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OVERVIEW OF THE HIGH-ENTROPY ALLOYS CONCEPT

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The article presents a review of the results of research on high-entropy alloys, describing the principles of their formation, the basic concepts and properties of high-entropy alloys. The existing categories of the entropic alloys are listed. The effects resulting from the formation of high-entropy alloys are described: high entropy, lattice distortion, sluggish diffusion and cocktail effects. It is noted that the traditional thermodynamic representations for multicomponent alloys require additions. It is also noted that the application of Hume-Rothery rules to predict the phase composition of high-entropy alloys shows some difficulties, since it is difficult to select a large number of elements having the same type of lattice and valence. The results of the analysis of a number of parameters and conditions which, according to the researchers' opinion, affect the structural state created by high-entropy alloys, taking into account which could allow to correctly predict the formation of structures in high-entropy. Analysis of the literature data has shown that at present there is no universal parameter that could allow the correct prediction of the formation of structures in multicomponent alloys systems. Methods for the preparation of powders of high entropy alloys are presented. The results of frequently used methods of obtaining coatings on the basis of high-entropy alloys, such as laser cladding, magnetron sputtering, electro-chemical deposition and thermal spraying are reviewed. Disadvantages of obtaining coatings methods are described.

Keywords: *high-entropy alloys, multicomponent alloys, thermodynamics, structure, coatings methods.*

1 INTRODUCTION

Over the past 20 years, the development of theories and technologies in the field of new materials has moved toward the use of many elemental alloys. This class of metallic compounds is called high-entropy alloys (HEA). HEA have properties such as high corrosion resistance [1, 2], high wear resistance [3] and specific mechanical properties [4, 5], which are optimal in heavy (aggressive) operating conditions [6]. CrMnFeCoNi-based HEA (Cantor alloys) [7], CoCrNi-based medium-entropic alloys [8, 9], CoCrFeNb eutectic HEA [10], CoCrFeNiTiAl alloys [11], HfMoNbTiTiZr [12] and TiZrNbWMo [13] are mainly being studied. These new alloys are also known as multicomponent alloys or alloys with several major elements, which differ from traditional alloys containing one major element, such as alloys of Al, Cu, Fe, Ti, etc. HEA are mainly composed of five or more elements in equiatomic or nonequiatomic ratios, where the composition of each element varies from 5 to 35 at.% [14]. Mixing of elements in this composition yields simple solid solution phases such as face-centered cubic (FCC), body-centered cubic (BCC), and hexagonal close packed (HCP) structures [7]. HEA were believed to possess exceptional characteristics, such as high mixing entropy, lattice distortion, delayed diffusion and cocktail effect, which account for their high mechanical strength, resistance to high temperature, wear resistance and corrosion resistance. Current researches on high entropy alloys is focused on microstructure evolution, hardness prediction, the effect of HEA nanoparticles on the efficiency of high-performance lithium-ion batteries, the mechanical properties of eutectic HEA, and the use of machine learning to predict HEA phase formation by composition. as well, work is underway to investigate transition metals (TM HEA) are considered to be promising structural materials for high-temperature (HT) applications. Nevertheless, their high-temperature oxidation is a critical problem and needs to be addressed for hot components. Therefore, HEA have exciting potential in various engineering structures [15–18]. However, the excessive cost of the elements of these systems limits their use in large-sized equipment and components. The application of surface coating technology effectively reduces the cost of production and maintenance of large equipment.

HEA-based coatings have become a potential surface protector because of their high surface protection capabilities [19]. In recent years, there have been tremendous changes in the methods used to produce HEA-based coatings and applications. This review focuses on the basic understanding of HEA and related HEA concepts, and summarizes the main problems associated with the development of HEA coatings, offering system-level recommendations for future research in this area.

Previously published reviews considered separately the basic properties, characteristics of HEA and the ability to form HEA phases (BCC, FCC, HCP, amorphous and multiphase) by entering five calculated parameters, (VEC, $\Delta\chi$, δ, $\Delta H_{mixture}$ and $\Delta S_{mixture}$). This review article systematizes the data of previous reviews, discusses the current methods for producing HEA powders and the characteristics of the possessing coatings based on them. This review article is the basis for the dissertation work, which will investigate the phase formation of HEA obtained by mechanical alloying at certain temperature regimes.

2.1 Formation of HEA

In order to achieve the necessary properties, researchers are developing new composite alloys. One of the ways to achieve such objectives is the concept of increasing thermodynamic entropy by increasing the number of components taken in certain proportions. Therefore, these alloys are called high entropy alloys.

Existing metallurgical knowledge and binary/triple phase diagrams suggest that such multi-element alloys can form several tens of phase types and intermetallic compounds, resulting in complex and brittle microstructures that are difficult to analyze and design, and likely have extremely limited practical value. The main feature of HEA is the formation of a single-phase stable thermodynamically stable solid solution. Stabilization of the solid solution is provided by high entropy of mixing of components in initial and liquid states. The entropy of mixing, or also called thermodynamic entropy, for pure elements depends only on the vibrational (thermal) motion of atoms. And in the cases of alloys, in addition to the vibrational motion of the atoms, the entropy of mixing is composed of the configuration, magnetic, and electronic components [20]. An increase in the number of components leads to an increase in configurational entropy. And based on the Boltzmann hypothesis, an increase in the configurational entropy of the alloy, both in the liquid and in the solidified state, contributes to the formation of a simple structure of a single-phase disordered solid substitution solution. In [21] it is reported that configurational entropy really favors the solid solution phases in comparison with the classical thermodynamic concepts, based on which the state of the system is determined through density, pressure, temperature and other local thermodynamic parameters.

In general, the HEA concept has expanded into three categories (Table), such as alloys with high entropy (from five elements up to thirteen), alloys with medium entropy (three or four basic elements) and alloys with low entropy (less than three basic elements) [22, 23].

Table. Classification of HEA according to their constituent elements [24]

Alloys	Number of elements	Examples
High entropy alloys	$5 \leq n \leq 13$	AlCoCrFeMnNi, CoCrFeMnNi, AlCrCuFeNi, MoNbTaW
Medium entropy alloys	$3 \leq n < 5$	CoCrNi, CoCrFeNi
Low entropy alloys	$1 \leq n < 3$	AlMg, AlCu, NiCo

Due to the considerable number of elements that set the composition of the matrix, to describe the HEA consider the factors of influence on the microstructure and properties of the alloys. HEA has four effects, [14, 25, 26] which are less pronounced in conventional alloys, in the literature they are called “core effects”. The core effects that are observed during the formation of HEA are: high entropy, lattice distortion, sluggish diffusion and mixing effect (the “cocktail” effect) [14]. Three of them are hypotheses, and the mixing effect is a separate characteristic of the HEA.

The high entropy effect. High entropy effect. The high entropy effect is a distinctive feature of HEA, and it is assumed that the increased configuration entropy in nearly equimolar alloys with five or more elements may favor solid solution phases and prevent the formation of intermetallic compounds. According to Gibbs' rule of phases in a five-element system, the number of phases in the alloy at constant pressure and temperature should be six equilibrium phases. However, HEA form a single-phase solid solution, which contradicts Gibbs' rule of phases. A solid solution, in terms of thermodynamics, forms at the minimum Gibbs free energy. Based on the Gibbs free energy equation (2), high entropy reduces the Gibbs free energy, thereby contributing to the stabilization of the solid solution and preventing the formation of intermetallic phases during crystallization [27, 28].

There is much evidence for the high entropy effect [27, 29–34]. Figure 1 shows a diffractogram of a multicomponent alloys obtained by sequentially adding an additional element, with an increase in the number from two to seven components. Multicomponent systems form solid solutions with FCC and BCC structure, which is contrary to common expectations: the formation of various kinds of binary/triple compounds [35].

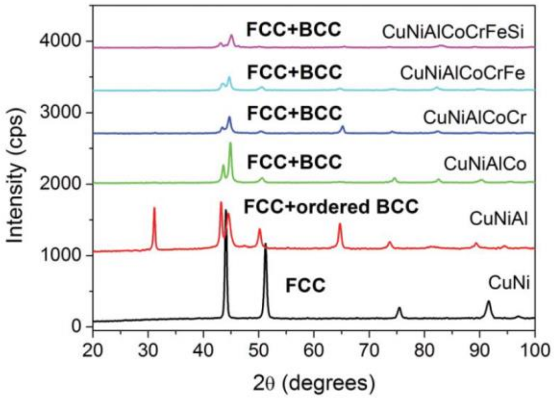

Figure 1. Diffractogram of multicomponent alloys obtained by sequential addition of an additional element [35]

The lattice distortion effect. The HEA crystal lattice consists of different elements, hence having different atomic radii. During the formation of an HEA, each atom has an equal probability of occupying one or another node of the crystal lattice, which leads to a serious distortion of the lattice. Since the size of the atoms in the HEA lattice differs from each other, the formation of a solid solution is accompanied by a distortion of the crystal lattice. The displacement of an atom in each node of the lattice leads to an indeterminate position of the atoms, hence the configuration entropy increases.

A schematic illustration of the effect of lattice distortion on the intensity of X-ray diffraction peaks is shown in Figure 2. The heterogeneity of the constituent elements significantly reduces the degree of crystallinity and X-ray scattering, which leads to an anomalous decrease in the height of the peaks [36]. Lattice distortion prevents the movement of dislocations and leads to a pronounced solid-solution hardening, as well as a decrease in electrical and thermal conductivity due to increased scattering of electrons and phonons. It is known that temperature changes affect the oscillation of atoms; when the temperature increases, the amplitude of oscillations increases, which leads to a decrease in the intensity of the reflected X-rays [37- 39]. A similar phenomenon is observed in the deformation of the crystal lattice, leading to the distortion of atomic planes. However, it is worth noting that when the number of components increases, the diffraction peak height decreases markedly more intensely than for the thermal effect (Figure 1,2).

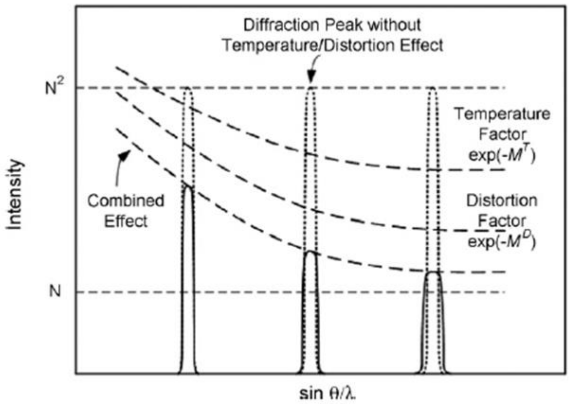

Figure 2. Schematic illustration of the effect of lattice distortion and temperature on the intensity of the XRD peaks [36]

The sluggish diffusion effect. In HEA, the potential energy between the nodes of the crystal lattice changes because of the heterogeneity of the atoms. In ordinary alloys with a low concentration of dissolved matter, the local configuration of the atoms before and after the vacancy jump is the same in most cases, while in HEA the difference in local atomic configurations leads to different bonds and, consequently, different local energies. In [40] the calculation of the activation energy of diffusion of elements in different matrices of CoCrFeMnNi HEA is presented. It is known that the fluctuations of the potential energy of interatomic bonding affect the kinetics and activation energy of diffusion. In pure metals, the fluctuation of potential energy after the migration of atoms is zero. With substantial changes in potential energy, at which the potential barrier and activation energy of diffusion increase, trapping of atoms by "traps" is likely and, consequently, the diffusion coefficient is lower. Properties of HEA, such as slow grain growth [41], strength at hot temperature [28, 42], and formation of nanostructures and nanoscale separations [30, 43-45], are associated with low values of the diffusion coefficient of atoms in HEA.

The cocktail effect. Compared to the other "core effects", the "cocktail" (mixing) effect is not a hypothesis and does not require proof. The essence of the "cocktail effect" is that exceptional properties of materials are often the result of unexpected interactions that can occur due to unusual combinations of elements and microstructures in multi-element systems.

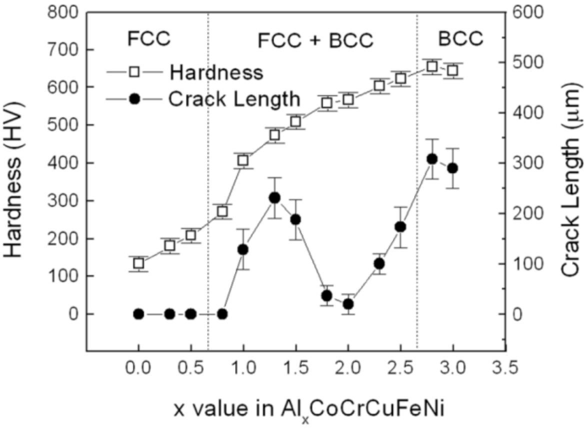

Figure 3. Hardness of $Al_xCoCrCuFeNi$ alloys as a function of Al concentration [29]

It should be noted that the properties of multicomponent alloys are due not only to the constituent elements, but also to the formation of phases. Depending on the composition, method of production and processing of the alloy, ordered and disordered phases of the solid solution are formed in the HEA. Both the atoms of the individual elements and the phases formed contribute to the properties of the HEA, which is the "cocktail" effect. In addition to the properties of the individual elements, the interaction between the constituent elements should also be considered. For example, Al is a soft element with a low melting point, but the addition of Al can improve the strength of the HEA. Fig. 3 shows the hardness of $Al_xCoCrCuFeNi$ alloys as a function of Al concentration. The increase in the strength of $Al_xCoCrCuFeNi$ alloys is due to the formation of the solid phase of the BCC and stronger cohesive bonding between Al and other elements.

The macroscopic properties of HEA are determined not only by the averaged properties of its constituent elements, but also include the effects of excess quantities resulting from inter-element reactions and distortion of the crystal lattice.

2.2 Thermodynamics of HEA

According to the second law of thermodynamics, the course of phase transformation processes is determined by the action of enthalpies (energy) and entropies (structural). Enthalpy determines the tendency of the system to go to the state with the lowest energy. Entropy determines the tendency of the system to go to the state with the maximum degree of disorder. As noted earlier, high entropy is the main factor influencing the formation of single-phase solid solutions, as was shown in [46].

For pure elements, the entropy depends only on the vibrational (thermal) motion of the atoms. And in the cases of alloys, in addition to vibrational S_v, the entropy of mixing is composed of the configurational S_{conf}, magnetic S_m, and electronic S_e components [20]. The mixing entropy also has the property of additivity. Consequently, S_{mix} can be mathematically expressed as:

$$\Delta S_{mix} = \Delta S_{conf} + \Delta S_v + \Delta S_m + \Delta S_e. \quad (1)$$

Also, in [46, 47] it is noted that for the case of multicomponent HEA, consisting of five or more elements, the configuration entropy dominates over the other three components. Since it depends on the many ways of distribution of atoms in the lattice and the resulting vacancies. An increase in the number of components leads to an increase in the configurational entropy, which leads to a decrease in the Gibbs free energy (2).

$$\Delta G = \Delta H - T\Delta S. \quad (2)$$

where G – Gibbs energy; H – enthalpy; S – entropy; T – absolute temperature.

In [31] it was shown that the multicomponent AlCrFeNiCu system with high mixing entropy has lower Gibbs energy (ΔG) values than intermetallics. It was believed that with increasing mixing entropy the possibility of formation of stable disordered solid solutions is higher than for ordered or intermetallic compounds.

It is possible to prove this statement based on Boltzmann hypothesis [48], therefore, in the case of equal molar fractions of each element the entropy of mixing is expressed as (3):

$$\Delta S_{mix} = -R\,(1/n\ \ln 1/n + 1/n\ \ln 1/n + \ldots + 1/n\ \ln 1/n) = \\ = -R \ln 1/n = R \ln n \quad (3)$$

Thus, the mixing entropy of solid solution formation increases as the number of elements increases (Figure 4).

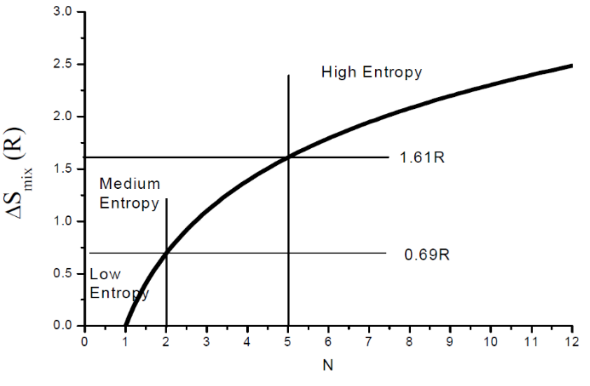

Figure 4. Entropy of mixing of alloys with the equiatomic ratio depending on the number of components [49]

There are different HEA structures: based on solid solution [7, 50, 51]; mixtures of intermetallic phases [52, 53]; amorphous phase [54, 55]; alloys with a more complex multiphase structure [30, 56–58]. This indicates that, in addition to high entropy, there are other factors affecting the formation of HEA. In [46, 59, 60], the authors identified three main parameters responsible for the formation of amorphous phases and solid solutions in multicomponent alloys. These parameters included the atomic size difference (δ), enthalpy (ΔH_{mix}), and entropy (ΔS_{mix}) of mixing. The obtained calculated formulas of the parameters are presented below:

– entropy of mixing [61]:

$$\Delta S_{mix} = -R\sum c_i ln c_i \quad (4)$$

R – the universal gas constant, c_i – the concentration (at. %) of the i-th element in the alloy;

– enthalpy of mixing [62]:

$$\Delta H_{mix} = \sum 4\Omega_{ij} c_i c_j \quad (5)$$

c_i and c_j – the concentration (at. %) of the i-th and j-th element in the alloy, respectively, $\Omega_{ij} = 4\Delta H_{AB}^{mix}$ – concentration-dependent parameter characterizing the interaction between elements in solid solution;

– average difference in atomic radii:

$$\delta r = 100\%\sqrt{\sum c_i \left(1 - r_i / \bar{r}\right)^2} \quad (6)$$

c_i – the concentration (at. %) of the i-th element in the alloy, r_i – atomic radius of the i-th element in the alloy, $\bar{r} = \sum c_i r_i$ – average atomic radius of the alloy.

The authors state [60–63] those solid solutions formed when the difference of atomic sizes is not great, the enthalpy of mixing has values close to zero, and the entropy of mixing has high values. In contrast, the amorphous phase tends to form when the difference in atomic size is significant, the enthalpy of mixing is negative, and the entropy of mixing is low. However, further studies have shown that the use of these parameters to predict the phase composition of the HEA does not always yield clear and accurate results. Based on these three parameters, it is impossible to say for sure whether a solid solution or an amorphous phase will form.

In order to predict the structure of HEA, the above factors have been considered in a number of works [60, 61, 64, 65]. For example, in [60] it was shown that the solid solutions in HEA (alloys $CrFeCoNiAlCu_{0.25}$, VCuFeCoNi, $Al_{0.5}CrFeCoNi$, $Ti_2CrCuFeCoNi$, ZrTiVCu NiBe and AlTiVYZr) are formed at values -15 kJ/mol $\leq \Delta H_{mix} \leq 5$ kJ/mol and $1 \leq \delta r \leq 6\%$. The required mismatch parameter between the atomic radii of the elements for solid solution formation agrees well with the Hume-Rothery rule.

Then in [63] it was proposed to introduce an additional thermodynamic parameter Ω, to predict the formation of the HEA structure

$$\Omega = T_m \Delta S_{mix} \left|\Delta H_{mix}\right| \quad (7)$$

$T_m = \sum c_i T_{mi}$ and T_{mi} – is the melting temperature of the elements. Calculations of Ω together with δr showed that the formation of simple solid solutions in HEA is observed, to a greater extent, when the condition $\Omega \geq 1.1$ and $\delta r \leq 6.6\%$ is fulfilled, and the formation of multiphase structure consisting of solid solutions and intermetallic phases is fulfilled when the condition $1.1 < \Omega < 10$ and $\delta r > 3.8\%$. Despite the error, the parameter Ω can function as a sufficiently reliable "tool" to distinguish between the formation of solid solutions or intermetallic phases in multicomponent systems. The high value of the parameter $\Omega > 1.1$ and the small value of $\delta r < 6.6\%$ predict the formation of solid solutions in HEA.

The application of Hume-Rothery rules to predict the phase composition of HEA shows some difficulties, since it is difficult to select a large number of elements having the same lattice type and valence values. For example, based only on Hume-Rothery rules in the Co(FCC)-Cr(BCC)-Fe(BCC)-Ni(FCC)-Mn(FCC) alloy, consisting of elements with completely different crystal lattices, the structure of a simple solid solution is formed, a multiphase structure should be formed [66–70]. A similar system Al(FCC)-Co(FCC)-Cr(BCC)-Cu(FCC)-Fe(BCC)-Ni(FCC) formed a multiphase structure [57].

In [71], it was proposed to complement the Hume-Rothery rules for multicomponent alloys. The following factors determining the formation of phases were considered:

- size factor associated with the difference in atomic radii (δr) of the constituent components;
- electrochemical factor associated with the difference in electronegativity ($\Delta\chi$) of the components;
- electron concentration per atom, e/a;
- the ability to complete the electron shells of element atoms near the end of short periods;
- orbital limitations.

Another approach was proposed by the authors Miracle and Senkov [50, 72] in the study of a single-phase alloy of the TaNbHfZrTi system with the BCC lattice, which they had previously applied to predict structure formation in amorphous alloys [54]. The peculiarity of the approach is that dissolved substances can destabilize the matrix lattice due to internal stresses and change the coordination number. The approach is based on the calculation of local changes in the interatomic distance and the elastic modulus arising near atoms of a certain sort in the assumed equiatomic solid solution. In a BCC lattice, they can be estimated as follows. Each element in this lattice has 8 nearest atoms, thus forming a cluster of 9 atoms. It is possible to roughly estimate the local environment of the atom of element *i* if we assume that the local chemical composition corresponds to the composition of the alloy. Thus, element *i* has $Nj = 9cj$ neighboring atoms of element *j* and $Ni = 9ci$ neighboring atoms of element *j*. Then the change of interatomic distances δr_i, and elastic modulus δG_i, around element i is estimated as an average difference of atomic radius and elastic modulus of this element with its neighbors:

$$\delta r_i = \frac{9}{8} \sum c_i \delta r_{ij} \qquad (8)$$

$$\delta G_i = \frac{9}{8} \sum c_i \delta G_{ij} \qquad (9)$$

$\delta r_{ij} = 2(r_i - r_j)/(r_i + r_j)$ is the difference in atomic radii, $\delta G_{ij} = 2(G_i - G_j)/(G_i + G_j)$ – the difference in the elastic modulus of the elements *i* and *j*.

From the values of the modulus of elasticity, we can determine the element that causes the greatest local lattice distortion. It is assumed that local lattice distortions caused by the addition of elements can lead to lattice instability of the solid solution, hence the formation of other phases. This approach allows us to answer the question of whether the structure of disordered solid solution will form or not, but the question of which phases will form in the structure remains open.

Another parameter, the electronegativity difference, in multicomponent alloys [61] defined as:

$$\Delta\chi = \sqrt{\sum_{i=1}^{N} c_i \left(\chi_i - \overline{\chi}\right)^2} \qquad (10)$$

χ_i – Pauling electronegativity for the *i*-th component, $\overline{\chi} = \sum_{i=1}^{N} c_i \chi_i$ – average electronegativity.

In [73] an attempt was made to predict the formation of phases by applying the parameters δr and $\Delta\chi$. Instead of the empirical Pauling scale, the determination of electronegativity by the method described in [64] was used:

$$\Delta\chi_{Allen} = \sum_{i=1}^{N} c_i \left(1 - \chi_i / \overline{\chi}\right). \qquad (11)$$

The electronegativity in this case is represented as the average ionization energy of the valence electrons of free atoms in the ground state, and its values often coincide with the Pauling electronegativity, however, they differ greatly for transition metals. It has been shown that at $3 < \Delta\chi_{Allen} < 6$ and $1 < \delta r < 6\%$ the formation of only solid solutions and not intermetallic phases (including the σ-phase) is observed. In particular, there is a tendency that solid solutions with a BCC lattice can exist with a larger parameter of atomic radius mismatch of elements and a smaller difference in electronegativity than solid solutions with a FCC lattice. At higher values of $\Delta\chi_{Allen}$ such an area is observed in which intermetallics prevail. At $\delta r \approx 6\%$ and a high electronegativity difference, a boundary state between the formation of solid solutions and intermetallic phases is observed. These data show that such parameters as δr and $\Delta\chi_{Allen}$ cannot be used for unambiguous prediction of the phase composition of HEA.

The Hume-Rothery rules define the electronic concentration per atom (e/a ratio), which has an obvious influence on the crystal structure of the so-called Hume-Rothery electronic compounds. In [65] it is described that there are two basic definitions of electronic concentration: the average number of free electrons per atom e/a, and the total number of electrons, including d-level electrons located in the valence band, the number of valence electrons (*VEC*). The e/a or *VEC* for a multi-component alloys can be determined as:

$$VEC = \sum_{i=1}^{n} c_i \left(VEC\right)_i \qquad (12)$$

However, HEA consist mainly of transition metals (alloys of the CoCrFeNiAlCu system), for which the e/a ratio is ambiguous, and in most cases impossible. For this reason, the authors [65] proposed to study the influence of *VEC* on the phase composition of HEA. It was shown that the *VEC* can be used to quantitatively predict the phase stability of the FCC and BCC phases in the HEA. Thus, at $VEC \geq 8.0$ – a single-phase FCC structure is formed; at $6.87 \leq VEC < 8.0$ – BCC and FCC phases coexist together; at $VEC < 6.87$ – a single-phase BCC structure is observed. It should be noted that at $VEC = 8$ the BCC phases can be formed, however, they are secondary.

Based on the analysis of the literature data, we can say that at present there is no universal parameter that could allow to correctly predict the formation of structures in multicomponent alloys systems. Thus, the definition of the conditions governing the stability of phases in HEA, through statistical analysis of the general behavior of the constituent elements in multicomponent alloys, is an actual task.

3.1 Methods for the preparation of HEA powders

Pre-alloyed HEA is the production of a fine metal powder, where the alloy consists of five or more basic metal powders. From various research articles it appears that experimenters and researchers use pre-alloyed HEA powders as well as HEA powders obtained by traditional metal powder mixing followed by cold isostatic pressing or spark plasma sintering to give the desired shape [74–81]. There are also alternative ways of producing HEA powders. In fully pre-alloyed powders, two methods are mainly used, i.e., atomization (gas atomizing) or mechanical alloying (MA) (by using a ball milling machine), as well as other methods of producing HEA by melting and then grinding into powder using high-energy milling [24, 82]. In recent studies, the most pre-doped HEA powders are usually obtained by mechanical crushing and pulverizing and then consolidated by various consolidation methods. Scheme for the preparation of HEA powders and known consolidation methods shown in the Figure 5.

The HEA powders obtained by different methods have the same microstructure, a confirmation are the works where HEA WMoNbZrV obtained by arc melting [27, 28] and MA [83], obtained typical dendritic microstructure. MA is more optimal for high melting point alloys that are difficult to process by casting methods. MA is widely recognized as an important nonequilibrium solid-phase process route for the synthesis of various alloys with different phase composition and microstructure [84].

At present, there is a high level of research on HEA-based alloys and coatings due to their properties and unique application possibilities, especially the development of HEA-based coatings. Based on recent developments in this direction, HEA coatings can be divided into three groups [85, 86]: (1) metallic HEA, (2) ceramic HEA (or high-entropy ceramics), and (3) high-entropy composites.

Metallic HEA coatings include Cantor-based alloys [7] and their derivatives with other lightweight HEA based on transition elements Al, Cr, Co, Mn, Fe, Ti, Ni, V, refractory Ta, Mo, Hf, Nb, Zr, W, Ti, V, Cr, etc. [28]. HEA metal coatings are aimed at high-temperature applications where oxidation, wear and corrosion are of paramount importance. At the same time, lightweight metal alloy coatings are being researched for density reduction for various lightweight structural applications such as AlCuSiZnFe, AlLiMgScTi, AlCuFeMnMgTi, AlLiMgTiX, etc. [87-90].

High-entropy ceramics mainly consist of metal nitrides and carbides of transition elements [85]. In HEA solid ceramic coatings, constituent impurities such as O, C, N, H are manifested in solid solution and exhibit a high entropy effect. For hard ceramic HEA coatings such as Cr, Si, Zr, various ceramic forming elements Ti, Al, etc. are used. These HEA coatings have exceptional surface protection ability in terms of high strength, thermal stability, anti-corrosion characteristics and low diffusion rate [91, 92]. Because of these characteristics, these coatings are used for thermodiffusion, oxidation-resistant and tough radiation-resistant coatings in nuclear power plants [93].

High-entropy composite coatings are being developed based on metallic and ceramic coatings [94–96]. Similar to conventional composites, HEA composites can be synthesized by reinforcing the HEA matrix with suitable ceramic reinforcing elements. There are developments of ceramic reinforcing materials such as SiC, Al_2O_3, WC, TiC, TiN, TiB_2, NbC [94–96] that show better hardness, wear resistance, chemical stability and adhesion to the matrix coatings of HEA.

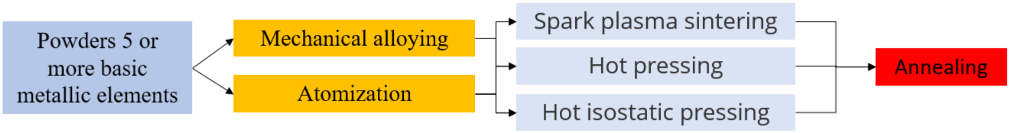

Figure 5. Scheme for the preparation of HEA powders and known consolidation methods

Coatings have been obtained through the development and evolution of various methods of producing HEA coatings. Significant progress has been made in these HEA composite coatings made by laser cladding, plasma arc cladding, magnetron sputtering, etc.

3.2.1 Laser cladding method

Laser cladding (laser doping of the surface) is a rapidly developing surface treatment method with the exceptional advantage of a high solidification rate (10^3–10^6 K·s^{-1}). This method is a frequently used method for HEA coating. The authors note that a feature of the method is high heat input, fast process, less material loss and environmentally friendly process [97–99]. Laser cladding allows the capture of nonequilibrium dissolved substances and avoids the stratification of components. This method can be used to produce HEA coatings with a thickness of about 1–5 mm, which is much larger than that of films produced by magnetron sputtering. Laser cladding provides a metallurgical bond strength between the coating and the substrate, which is higher than that of the thermal spraying method. In [100], Al_2CrFeNiCoCuTix coatings were obtained by laser cladding. The typical morphology of the microstructure of Al_2CrFeNiCoCuTix coatings is presented in [100], which shows that the cladding layer of the coatings consists of a cladding zone, a boundary zone, a thermal influence zone, and a substrate zone. The cladding zone mainly consists of equiaxed crystals away from the substrate and columnar crystals near the substrate. The coverslips consisted of FCC, BCC, and Laves phases because of the high entropy effect.

This process favors the strong adhesion of the coating to the substrate, homogeneous microstructure, and formation, but has less thermal damage caused by the fast heating and cooling rates involved in the process. The main disadvantages of laser doping of the surface are the formation of the peri-seam zone, high residual stresses and elemental dilution of the HEA coating and the substrate [101].

There is also a problem arising during laser processing, in the process of obtaining a coating on a substrate with a low melting point. The problem is that there is often a significant dilution of elements from the substrate. In [102], AlCoCrCuFeNi coatings on pure magnesium substrates were obtained by laser cladding. It was found that the Mg element in the matrix melts into the lower layer of HEA coatings. [103–104] synthesized AlFeCoCrNi coatings by laser surfacing on an aluminum substrate and found that laser treatment results in a composite microstructure of a HEA-rich BCC phase embedded in an aluminum-enriched matrix.

3.2.2 Magnetron sputtering method

Magnetron sputtering (MS) is also a frequently used method for producing HEA films and coatings. In MS, the main parameters affecting the properties of the coatings are the target composition, the bias voltage, and the gas flow rate. HEA coatings obtained by MS have excellent properties, such as high hardness and Young's modulus, wear resistance, thermal stability at elevated temperatures, and diffusion retardation [105, 106]. The properties of HEA coatings can be further improved by nitrogen doping, as the structure of HEA coatings transforms from amorphous to FCC [107–111]. In [112] it is noted that this method is considered one of the most effective methods of deposition of thin films based on HEA. In [113] the results of the study of FeCrNiTiZrAl coating obtained by MS, the coating characteristics are not inferior to high-entropy equiatomic alloys.

Most magnetron sputtered HEA coatings are nitride, carbide and boride HEA coatings with excellent wear, oxidation and irradiation resistance properties [114–118]. Most of these sputtered coatings have BCC or FCC solid solution phases with an amorphous structure compared to other coatings deposited by thermal spraying.

The main disadvantage of this method of coating production is the problem of working with strongly magnetic substances (Ni, Co, Fe). In MS it is impossible to achieve high-speed sputtering at low temperature for strongly magnetic materials, because almost all the magnetic flux cannot pass through the magnetic target, so it is impossible to add an external amplifying magnetic field near the surface of the target.

3.2.3 Electrochemical deposition

Electrochemical deposition makes it possible to deposit HEA films on substrates with complex geometry, conduct it at low processing temperatures and low energy consumption. Since electrochemical deposition does not require sophisticated equipment and expensive raw materials, it enables inexpensive synthesis of HEA films. In addition, by varying the deposition parameters, electrodeposition can easily control the composition, morphology, and thickness of the films [119–121].

In [120], the use of the pulsed electrochemical deposition method successfully led to the synthesis of thin films of the highly entropic CoCrFeMnNi alloy. It was noted that the morphology of the film surface and the composition of the HEA depended on various parameters of the applied pulse, such as the filling factor and frequency. The structure and surface morphology of electrodeposited CoCrFeMnNi coatings are described for the first time in the present work. The unified solid solution structure of the FCC type was identified by X-ray diffraction analysis. It can be concluded that the application of the strategy of HEA production by electrodeposition can provide a promising approach to the development of new metallic materials with remarkable properties. The main drawback of this method is the need to use toxic, environmentally unsafe electrolytes.

3.2.4 Thermal spraying

Recently, according to published articles in obtaining HEA gas-based coatings by thermal spraying, most researchers use plasma arc cladding, plasma spraying, high velocity oxy-fuel spraying (HVOF), and cold spraying methods. It is noted that not only the chemical composition of materials in HVOF-based thermal spraying is the main factor influencing the characteristics of the coating, but also the parameters of thermal spraying will play a significant role.

The plasma arc cladding process has many advantages for the synthesis of HEA films and coatings, such as high energy exchange efficiency, low thermal deformation of the part, low dilution of the substrate material, and good metallurgical bonding to the substrate [96, 127]. Typically, the HEA arc melting method is mainly fabricated or coated to avoid segregation during solidification [21].

Plasma spraying is widely used in the aerospace, automotive, petrochemical and mining industries for coatings and repair of parts [122–124]. The heat source of plasma spraying is a DC plasma arc, which is high enough to atomize and apply all types of coatings, including ceramics with high Tm. The plasma deposition method, a high-energy coating deposition procedure, is also widely used in HEA-based coating applications [125]. Compared to laser surface doping, plasma cladding has a higher blowing force, through which the molten coating materials are melted and mixed to achieve microstructure and characterization homogenization [125].

In contrast to plasma spraying, the heat source in HVOF is the heat of combustion of fuel and oxygen. Meanwhile, the HVOF sputtering process is characterized by a high particle jet velocity and a relatively low processing temperature [128]. So far, studies concerning HVOF-based coating materials have mainly involved the characterization of microstructural evolution, phase composition, and wear and oxidation behavior [129, 130].

In contrast to the traditional fusion-based coating manufacturing method, cold spraying (CS) is a newly developed solid-state coating method, so there is no oxidation, phase transformation or residual thermal stress. To date, HEA-related coatings deposited by the CS method are limited to HEA materials with low strength and high ductility because of the internal mechanism [131] of the CS method, such as FeCoNiCrMn type FCC [132].

Although these coating methods have demonstrated great potential related to HEA, much work is still needed from theory and applications to further expand the applicability in HEA systems, such as adapting processing parameters and using post-processing [133–134] after coating fabrication. Also, in recent works related to HEA-based coatings, a detonation spraying method has been started [135–137], this method is also related to the gas-thermal method.

New developments in terms of innovations, both in terms of starting materials for HEA-based coatings and in terms of the appropriate regulation of thermal spray processes, need to be investigated and developed to further expand the application in the production of HEA-based coatings with special microstructural characteristics and outstanding properties.

4 CONCLUSIONS

In this article the concept of HEA was considered, based on the analysis of the literature data we can make the main conclusions:

− Currently, there is no universal parameter that could predict the formation of structures in multicomponent systems of alloys. Determining the conditions governing the stability of phases in HEA by statistical analysis of the overall behavior of the constituent elements in multicomponent alloys is an urgent task.

− Materials related to HEA, both as bulk materials and as coatings, have a wide range of applications due to their outstanding physical, chemical and mechanical properties. A further trend of development will be the study of non-equimolar HEA and non-Cantor alloys. But the goal remains the same to use entropy and develop a certain combination to achieve the desired mechanical and functional properties. Just the same research on the nature of physical and chemical processes occurring during mechanochemical synthesis of powders and the development of ideas about the formation of the structure and properties of HEA under conditions of cryogenic mechanochemical alloying is a relevant area for research.

− The use of HEA in general seems promising and requires additional research both in the direction of finding strategies for creating practically significant compositions, and in the direction of increasing the properties of alloys for their further operation at elevated temperatures in aggressive environments.

At the moment, work is being done on the development and application of stable HEA structures. As described in many literatures HEA will be used to a greater extent in structural materials for operation at low and high temperatures. Since the diffusion processes at low temperatures are very slow, and the HEA will be in a metastable state for a long period of time, the same properties have structural alloys.

To design and develop HEA that have better mechanical properties under different environmental conditions stability in phases or strengthening mechanisms is an important aspect. From the reviews of research papers, it is only by using MA that solid solution phases with multiple phases and complex structures are formed. The transformation of phases into complex structures and multiple solid solution phases observed during consolidation and annealing indicate metastable phases formed during MA. This means that the research in this direction should be developed.

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ЖОҒАРЫ ЭНТРОПИЯЛЫҚ ҚОРЫТПАЛАР ТҰЖЫРЫМДАМАСЫНА ШОЛУ

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Мақалада жоғары энтропиялық қорытпаларды зерттеу нәтижелеріне шолу жасалады, олардың пайда болу принциптерін, жоғары энтропиялық қорытпалардың негізгі түсініктері мен қасиеттерін сипаттайды. Энтропиялық қорытпалардың қолданыстағы санаттары келтірілген. Жоғары энтропиялық қорытпалардың пайда болуынан туындайтын әсерлер сипатталған: жоғары энтропия, тордың бұрмалануы, баяу диффузия және араластыру әсері. Көп компонентті қорытпаларға арналған дәстүрлі термодинамикалық көріністер толықтыруды қажет етеді. Сондай-ақ, жоғары энтропиялық қорытпалардың фазалық құрамын болжау үшін Юм-Розери ережесін қолдану кейбір қиындықтарды көрсетеді, өйткені тор мен валенттіліктің бірдей түріне ие көптеген элементтерді таңдау қиын. Зерттеушілердің пікірінше, жоғары энтропиялық қорытпалардағы құрылымдардың қалыптасуын дұрыс болжауға мүмкіндік беретін жоғары энтропиялық қорытпалардың құрылымдық күйіне әсер ететін бірқатар параметрлер мен жағдайларды талдау нәтижелері сипатталған. Әдеби деректерді талдау көрсеткендей, қазіргі уақытта көп компонентті қорытпалар жүйелерінде құрылымдардың қалыптасуын дұрыс болжауға мүмкіндік беретін әмбебап параметр жоқ. Жоғары энтропиялық қорытпалардың ұнтақтарын дайындау әдістері ұсынылған. Лазерлік балқыту, магнетронды бүрку, электрохимиялық тұндыру және газ-термиялық бүрку сияқты жоғары энтропиялық қорытпаларға негізделген жабындарды алудың жиі қолданылатын әдістерінің нәтижелері қарастырылған. Осы әдістермен жабынды алу кезіндегі кемшіліктер сипатталған.

Түйін сөздер: *жоғары энтропиялық қорытпалар, көп компонентті қорытпалар, термодинамика, құрылым, жабу әдістері.*

ОБЗОР КОНЦЕПЦИИ ВЫСОКОЭНТРОПИЙНЫХ СПЛАВОВ

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В статье представлен обзор результатов исследований высокоэнтропийных сплавов, описывающие принципы их образования, базовые понятия и свойства высокоэнтропийных сплавов. Перечислены существующие категории энтропийных сплавов. Описаны эффекты, возникающие вследствие образования высокоэнтропийных сплавов: высокая энтропия, искажение решетки, замедленная диффузия и эффект перемешивания. Отмечается, что традиционные термодинамические представления для многокомпонентных сплавов требуют дополнения. Также отмечается, что применение правил Юм-Розери для предсказывания фазового состава высокоэнтропийных сплавов показывает некоторые затруднения, так как трудно подобрать большое количество элементов, имеющих одинаковый тип решетки и валентности. Описаны результаты анализа ряда параметров и условий, которые, по мнению исследователей, влияют на структурное состояние создаваемых высокоэнтропийных сплавов, учет которых мог бы позволить корректно предсказать формирование структур в высокоэнтропийных сплавах. Анализ литературных данных показал, что в настоящее время не существует универсального параметра, который мог бы позволить корректно предсказать формирование структур в многокомпонентных системах сплавов. Представлены методы подготовки порошков высокоэнтропийных сплавов. Рассмотрены результаты работ часто используемых методов получения покрытий на основе высокоэнтропийных сплавов, таких как лазерная наплавка, магнетронное напыление, электрохимическое осаждение и термическое напыление. Описаны недостатки при получении покрытий данными методами.

Ключевые слова: *высокоэнтропийные сплавы, многокомпонентные сплавы, термодинамика, структура, методы покрытия.*