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High Entropy Alloys: Structure, Mechanical Properties, Deformation Mechanisms and Applications

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Abstract—A brief review of publications by foreign researchers on the study of the structure, phase composition and properties of five-component high-entropy alloys (HEAs) in different structural states in a wide temperature range over the past two decades has been made. HEAs attract the attention of scientists with their unique and unusual properties. Difficulties in conducting a comparative analysis and summarizing data due to different methods for obtaining HEAs, modes of mechanical tests for uniaxial compression and tension, sample sizes and shapes, types of heat treatments, and phase composition (BCC and FCC lattices) are noted. It is noted that HEAs with the BCC lattice have predominantly high strength and low ductility, while HEAs with the FCC lattice have low strength and increased ductility. It is shown that a significant increase in the properties of HEA FeMnCoCrNi with the FCC lattice can be achieved by doping with boron and optimizing the parameters of thermomechanical treatment when doping with carbon in an amount of 1% (at %). The deformation curves analyzed in the temperature range of –196...800°C indicate an increase in the yield strength with a decrease in the grain size from 150 to 5 μm. As the temperature decreases, the yield strength and relative elongation increase. The effect of the deformation rate on the mechanical properties consists in an increase in the tensile strength and yield strength, which is most noticeable at high rates of 10^{-2}–10^{3} s^{-1}. The features of the deformation behavior of HEAs in single- and polycrystalline states are noted. The complex of high operational properties of HEAs provides the possibility of their application in various industries. The prospects for using energy treatments to modify surface layers and further improve the HEAs properties are noted.

Keywords: high-entropy alloys, structure, mechanical properties, dislocations, twinning

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INTRODUCTION

One of the fundamental and important practical aspects of condensed matter physics and physical materials science is the development of physical foundations for the creation of new metallic materials and technologies for their production with a set of necessary physical, mechanical and operational characteristics. At the very end of the 20th century, the first works appeared on the creation and comprehensive study of new, so-called high-entropy polymetallic alloys (HEAs), including up to 5—6 basic elements, each in a high concentration (for example, from 5 to 35%). Alloys of the systems AlCoCrCuFeNi, CoCrCuFeNiTi, and others were chosen as the first candidates for such materials. These materials, along with the characteristics typical of metal alloys, have unique and unusual properties inherent, for example, in metal-ceramics: high hardness and resistance to temperature softening, precipitation hardening, positive temperature hardening coefficient, high strength characteristics at elevated temperatures, high wear and corrosion resistance, and a number of other characteristics. The main feature of HEAs is the formation of a single-phase stable, thermodynamically stable and high-strength substitutional solid solution predominantly with the FCC or BCC lattice [1]. Stabilization

of the solid solution and prevention of the formation of intermetallic phases during crystallization are ensured by the high entropy of mixing of the components ($S_{mix} > 1.61R$, where R is the universal gas constant) in the initial (charge) and liquid (melt) states. The maximum value of entropy is achieved at an equimolar element ratio. In recent years, several reviews [1—6] have been published (mainly in foreign literature), which reflect the authors view of the most interesting HEAs properties. These works describe the thermodynamics of HEAs, and consider the results of modeling their structure and new variants of methods for obtaining multicomponent alloys.

Already the first studies revealed significant differences in the structure of various high-entropy alloys (HEA). HEAs based on a solid solution [7, 8], mixtures of intermetallic phases [9, 10], an amorphous phase [11, 12], as well as alloys with a more complex multiphase structure [4—13], were discovered.

In the last two decades, there has been an exponential increase in the number of publications devoted to the creation of single-phase equiatomic five-component HEAs of the CrMnFeNi, AlCoCrFeNi types [14], as well as complexly concentrated alloys (CCAs). Despite a significant number of works, it is extremely difficult to conduct a rigorous direct comparative analysis of the structural-phase states and mechanical properties of HEAs and CCAs of the same composition. It was noted in [15] that: "A direct comparison of data is difficult due to differences in the type and concentration of the main elements, the type of thermomechanical treatment, the temperature and duration of heat treatment." Let us analyze, however, the latest fundamental works on five-component alloys.

RESULTS AND DISCUSSION

HEAs Mechanical Properties

The properties of HEAs are determined by their structure and elemental composition. Thus, HEAs with the BCC lattice have predominantly high strength and low ductility, while materials with the FCC lattice have low strength and high ductility. When analyzing the mechanical properties under uniaxial compression, the values of the main HEAs parameters differ little from the parameters of industrial alloyed steels. Activation of twinning mechanisms for the $Al_{0.3}$CoCrFeNi alloy provides deformation up to 97%, and the strength is 1378 MPa [16]. In the two-phase AlCoCrFeNi alloy, the ultimate strength can reach 4390 MPa [19]. These values, in the opinion of the authors of review [14], can be clearly overestimated due to the peculiarities of the compression unit, the shape, and dimensions of the samples.

It is also inappropriate to automatically transfer unique data on compression to data on tension. The reasons for such errors may be the failure to consider the presence of nanosized intermetallic compounds and precipitates, short-range order in single-phase BCC or two-phase FCC + BCC alloys.

In this paper, the most interesting and curious results of numerous studies of the mechanical properties of HEAs at different temperatures are considered. In [7], when studying the mechanical properties of CrMnFeCoNi and CrFeCoNi alloys subjected to tension in the temperature range from −196 to 1000°C, their high ductility at room temperature was revealed. Thus, plasticity and hardness increased with decreasing temperature, while the tensile strength almost doubled, and the relative elongation to failure increased by 1.5 times (up to 60%).

The excellent mechanical properties of the high-entropy AlCoCrFeNiTi alloy at room temperature are described in [17]. The improvement of mechanical characteristics, consider the large radius of the titanium atom, was explained by the authors by solid solution hardening. The $AlCoCrFeNiTi_{0.5}$ alloy showed the best mechanical characteristics: the yield strength is 2.26 GPa, the tensile strength is 3.14 GPa, and the plastic deformation is 23.3%.

A detailed consideration of the mechanical characteristics of the HEAs at low temperatures was carried out in [18, 19]. The AlCoCrFeNi alloy has excellent mechanical characteristics both at room and cryogenic temperatures [18]. An increase in the yield strength and durability with a decrease in temperature was found from 298 to 77 K by 29.7 and 19.9%, respectively. According to the authors' interpretation, the mechanical characteristics are improved in the presence of a single-phase BCC solid solution structure.

The values of the mechanical properties of single-phase FCC HEAs are close to those for nickel-based alloys and austenitic stainless steels with the same alloying elements. It is noted in [20] that significantly improved properties of FeMnCoCrNi HEA (920 MPa and 52%) are achieved by doping with boron. This leads to segregation of boron at grain boundaries, a decrease in grain size, and a weakening of interfacial strengthening. Doping with carbon (810 MPa, 66%) [21] in the interstices increases the stacking fault energy for this HEA. It is noted that the optimization of the parameters of the thermomechanical treatment of the CoCrFeNiMn HEA doped with 1% C (at %) significantly improves the mechanical properties. The key role here belongs to intermetallic compounds of the $Me_{23}C_6$ type. The temperature dependence of the specific strength for HEA CoCrFeMnNi [22] at a temperature above 600°C shows a drop, which can be eliminated by simultaneous alloying with aluminum and titanium and short-term annealing. Unfortunately, in the literature, there is a limited number of publications on the HEAs behavior at low temperatures. Thus, with these purposes, the CrMnFeCoNi alloy was created at the beginning. At low temperatures, it has enhanced strength, plastic properties and high impact strength [7—23].

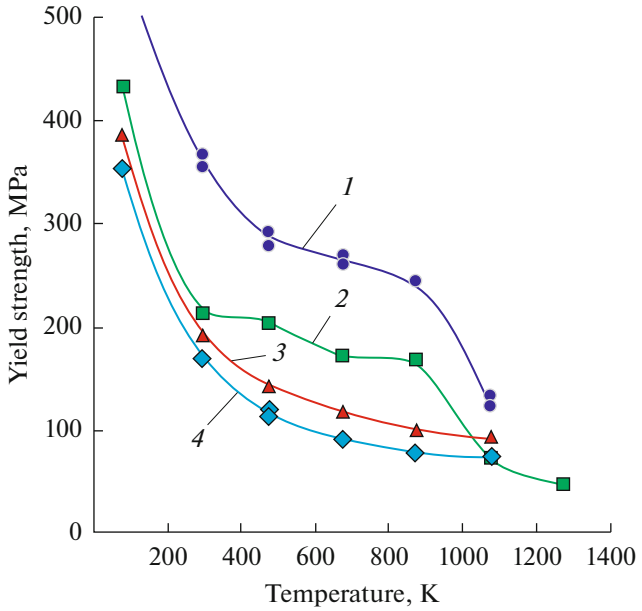

Fig. 1. Temperature dependence of the yield strength of Cr MnFeCoNi HEA for different grain size values [26, 27], μm: (*1*) 4; (*2*) 35; (*3*) 50; (*4*) 155.

Pure FCC metals have almost no temperature dependence of the yield strength in the range of 77–500 K [24]. For FCC coarse-grained Cantor HEAs in this temperature range, this parameter increases up to four times as the temperature decreases [25].

As a rule, HEAs have greater strength than conventional alloys with fewer elements [25]. It should be kept in mind that in concentrated solid solutions, the effective radius of constituent elements differs significantly from its value in pure elements [26]. Therefore, to determine their contributions to solid solution strengthening, it is necessary to calculate from first principles the energy of interaction with dislocations for an individual dissolved element. Regarding the question to what characteristics are discussed when compiling the HEAs composition, it may be said that this is a discrepancy between the volumes of alloying elements. Based on traditional solid solution theories, this is easier said than done. How can volume mismatch be measured? The point is that the HEA atoms are displaced to different degrees. For single crystals, X-ray diffraction studies make it possible to quantify the atomic displacement parameter (ADPs) [26] (ADP is the sum of the areas of deviation from the equilibrium position due to thermal vibrations (dynamic disorder) and statistical displacement due to the size effect).

In tensile tests of single crystals of the CrMnFeCoNi alloy, depending on the test temperature (77–423 K) and orientation, the values of the critical shear stress vary from 53 to 172 MPa [27–29]. These values are at

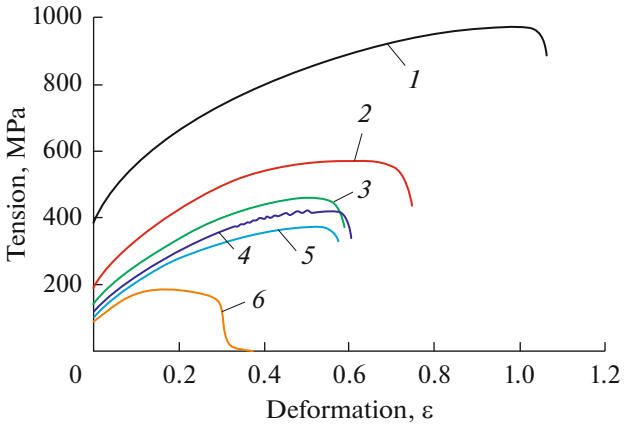

Fig. 2. Stress-strain curves for CrMnFeCoNi HEA at different temperatures [27], °C: (*1*) –196; (*2*) 20; (*3*) 200; (*4*) 400; (*5*) 600; (*6*) 800.

least an order of magnitude higher than for FCC pure nickel.

When analyzing the thermally activated process of plastic deformation, an important parameter is the activation volume (it is approximately $60b^3$ at 77 K and $360b^3$ at room temperature). This is somewhat less than in solid solutions (~$1000b^3$) (where b is the Burgers vector). During quasi-static tests (10^{-1}–10^{-5} s^{-1}) for an alloy with a grain size of 35 μm, the effect of the deformation rate is insignificant [7] (Fig. 1). In the range 10^{-3}–10^{2} s^{-1}, the yield strength increases by about 35% (from 484 to 650 MPa) for an alloy with a grain size of 10 μm, with a corresponding increase in the tensile strength from 853 to 968 MPa. At high rates, the deformations are even more significant: from 325–360 MPa at rates of 10^{-4}–10^{-2} s^{-1} to 590–680 MPa at rates of 3000–4700 s^{-1} [30]. This is due to the phonon drag [30].

Typical curves σ(ε) for the CrMnFeCoNi HEA with a grain size of 50 μm for different temperatures are shown in Fig. 2. During compression of the CrMnFeCoNi alloy with a grain size of –150 μm, the flow stresses increase with decreasing size, except for high temperatures (800°C), where the alloy with a grain size of 5 μm exhibits deformation softening after yielding mainly by the grain boundary deformation mechanism. The yield stress, flow stress, and elongation increase with decreasing temperature down to liquid nitrogen temperature [7].

Dislocation and Twin Plasticity

As for FCC metals, the dislocation substructure of the HEAs exhibits a transition from chaos at low degrees of deformation to coils and cells with an increase in the degree of deformation (Fig. 3). In this case, there is no difference in the increase in the dislocation density at room temperature and 77 K, which

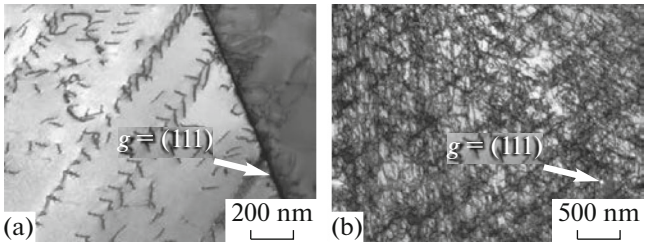

Fig. 3. Planar dislocation clusters on the {111} plane of CrCoNi HEA at different deformation degrees: (a) $\varepsilon = 5\%$ at 77 K; (b) $\varepsilon = 22\%$ at room temperature (cellular-reticular dislocation substructure) [40].

corresponds to the temperature independence of the hardening rate of FCC metals at small deformations. At $\varepsilon > 6\%$, the twinning mechanism starts working, which becomes dominant at $\varepsilon > 9\%$. The twin mechanism of plasticity can simultaneously increase strength and plasticity [31]. The axial stresses for the appearance of twins at 77 K are approximately 720 MPa [32]. The general opinion of the authors is that these stresses are basically independent of temperature.

Another factor related to twinning is the stacking fault energy: for the ternary equiatomic alloy CrCoNi, it is 25% lower than for CrMnFeCoNi (22 and 30 mJ/m^2, respectively) [33]. The higher level of yield strength for the CrCoNi alloy allows the critical twinning stress to be reached earlier compared to the CrMnCoNi alloy.

The high-entropy alloy $Fe_{36}Mn_{21}Cr_{18}Ni_{15}Al_{10}$ in the cast state has the BCC structure with embedded B2 particles of a cuboidal shape, consisting of mainly aluminum and nickel. The compressive and tensile yield strengths at room temperature are 750 and 990 MPa, respectively. After 600°C, there is a noticeable decrease to 310 and 360 MPa. When compressed, the HEA demonstrates a noticeable plasticity, starting from room temperature. In tension, the elongation to failure is only 2.5%. An increase in temperature leads to a moderate increase in ductility upon fracture. Annealing at 1000°C leads to the formation of the FCC phase. The microhardness noticeably decreases from 410 HV in the cast state to 310 HV after annealing at 1000°C [34]. In the opinion of the authors of this work, the transformation of the BCC structure into the FCC is associated with the presence of aluminum. This 10% Al changes the Valence Electron Concentration (VEC) compared to alloys of the same composition based on Co–Cr–Fe–Ni–Mn.

Single Crystal Alloys

Numerous works on HEAs testify to the decisive role of the orientation of single crystals during testing. In this case, it is important which exact methods for determining twinning were used. TEM methods allow, at $\varepsilon > 5\%$ for samples oriented for multiple glide, to fix the onset of twinning. When oriented for single glide, initially up to 27%, the dislocation activity plays the major role. With the multiple glide orientation [001], no twinning was observed at all. It is also known that twinning becomes more difficult with decreasing grain size [35]. In single crystals, it occurs at lower tensions. It should be noted that backscattered electron diffraction and other methods are inferior to TEM in assessing the role of twinning.

HEAs with BCC and FCC Lattices

Decreasing the VEC in molybdenum and niobium alloys reduces the shear strength [36], so that low VEC alloys are easier to deform and become more ductile. Compared to FCC alloys, BCC HEA alloys are less studied due to brittleness at low homologous temperatures. The exceptions are NbMoTaW, VNbMoTaW, TiVZrNbHf, and especially TiZrNbHfTi alloys [37]. After arc melting and pressing (1473 K, 207 MPa, 3 h), a dendritic structure is formed in it, which disappears after thermomechanical treatment (cold rolling + recrystallization for 2 h at 1000°C) with the formation of a homogeneous grain structure and a homogeneous chemical composition.

Compared to the FCC HEAs (for example, CrMnFeCoNi), the BCC TiZrNbHfTa alloy is less hardened. The estimate of the parameter α in the expression $\Delta\tau = \alpha Gb\sqrt{\rho}$ indicates the decisive role of the Peierls barrier. For the CrMnFeCoNi alloy with the FCC structure, the forest dislocations make the main contribution. For grains with sizes of 38.81 and 128 μm, the tensile yield strength of HEA TiZrNbHfTa is approximately the same, 950 MPa [38], which is less than for grains with sizes of 22 μm (1145 MPa) [37].

Theory and Models for Describing the Mechanical Properties of HEAs

Molecular dynamics methods used to describe mechanical properties based on model concepts are given in [37–40]. Difficulties in interpreting HEAs plasticity are related to the need to understand how dislocations move through compositionally misoriented lattices. A quantitative theoretical analysis of mechanical properties (such as cold hardening, ductility, twinning, fracture, fatigue) is difficult for HEAs, since there is still no unambiguous approach even for diluted alloys. Difficulties are due to the multicomponent nature of the HEAs composition. First of all, the difficulties in determining the role of the short-range order in the mechanical properties of the HEA should be overcome. The mechanisms of Taylor hardening (dislocation interaction), interfacial interaction (interaction of dislocations with grain boundaries, twins, particles) are studied much better.

HEAs Application Areas

HEA can be used, in particular, in the nuclear power industry, since they have a high resistance to radiation [41], as refractory, magnetically soft materials, as well as materials used in the metalworking industry. In the aerospace industry, HEAs can find application due to their corrosion resistance, as well as materials for hydrogen storage [42, 43]. Paper [44] presents the technology developed by the authors and describes the properties of a new class of superhard coatings based on HEA. The starting material for deposition is a new class of materials—HEA. The use of advanced technological modes of vacuum-arc deposition made it possible to obtain superhard coatings based on solid-soluble HEA with thermal stability up to 1100°C [44]. It was revealed that high-entropy single-phase nitride coatings based on five nitride-forming elements are characterized by high values of hardness (50–60 GPa) and elasticity modulus (more than 600 GPa). The formation of the lattice type of nitride coatings is most affected by the heat of formation and the predominance of nitrides with one type of crystal lattice. It was established that high-entropy single-phase nitride coatings are quite thermally stable [44].

One of the promising fields of application of nitride coatings based on HEA is biomedicine. Protective coatings for biomedical applications should have a low modulus of elasticity, high chemical stability, wear resistance and corrosion resistance in physiological environments, low coefficient of friction, biocompatibility, and excellent adhesion to the surface on which protective coatings are deposited [45]. Particularly noteworthy is work [6], which predicted possible areas of application of HEAs in various industries.

A significant increase in the characteristics of the surface layers of metals, steels, light alloys can be achieved through the use of external energy effects. The most effective of them are high-power electric currents, electroexplosive alloying, processing by high-current low-energy electron beams [46–48]. Such external energy impacts, due to the modification of surface layers, significantly increase the cyclic durability and wear resistance of titanium and titanium alloys [48], stainless steels [49], light alloys [46], and the electroerosive resistance of copper-based high-power electric circuit switches [50]. The data accumulated and analyzed in the presented works make it possible to expand the fields of application of HEAs in various industries.

CONCLUSIONS

The last two decades are characterized by the appearance of a significant number of works on the creation, study and use of HEA containing up to 5–6 basic elements with a concentration of 5 to 35%. Compared to typical metal alloys, they exhibit unique properties due to their high hardness, thermal and corrosion resistance, thermal stability at elevated temperatures, and others. An attempt is made to perform a brief analysis of the latest publications of foreign authors on the formation of structural-phase states and properties of mainly five-component alloys of the AlCoCrFeNi type and areas of their possible application. It is noted that despite the significant number of works, the rigorous comparative analysis is difficult due to a number of objective reasons. There is an accumulation of experimental data and its comprehension. High-entropy BCC alloys have predominantly high strength and low ductility, while FCC materials have low strength and high ductility. It is shown that significantly improved properties of the FCC FeMnCoCrNi HEA are achieved by doping with boron. The same can be achieved by optimizing the parameters of thermomechanical treatment with the introduction of 1% C (at %). An analysis of the deformation curves indicates an increase in the yield strength and relative elongation as the temperature is lowered to the temperature of liquid nitrogen.

For FCC HEAs, at a degree of deformation $\varepsilon > 6\%$, along with the dislocation mechanism, the twinning mechanism starts working. When testing single-crystal HEAs, orientation plays a decisive role. It is best to fix the onset of twinning in multiple and single glide using TEM methods.

Depending on the set of HEAs properties, the areas of their application are indicated. Superhard HEA-based coatings are thermally stable. Possible promising areas for the use of external energy effects for modifying the surface layers of HEAs are shown.

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