}$	FCC
	$(\text{FeMnCoNi})_x\text{O}$	Cubic + Spinel
	$(\text{CrMnFeCoNi})_x\text{P}$	Hexagonal
O_2 evolution reaction (OER)	CrMnFeCoNi	FCC
	AlMoCuPdAu	FCC
	CoFeLaNiPt	FCC
	AlMoCoIrMo	FCC
	$(\text{FeMnCoNi})\text{O}_x$ ($x \sim 1.2$)	Cubic + Spinel
	$\text{FeMnCoNi} + (\text{FeMnCoNi})\text{O}_x$	FCC
	$(\text{CrMnFeCoNi})_x\text{P}$	Hexagonal
	$\text{K}(\text{MgMnFeCoNi})\text{F}_3$, $\text{K}(\text{MgMnCoNiZn})\text{F}_3$	Perovskite
Reduction oxidation reaction (ORR)	CrMnFeCoNiNb, CrMnFeCoNiMo	FCC
	AlNiCuPtPdAu	FCC
	AlCuNiPtMn	FCC
Methanol oxidation reaction	$\text{Ir}_{0.19}\text{Os}_{0.22}\text{Re}_{0.21}\text{Rh}_{0.20}\text{Ru}_{0.19}$	FCC
	AlMoCuPdAu	FCC
Ethanol oxidation reaction	RuRhPdOsIrPt	FCC
Water oxidation	$(\text{Co}, \text{Cu}, \text{Fe}, \text{Mn}, \text{Ni})_3\text{O}_4$	Spinel
Oxidation of aromatic alcohols	$(\text{Mg}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2}\text{Cu}_{0.2}\text{Zn}_{0.2})\text{O}$	Cubic
Li-ion battery	$(\text{Co}_{0.2}\text{Cu}_{0.2}\text{Mg}_{0.2}\text{Ni}_{0.2}\text{Zn}_{0.2})\text{O}$	Cubic
	$(\text{MgCoNiZn})_{1x}\text{Li}_x\text{O}$ ($x = 0.05; 0.15; 0.25; 0.35$)	Cubic
	$(\text{Mg}_{0.2}\text{Ti}_{0.2}\text{Zn}_{0.2}\text{Cu}_{0.2}\text{Fe}_{0.2})_3\text{O}_4$	Spinel
	$(\text{Mg}, \text{Ti}, \text{Zn}, \text{Cu}, \text{Fe})_3\text{O}_4$	Spinel
	$[(\text{Bi}, \text{Na})_{1/5}(\text{La}, \text{Li})_{1/5}(\text{Ce}, \text{K})_{1/5}\text{Ca}_{1/5}\text{Sr}_{1/5}]\text{TiO}_3$	Perovskite
	$\text{NaNi}_{0.12}\text{Cu}_{0.12}\text{Mg}_{0.12}\text{Fe}_{0.15}\text{Co}_{0.15}\text{Mn}_{0.1}\text{Ti}_{0.1}\text{Sn}_{0.1}\text{Sb}_{0.04}\text{O}_2$	O3
Sodium-ion battery	FeNiCoMnMg	FCC
Supercapacitors	AlCoCrFeNi	FCC
	$(\text{TiNbTaZrHf})\text{C}$	FCC
	$(\text{CrMoVZrNb})\text{N}$	FCC
	$(\text{Mg}, \text{Co}, \text{Ni}, \text{Cu}, \text{Zn})_{1x}\text{Li}_x\text{O}$	Cubic

capacity of 446 (mA/h)/g and 256 (mA/h)/g after 100 cycles. Sodium-ion batteries showed 83% capacity retention after 500 cycles.

Supercapacitors. A synthesized nanoporous HEA AlCoCrFeNi, used as an electrode, has a high capacity (700 F/cm³) and cyclic stability (>3000 cycles).

Dielectric materials. Due to polarization in an external electric field, high-entropy dielectrics can be used in capacitors and powerful electronic converters. Highly entropy oxides of the (MgNiCoCuZn)_{0.95}Li_{0.05}O type have high dielectric properties in a wide (100 Hz–2.3 MHz) frequency range.

One of the promising areas of HEA application is the shipbuilding industry [28]. Valves, pumps, shafts, screws and other mechanisms operating in seawater are subject to corrosion and wear. Polymeric and ceramic materials are used as protective coatings, which are not perfect, in particular, ceramic coatings are fragile, and polymer ones have unstable dimensions. The developed and tested coating made of an AlCrFeNiW_{0.2}Ti_{0.5} HEA has a high hardness (~692 HV) and increased tribological properties [28]. HEAs AlCoCrFeNiCu and AlCoCrFeNiTi, which have increased tribological properties at high temperatures, have prospects for application in the aerospace industry [29].

Welding production also falls into a wide range of HEA applications. In modern nuclear reactors, there are a significant number of welded joints made of dissimilar materials. They are subject to high requirements for high-temperature (up to 1025 K) structural stability, anti-corrosion and mechanical properties. In the opinion of the authors of [30], the solution to the problem is possible when using joints made of a Cantor HEA (CoCrFeMnNi) and duplex stainless steel obtained by laser welding. There were attempts made to create high-entropy refractory superconductors [31, 32], superconductors based on rare-earth elements [33]. Other areas of possible use of HEAs are summarized in monographs and reviews [2, 23, 34–36]. There is every reason to believe that the fields of HEAs application will expand along with the development and creation of new compositions and the study of their properties.

CONCLUSIONS

The brief review of the works of domestic and foreign researchers over the past five years on the use of HEAs in various science-intensive industries was carried out. The promising areas of application are: biomedicine; materials with special magnetic properties, including those with a magnetocaloric effect; shipbuilding; aerospace industry; welding; and superconductors. The application of HEAs in the energy-related industries was analyzed in detail. The forecast is made for the expansion of HEAs along with the creation of new alloys and their research.

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