



Research progress of high entropy alloy: Surface treatment improves friction and wear properties

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ABSTRACT

High entropy alloy (HEA) has high hardness, strength and excellent friction and wear properties, which also has broad application prospects in wear-resistant and high temperature coating, aerospace and military fields. However, one third of the world's total energy is consumed by various forms of friction or wear and tear, causing untold economic losses each year. The important way to reduce the wear is to increase the hardness of materials. The surface modification of high entropy alloy is an effective way to improve the friction and wear properties. In this paper, the research progress of high entropy alloys at home and abroad in recent years is reviewed, especially the surface treatment technology which can improve the friction and wear properties of high entropy alloys is introduced, and the influence of surface strengthening treatment on the friction and wear properties and mechanism of high entropy alloys is discussed. Surface treatment mainly includes surface mechanical strengthening, surface heat treatment strengthening, chemical heat treatment strengthening, surface laser cladding treatment, surface plasma treatment, surface electron beam treatment, vapor deposition technology. The purpose of this work is to provide a reference for the research on the friction and wear property of high entropy alloy, to provide corresponding ideas for the possible industrial demand in the future, and to provide the key research direction for future application.

1. Introduction

High entropy alloy is composed of a variety of alloy elements in equal or unequal atomic ratios and has many unique properties. Its concept has disrupted the traditional alloy design concept and innovated the alloy design method, resulting in a new type of multi-principal alloy with superior potential [1]. High entropy alloy has excellent wear and corrosion resistance, good plasticity and toughness, heat resistance, soft magnetic properties, radiation resistance, catalytic properties, bio-compatibility, and other properties. They have a wide range of applications and can be used as materials in a variety of fields, including tools, aerospace, chemical engineering and shipping, electronic communication, nuclear energy, and the petroleum industry [2–4]. Because of the high entropy effect, lattice distortion, and slow diffusion effect, high entropy alloy has excellent mechanical properties and is

suitable for wear conditions [5,6].

By enhancing the surface structure and residual stress distribution, surface strengthening is the improvement of the surface properties of metallic materials. The wear resistance of the material surface can be increased by using surface technology to create a specific coating or work hardening layer on the surface of the alloy to accomplish surface strengthening. Numerous researches [7–10] have shown that employing HEA coating can improve wear resistance and corrosion resistance. For example, the AlCoCrFeNi coating [11] prepared by electric spark deposition technology clearly demonstrated anti-friction and wear resistance properties compared with the hard Cr coating of traditional electroplating. According to the findings, the wear rate of the AlCoCrFeNi HEA coating was reduced by around 41 %, and the friction coefficient was only 0.25 to 0.33 as opposed to 0.6 to 0.73 for the hard Cr coating. Currently, adding and synthesizing alloy components with high

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hardness or strength to strengthen the matrix is the main focus of the review of high-entropy alloys in order to increase wear resistance, but there is still work to be done on the relationship between surface strengthening and wear resistance. Surface mechanical strengthening, surface heat treatment strengthening, chemical heat treatment strengthening, surface laser cladding, surface plasma technology, surface electron beam technology, and vapor deposition technology are used in combination with the wear mechanism of high entropy alloy. This paper summarizes the research status of strengthening technology to improve the surface wear resistance of high entropy alloy, to serve as a useful resource for the subsequent research.

2. Wear mechanism

One of the three main ways that materials fail is listed as wear. It broadly refers to the surface loss brought on by the relative motion of objects in an open or closed system of different media while being subjected to loads [12]. According to classical Archard law [13], the higher the hardness of the alloy, the better the wear resistance. Through the well-known Hall-Petch relationship, significant strengthening effect can be produced in high entropy alloys by refining grains to ultra-fine crystals, thus effectively improving the hardness [14]. In addition, toughness is also a key factor affecting wear, which represents the ability of the worn surface of an alloy to accommodate plastic deformation [15]. The wear mechanisms of high entropy alloy can be classified into abrasive wear, adhesive wear, oxidation wear, surface fatigue wear, and other types of wear based on the features of the surface failure mechanism.

The material loss was mainly caused by wedge-shaped ploughing due to strong plastic deformation caused by hard frictional subsets

(especially oxide particles generated during wear). Mu et al. [16] found the CoCrFeNiMo HEA coating had three different wear mechanisms: oxidation wear, adhesion wear, and abrasive wear. The surface roughness induced by extrusion and metal tearing parallel to the wear trajectory of the extrusion surface can effectively increase the coefficient of friction (COF). Erdogan et al. [17] detected the wear mechanism of fatigue-induced extrusion on the wear surface of CoCrFeNiTi_{0.5}Al_{0.5} HEA, so the alloy obtained an unusually high COF at 500 °C. According to the experiment [18] studying the effect of annealing on FeCoCrNiAl HEA coating, the wear mechanism of a non-annealed coating was primarily abrasive wear, that of a coating annealed at 500 °C was abrasive wear, adhesive wear, and adhesive wear. Jiang et al. [19] prepared three coatings of FeMnCoCr, FeMnCoCr/TiC and FeMnCoCr/TiC/CaF₂ on copper. In terms of wear morphology, the primary wear mechanism for FeMnCoCr coating was abrasive wear, the FeMnCoCr/TiC coating was abrasive wear and adhesive wear, and adhesive wear was the mainly wear mechanism for FeMnCoCr/TiC/CaF₂ coatings.

Tong et al. [20] investigated the effect of LSP (laser shock peening) treatment on the wear process of the CrMnFeCoNi HEA, as shown in Fig. 1. After LSP, the rough surface was smoothed down and had the high microhardness and residual which make it resistant to the incursion of the counter ball. Under the same load, the hard interface between the count ball and the treated specimens was smaller than the untreated soft interface, resulting in the reduction of the total friction coefficient and wear. The main wear mechanisms of untreated and treated specimens were abrasive wear and adhesive wear. The wear mechanism of the FeCoCrAlCu HEA containing TiC [21] was shown in Fig. 2. The oxide layer on the coating surface was gradually damaged due to the plow

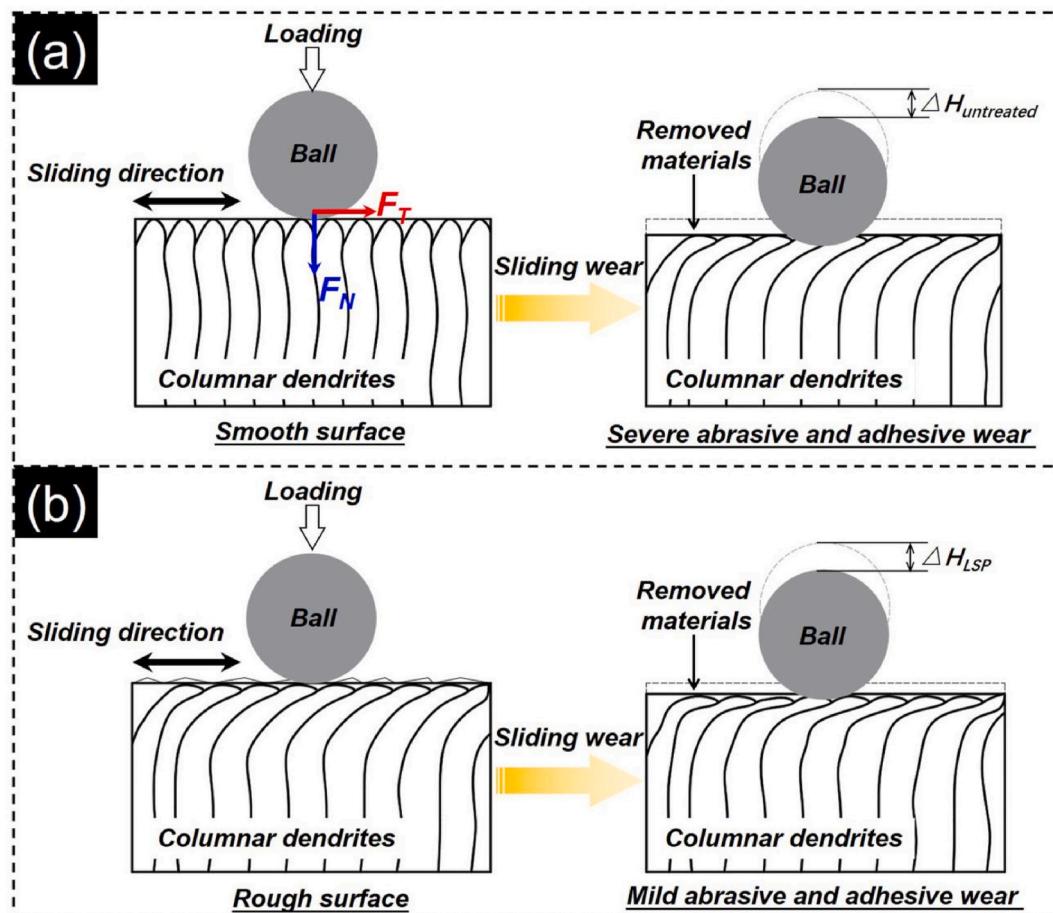


Fig. 1. Schematic presentation of wear processes of (a) untreated and (b) LSP treated specimens.

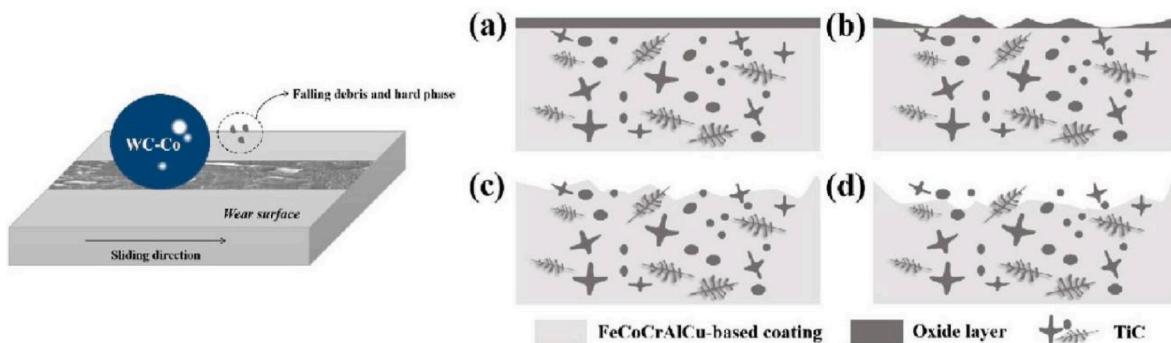


Fig. 2. Wear mechanism of TiC reinforced the FeCoCrAlCu-based HEA coating.

sliding of the friction pairs, resulting in scratches, plastic deformation and debris. After the contact between the HEA matrix and the friction pair, there was slight plastic deformation and flaked off on the coating surface, and micro-cracks on the wear surface (Fig. 2(a) and (b)). Meanwhile, TiC, the reinforcing phase in the composite, was gradually exposed to the wear surface (Fig. 2(c)). With the decrease of the bonding force with the high entropy alloy, TiC particles were gradually precipitated from the coating, as shown in Fig. 2(d).

Refractory high entropy alloy (RHEA) exhibits extremely high hardness and superior wear resistance when compared to commercial superalloys (Inconel 718), structural steel (45 steel), stainless acid resistant steel (316L stainless steel), tool steel and so on [22–25]. The findings of the study demonstrated [24]: The wear mechanism of Al_xCrTiMo RHEA at 25 °C and 200 °C was mostly abrasive wear. The wear mechanism at 400 °C, was a comprehensive effect of abrasive wear, adhesive wear, and plastic deformation. At temperatures between 600 °C and 800 °C, adhesive wear predominated, with only minor amounts of abrasive wear and plastic deformation. In the experiment of doping Si element in the NbTaWMo RHEA [25], the change in the wear mechanism of the alloy from 25 °C to 800 °C can be glimpsed in Fig. 3. On the surface of the wear marks, a substantial amount of fine abrasive grains accumulated between 25 °C and 200 °C, while bigger abrasive grains accumulated at 400 °C. In the middle and low-temperature stages, abrasive wear was the main wear mechanism because of the cutting effect that dual silicon nitride balls had on the alloy. At 600 °C and

800 °C, there was not only the buildup of abrasive grains on the surface of the abrasive scar morphology, but also the formation of a black oxide enamel layer that inhibited the wear of high entropy alloys, as shown in Fig. 3(f), the main wear mechanism of which was the combined effect of abrasive and oxidative wear.

3. Surface treatment modification method

Surface treatment of materials can greatly reduce the friction and wear of surfaces in contact with each other or generating relative motion, thus reducing the consumption of materials and energy. Commonly used surface matrix strengthening treatment methods include surface mechanical strengthening treatment, surface chemical heat treatment, surface heat treatment and other strengthening; and surface protective layer strengthening mainly include surface laser cladding technology, surface plasma technology, vapor deposition technology and so on. The content of this part can be roughly understood in Fig. 4.

3.1. Surface mechanical strengthening treatment

Surface deformation strengthening can make metal parts surface plastic deformation and work hardening, causing changes in the surface microstructure, improving the fatigue strength, wear resistance and corrosion resistance of metal parts, and its process mainly has three processes such as blasting (peening), rolling and hole extrusion [26,27].

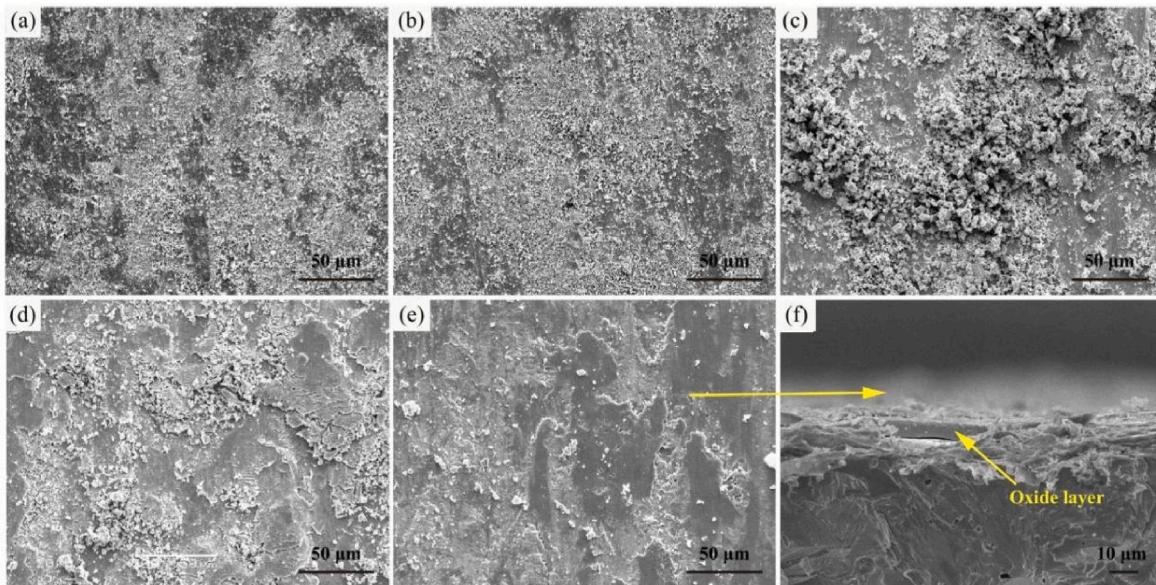


Fig. 3. SEM images of the wear marks of NbTaWMo RHEA at different temperatures: (a) 25 °C, (b) 200 °C, (c) 400 °C, (d) 600 °C, (e) 800 °C, (f) abrasion profile at 800 °C.

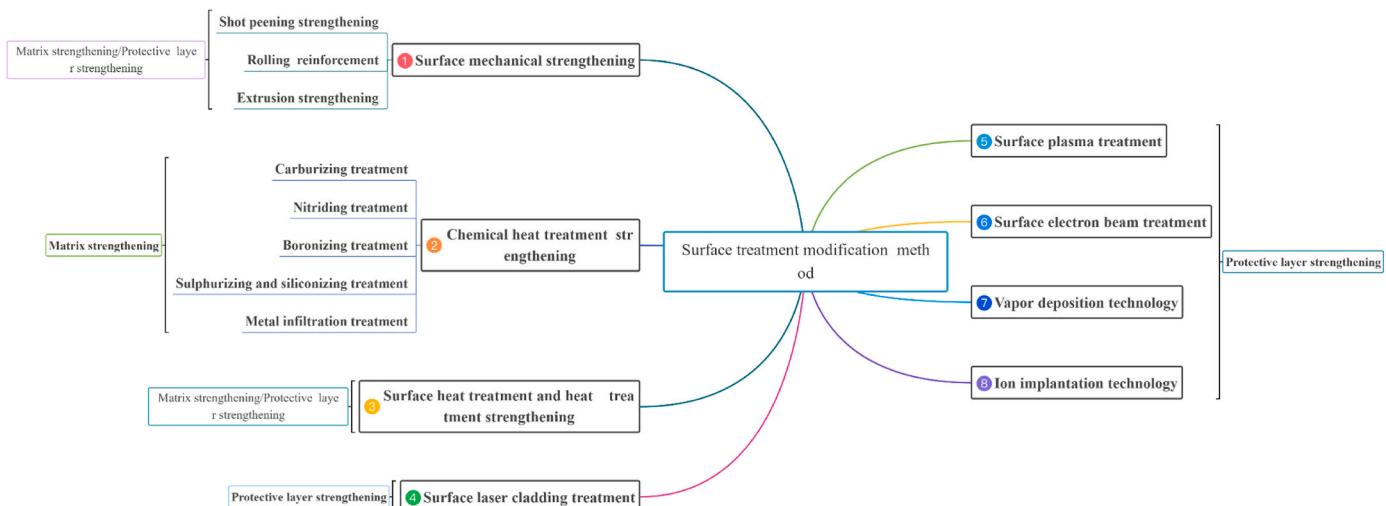


Fig. 4. Mind map of surface treatment modification methods for improving friction and wear properties of high entropy alloy.

3.1.1. Shot peening

Shot blasting is a surface strengthening method below the recrystallization temperature that can greatly increase the resistance to fretting wear and has the benefits of ease of use, low energy consumption, high effectiveness, a broad application range and others [27]. When high energy pellets attached with high entropy alloy powder impact the metal matrix surface, the matrix surface will produce plastic deformation under the action of high energy. The high entropy alloy powder is combined with the matrix surface through mechanical force to form a high entropy alloy film, so as to improve the mechanical properties and wear resistance of the metal matrix.

Liu et al. [28] prepared NiFeCuCrCo HEA film on magnesium alloy surface by surface high-energy shot peening method. Through mechanical deformation, NiFeCuCrCo HEA powder was tightly bonded on the surface of AZ31 magnesium alloy to form a protective high entropy alloy layer. The high entropy alloy coating greatly improved the surface mechanical properties and corrosion resistance of the magnesium alloy. The corrosion rate was about 1.5 % of that of the substrate, and the wear resistance was more than double. The wear mechanism of AZ31 magnesium alloy was abrasive wear and the high entropy alloy coating was adhesive wear.

Shot peening is taken as post-treatment, which plays an important role in refining surface grains and improving the strength, hardness, toughness and plasticity of materials [20,29,30]. Wang et al. [29] prepared FeCoCrMn HEA coating on Q235 steel matrix by plasma cladding method, and improved the mechanical properties of the coating by shot peening. In the process of shot peening, surface grain refines, dislocation density and lattice distortion increase, dislocation movement was

blocked and accumulated, resulting in strain hardening of alloy and hardness improvement of materials. The hardness of the material increased from 400 HV to 460 HV, the friction coefficient decreased from 0.53 to 0.38, the regional wear decreased from 0.005 g to 0.004 g, and the wear mechanism changed from abrasive wear and adhesive wear to adhesive wear. Tong et al. [20] prepared CrMnFeCoNi HEA by laser additive manufacturing and laser shock shot peening. After LSP, ultrafine grains and compressive residual stress were generated on the surface of the sample, as shown in Fig. 5. The high-density dislocation structure and deformation twins provided strong abrasion resistance. The friction coefficient and wear rate of the alloy were reduced obviously, showing excellent wear resistance. Tong et al. [30] prepared CrMnFeCoNi HEA by laser directed energy deposition technology, and improved its performance by LSP. After LSP, severe plastic deformation was induced on the surface of the alloy, which not only generated residual compressive stress, but also changed the surface stress state from tensile stress to compressive stress, increased the dislocation density, refined the grain, and significantly improved the strength and ductility of the alloy.

In addition to strain hardening and strengthening induced by LSP and strain hardening, the introduction of gradient structure and chemical gradient could also enhance the metal material strength and ductility, which indirectly serves the goal of enhancing materials hardness and wear resistance. Fu et al. [31] treated CrFeCo-NiMn_{0.75}Cu_{0.25} HEA using LSP, which showed significant improvement in its mechanical properties due to the introduction of gradient microstructures including sub-grain, dense dislocations and nano-scale twin around the surface. The CrCoNi medium entropy alloy [32] was

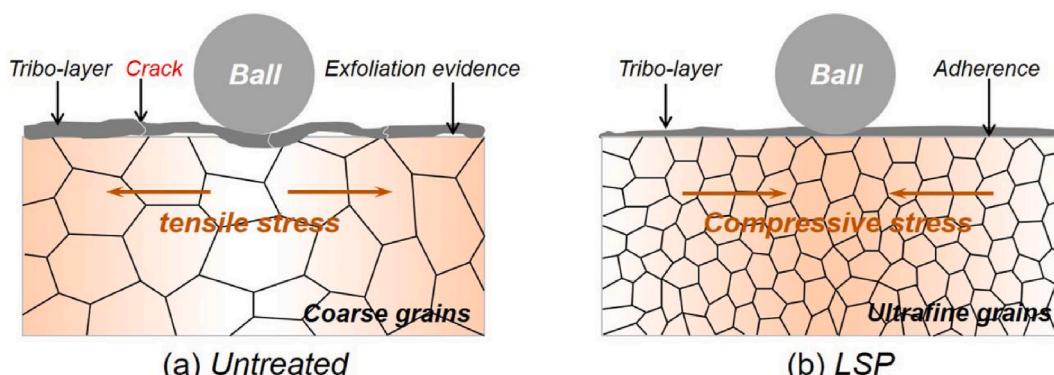


Fig. 5. Schematic diagram of wear and damage mechanism: (a) untreated samples, (b) treated samples.

successfully given a gradient nano-grain structure by using high energy shot peening technique. It gained outstanding ductility and strain hardening ability at high strength due to the heterogeneous deformation-induced hardening effect of heterogeneous structure, and CrCoNi MEA demonstrated excellent strength-plasticity synergistic performance. By using rotating accelerated shot peening technique, Hasan et al. [33] treated CrCoFeNiMn HEA to create various gradient structure profiles. By changing the process settings, various gradient structures, such as gradient twins, gradient dislocations, and gradient nano-twins, were included into the alloy. The findings demonstrated that CoCrFeNiMn HEA had simultaneously increased strength and ductility. By rotating accelerated shot peening technology, Liang et al. [34] applied high strain and high strain rate to the material surface and created gradient nanostructure, realizing surface nano-crystallization of the Ni₂FeCoMo_{0.5}V_{0.2} HEA. The explanations for the alloy's grain refining were deformation twins and dislocation activity. By using rotating accelerated shot peening technology, An et al. [35] encouraged the production of deformation twins and stacking faults in CoCrFeMnNi HEA, resulting in a graded grain size gradient microstructure with a significant number of nano-twins. The combined effects of twin strengthening, grain boundary strengthening, dislocation strengthening and heterogeneous deformation strengthening were what gave the HEA its high strength and ductility.

3.1.2. Rolling reinforcement

A non-cutting processing method known as rolling strengthening technology can greatly increase the fatigue strength of parts and lessen notch sensitivity [27]. It is appropriate for big components with a straightforward geometry, particularly the stress concentration of systems with abrupt size changes.

Cui et al. [36] treated FeCrCoNiMn HEA coating by ultrasonic rolling. The residual stress in the cladding layer was eliminated, and the grain of the coating surface was refined to form a dense alloy coating with high entropy. Compared with the substrate, the surface hardness and wear resistance of the coating were increased by about 1~1.5 times, and the corrosion resistance is increased by 2~2.5 times. Cui et al. [37] performed ultrasonic surface rolling treatment on the surface of CrMnFeCoNi HEA. After ultrasonic surface rolling treatment, nano-layer appeared in the high entropy alloy due to grain refinement and work hardening, and the maximum surface hardness was 352.28 HV, 1.5 times that of the untreated sample. The coefficient of friction decreased from 0.71 to 0.62, and the amount of wear decreased from 1.6 mg to 1.2 mg, showing better wear resistance. The wear mechanism was stratified wear, abrasive wear and oxidation wear. Qin [38] strengthened the surface of the CrMnFeCoNi HEA using ultrasonic surface rolling technology. The results show that after ultrasonic surface rolling treatment, the grain refinement and plastic strain can greatly improve the hardness of the surface layer, the average friction coefficient and wear quantity

were reduced, improving wear resistance. The main wear mechanisms in single layer, double layer and ultrasonic rolling CrMnFeCoNi HEA were layered wear, abrasive wear and adhesive wear (adhesive wear model was shown in Fig. 6), abrasive wear and layered wear, respectively. Li [39] treated CoCrFeMnNiTi HEA coating by ultrasonic surface rolling. After ultrasonic surface rolling treatment, due to the increase of fine grain strengthening, dislocation strengthening and residual stress, the microhardness of CoCrFeMnNiTi HEA was increased by 38 %, the abrasion depth was reduced by 30 %, the volume wear rate was reduced by 63 %, significantly improving the wear resistance. The wear mechanism changed from abrasive wear to oxidative wear and abrasive wear. After ultrasonic/mechanical rolling treatment, Xie et al. [40] and Chen et al. [41] respectively formed gradient microstructure on the surface of the high entropy alloy, showing excellent strength and ductility, and significantly improved wear resistance and fatigue properties under common faults.

3.1.3. Extrusion strengthening

To improve the material's fatigue resistance and stress corrosion resistance, hole extrusion uses the plastic deformation of metal under the influence of an external force to deform and strengthen the surface of the inner hole of the workpiece [27].

Constant channel Angle extrusion mainly refines the grains of high entropy alloy through almost pure shear in the deformation process, thus significantly improving the mechanical and physical properties of materials [42–45]. Picak et al. [42] activated CoCrFeMnNi HEA by equal channel angular pressing at high temperature to induce twin-induced plasticity and phase-change induced plasticity. The equal channel angular pressing mold channel is shown in Fig. 7. After equal channel angular pressing treatment, a composite microstructure was formed with high defect density, refinement grains, and deformation induced twinning and ε-martensite. Due to grain refinement, dislocation density, uneven grain size distribution, and accidental formation of deformation twins and ε-martensite, the alloy exhibited a good strength-toughness combination at room temperature. The (AlFeNiCrCoTi_{0.5})p/6061 Al-based composites [43] were created by using cold isostatic pressing and equal channel angular pressing. The diffusion interface occurred after equal channel angular pressing, and the strengthening phase was tightly integrated with the matrix, requiring an increase in material strength. Rymer et al. [44] developed super fine-grained CrFeCoNi HEA via equal channel angular pressing and spark plasma sintering. The mechanical properties (hardness and tensile properties) of the alloy were significantly enhanced after equal channel angular pressing. Hammond et al. [45] produced a the AlFeMgTiZn HEA with fully consolidated low-density by equal channel angular pressing, combining mechanical alloying and severe plastic deformation. It was the intense plastic deformation processing that made the alloy have reasonable mechanical properties and high hardness.

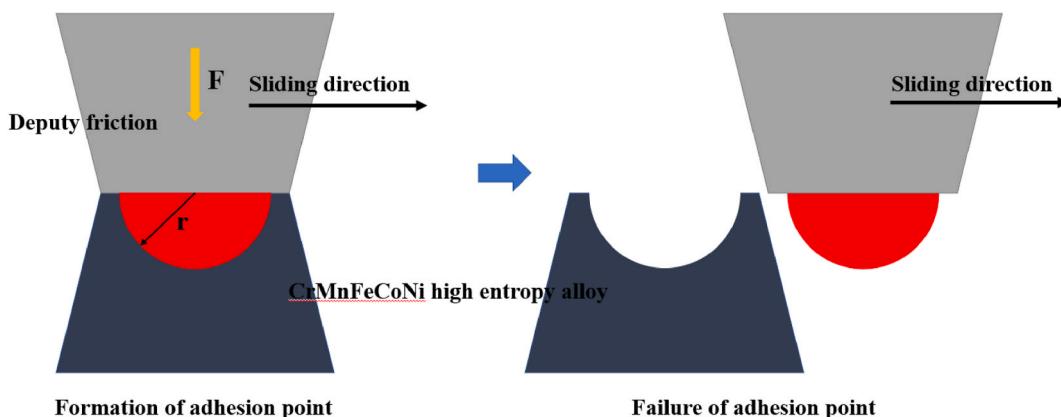


Fig. 6. Adhesive wear model of single layer CrMnFeCoNi HEA.

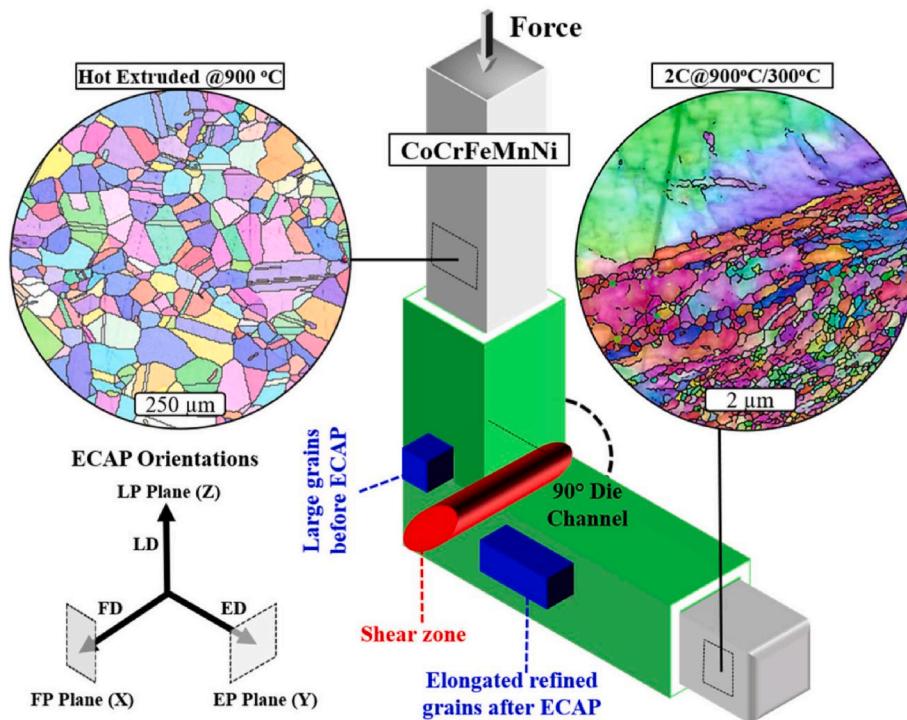


Fig. 7. The schematic diagram of the ECAP die channel of the CoCrFeMnNi HEA. LD: longitudinal direction, FD: flow direction, ED: extrusion direction. Illustration: Electron backscattering diffraction images of two ECAP channels hot extruded at 900 °C and processed at 900 °C/300 °C.

Li et al. [46] conducted forward extrusion, reverse extrusion and repeated reciprocating extrusion on AlCoCrFeNi HEA. In the process of extrusion, the grains were coarser and deformed continuously, so the grains were refined and the comprehensive mechanical properties of high entropy alloy were improved. For the CrMnFeCoNi HEA, Pu et al. [47] used quasi-static extrusion processing and short-time heat treatment to enhance the characteristics. The structural inhomogeneity gave the alloy a good strain hardening capacity at high flow stress levels due to the severe plastic deformation that refined the grain and the subsequent heat treatment that partially recrystallized the refined grain. This alloy had a much higher strength than uniform alloy while yet having good elongation.

Currently, research on the surface mechanical strengthening technology about the high entropy alloy can not only increase the hardness of the alloy's surface hardening layer to improve wear resistance, but can also introduce gradient microstructure or twinning into the high entropy alloy, and can optimize the friction and wear performance of the high entropy alloy by fine grain strengthening, dislocation strengthening, twinning strengthening, and heterogeneous deformation strengthening. A synergistic increase in strength and extensibility is achieved at the same time. Research in this field still has to be expanded, for example, ultrasonic nanocrystalline surface modification technology. The ultrasonic nanocrystalline surface modification treatment which is a technique for mechanical strengthening that has recently gained a lot of interest in industrial applications can increase surface hardness, refine subsurface layer grain size, and provide substantial compressive residual stress to improve wear resistance.

3.2. Chemical heat treatment strengthening

Chemical heat treatment strengthening is to infiltrate the active atoms in the medium into the surface layer of the alloy or form a coating layer of a certain compound, which can change its chemical composition and structure and achieve the purpose of surface strengthening [48]. Carburizing, nitriding, boriding, and sulfurizing treatment can effectively improve anti-wear and lubrication performance of the HEA.

3.2.1. Carburizing treatment

During the carburizing process, both the carbide and the solid solution of supersaturated carbon atoms precipitated from the high entropy alloy can effectively impede dislocation movement, and the strength and hardness as well as the wear resistance of the alloy are improved under the effect of dispersion strengthening and solid solution strengthening. At the same time, the carbon film itself has excellent low-friction characteristics, which is also conducive to improving the high-entropy alloy coating and achieving low friction.

Zhang et al. [49] used solid carburization to surface modify CoCrFeNiC HEA. The results showed that the solid carburizing process could improve the wear resistance of the entropy high alloy, as shown in Table 1, with the carburized layer having lower wear, wear rate and higher wear coefficient than the substrate. Nano-hardness (H_n) and reduced elastic modulus (E_r) qualitatively evaluate the wear resistance of carburized specimens, and the ratios H_n/E_r (related to resistance to elastic strain damage) and H_n^3/E_r^2 (indicating resistance to plastic deformation) can be used as parameters to describe wear resistance. Due to the precipitation hardening of the carbide precipitation phases M_7C_3 and $M_{23}C_6$ and the solid solution strengthening effect of the carbon atoms (Fig. 8(c)), the maximum hardness of the carburized surface was twice as high as that of the interior (Fig. 8(b)) and the ratios H_n/E_r and H_n^3/E_r^2 were also significantly higher on the surface than the matrix (Fig. 8(d)), resulting in a better wear resistance of the surface. Peng et al. [50] performed surface modification of CoCrFeNi HEA by low-temperature gas carburization. After low-temperature gas

Table 1

Average true depth (h_{true}), wear volume (V), wear rate (W) and wear resistance coefficient (R) of scratches calculated at different distances from the surface.

Location (mm)	h_{true} (nm)	V (mm ³)	W (10 ³ nm ²)	R (10 ¹¹ Pa)
20	54.4	0.223	22.3	1.35
60	67.2	0.259	25.9	1.16
120	68.0	0.263	26.3	1.14
200	86.3	0.339	33.9	0.85
500	94.6	0.366	36.6	0.82

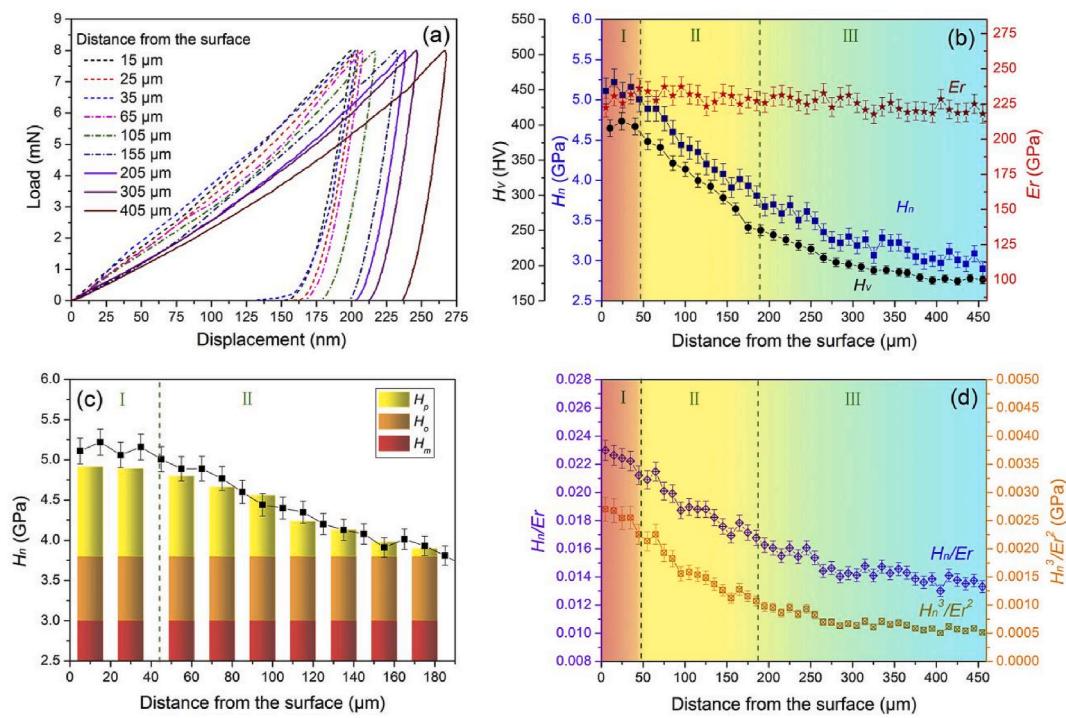


Fig. 8. (a) The nanoindentation load-depth curves at different distances from the surface of the carburized sample. (b) Distributions of the Vickers hardness (H_v), nano-hardness (H_n), and reduced elastic modulus (Er) from the surface to the matrix. (c) The hardness contributions from different hardening mechanisms in the region I and II. H_m is the hardness of the CoCrFeNi matrix; H_o is the hardness contributed by the strengthening of the solid solution of carbon atoms, H_p is the hardness contributed by the strengthening of the carbide precipitation phase. (d) Variations of the H_n/Er and H_n^3/Er^2 with different distances.

carburization, a carbide-free supersaturated interstitial solid solution of carbon with FCC structure was formed on the surface. Due to the solid solution strengthening of interstitial carbon atoms and strain hardening caused by lattice expansion, the surface hardness increased significantly, reaching 1221 HV, which was approximately five times that of the matrix.

The surface hardness and wear resistance of the high entropy alloy are closely related to the diffusion of carbon, the size and distribution of the carbide precipitation phase, etc. Li et al. [51] treated CuCoCrNiFeC HEA by solid carburizing method. After the carburizing treatment, the CrFe₇C_{0.45} and M₇C₃ carbides precipitated on the matrix of the alloy. Due to diffusion strengthening, the surface hardness of the alloy can be increased by up to 50 % compared to the matrix. As the carbide density decreases, the hardness gradually decreases from the surface layer to the core. Sun [52] carried out solid carburizing treatment on CuCoCrNiFe HEA. It was found that the hardness and wear resistance of the alloy increased under the diffusion strengthening effect of M₂₃C₆ and M₇C₃ carbides. The wear resistance of the alloy with fine carbides within the carburized layer was stronger than that of the sparse coarse carbides.

Ju et al. [53] used an encapsulated solid infiltration process for the surface modification of HfZrTiTa HEA, selecting activated carbon as the carburizing agent and holding it at 700 °C–900 °C for 2 h–10 h. The results showed that the maximum surface hardness of the alloy was 1590.3 HV, much higher than that of untreated HfZrTiTa HEA. Compared to the matrix, the surface hardness performance increased by 3–5 times and the high temperature oxidation resistance increased by about 23.07 %.

3.2.2. Nitriding treatment

High entropy alloy in the nitriding process will occur solid solution strengthening phenomenon, which can improve the strength of the alloy surface layer, but also in the surface of some hard nitride phase (such as AlN, CrN, TiN and FeN, etc.) to improve the wear resistance of the nitriding layer. In the frictional wear process, the nitriding layer to a

certain extent avoids direct contact between the frictional substrate and the matrix, which benefits to reduce the wear rate and improve the wear resistance of the alloy.

Liu et al. [54] used salt bath nitriding for surface strengthening of FeCrMnNiAl_{0.2}Ti_{0.1} HEA. The results showed that salt bath nitriding could effectively improve the wear resistance and tribological behavior of the high entropy alloy. After salt bath nitriding, a high hardness nitride layer was formed on the alloy surface, which played the role of surface strengthening so that only slight abrasive wear occurred in the nitriding sample. The hardness can be increased from a maximum of 311 HV in the as-cast phase to 1080 HV, and the wear rate was reduced from 0.107 mm³/(N·m) to 0.025 mm³/(N·m) and the friction coefficient from 0.729 to 0.631.

Numerous studies have made clear [55–63] that plasma nitriding technology can significantly improve the hardness and wear resistance of high entropy alloy surface. Hou et al. [55] found that the wear resistance of Al_xCoCrFeNi HEA was increased by 4–18 times after plasma nitriding treatment. Tang et al. [56] found that compared to conventional nitriding steels, the nitriding alloys exhibited 25–54 times more resistance to adhesive wear than conventional alloys. Ma et al. [57] used the plasma nitriding method to nitride AlCrW_xTa_yNb_z films. The results showed that under the bombardment of nitrogen ions, the densities of the films increased, the surface roughness decreased, and the microhardness and elastic modulus both increased gradually.

According to Leyland's theory, the corresponding H/E and H^3/E^2 both increased and the films exhibited good frictional properties. There are two main mechanisms by which plasma nitriding increases the hardness of the alloy: (i) solid solution strengthening effect. The nitrogen atoms diffuse within the alloy lattice, increasing the lattice distortion and playing a solid solution strengthening role. (ii) Diffusion strengthening effect. The nitrogen and other metal elements form a harder nitride structure, which plays a diffusion strengthening role.

The nitriding process was beneficial to the improvement of wear resistance of high entropy alloy [58]. The untreated sample had large

wear area and poor wear resistance, while the wear loss of nitriding sample was much lower than that of untreated sample. Zhou et al. [59] used plasma nitriding technology to treat Al_xCoCrFeNi HEA, and found that AlN and CrN formed on the surface of the coating after nitriding, resulting in a microhardness about twice that of the non-nitriding coating, and better wear resistance than the non-nitriding coating, as shown in Fig. 9. Al element is a strong nitrogen/oxygen reaction element, when the content of Al reaches a certain degree, oxidation friction film and a certain amount of nitride will be generated. Due to the increase of the number of hard nitride phase and the volume fraction of BCC phase, the coating hardness and wear resistance are greatly improved.

Wang et al. [60–62] used plasma nitriding to prepare nitriding layers on the surface of AlCoCrFeNi HEA and Al_{1.3}CoCuFeNi₂ HEA and investigated the frictional properties of the nitriding and cast high entropy alloys under different conditions. The results showed that the hardness of the nitriding alloys was increased due to the hard phases (AlN, CrN and Fe₄N) and solid solution strengthening, and the wear resistance of the nitriding alloys was better than that of the as-cast alloys under the same conditions.

Lan et al. [63] prepared nitriding layers on Ni₄₅(FeCoCr)₄₀(AlTi)₁₅ HEA using plasma nitriding method. The results showed that the nitriding treatment resulted in the formation of hard phases such as AlN, CrN, Fe₄N and TiN on the surface, with a significant increase in hardness from 8.7 GPa in the as-cast state to 14.5 GPa in the nitriding state. Seeing Table 2, it can be concluded that the wear rate and friction coefficient were lower than under air conditions due to the liquid flow, cooling and lubricating effects of deionized water and acid rain; and because of the corrosive effect of acid rain effect, which caused the protective oxide film to soften and formed a softer powder rather than hard particles during sliding, its wear rate and coefficient of friction were reduced [60–63].

3.2.3. Boriding treatment

The boron penetration layer exhibits excellent wear resistance due to the hard borides and severe solid solution strengthening, resulting in a higher surface hardness and a larger thickness of the high hardening layer.

Hou et al. [55] used solid boron penetration in Al_{0.25}CoCrFeNi HEA to prepare a boron layer of approximately 50 μm thickness. Due to the larger thickness and harder hardness of the boron infiltrated layer, with severe solid solution strengthening, the hardness of the boron alloy was nearly 1136 HV, which was six times that of the borated alloy. The friction coefficient and wear rate of the un-borated alloy were 0.80 and $1.468 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$, while those of borated alloy were 0.53 and $1.25 \times 10^{-5} \text{ mm}^3/(\text{N}\cdot\text{m})$, with twelve times the wear resistance of the

un-borated alloy. Hu et al. [64] treated CoCrFeNiMn HEA by powder-pack boriding. Found CoCrFeNiMn HEA surface microhardness significantly increased to $23.8 \pm 1.4 \text{ GPa}$, the friction coefficient was reduced from 0.86 to 0.48. Hou et al. [65] performed solid boriding on Al_{0.25}CoCrFeNi HEA with a boron layer thickness of 20~50 μm. It was found that the surface strength and yield strength of the borated alloy were increased due to the hard boride layer and the boron-induced solid solution strengthening. After 9 h of treatment at 900 °C, the surface hardness increased from 188 HV to 1265 HV, 6.7 times that of untreated, and the yield strength increased from 195 MPa to 265 MPa.

Lindner et al. [66] used powder-packed boriding to precipitate-harden CoCrFeNi HEA and CoCrFeMnNi HEA, and proposed that surface hardening of ductile high entropy alloys could improve wear resistance. Due to the formation of boriding layer, the microhardness and wear resistance of the alloy were significantly improved. The wear mechanism of untreated alloy was adhesive wear, and that of borated alloy was abrasive wear. Wu et al. [67] boronized Al_{0.1}CoCrFeNi HEA at 900 °C, forming a boron layer with a total thickness of about 17.3~58.5 μm. Due to the formation of a large number of hard borides on the surface and the interstitial solid solution strengthening effect of the boron atoms, the hardness of the borated alloy can reach 1398 HV, which was approximately seven times that of un-borated. With the increase of boron penetration time, the average friction coefficient and wear rate of the boron alloy under dry conditions were 0.41 and $5.86 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$, and the wear mechanism changes from delamination and abrasive wear to polishing action; the average friction coefficient and wear rate in water are 0.22 and $1.89 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$, and the wear mechanism changes from two-body sliding abrasive wear to three-body rolling abrasive wear.

Günen [68] boronized Co_{1.19}Cr_{1.86}Fe_{1.30}Mn_{1.39}Ni_{1.05}Al_{0.17}B_{0.04} HEA and formed a composite boride layer with a thickness of 31.95~64.36 μm and a surface hardness of 23.49~28.09 GPa. The borated alloy showed better wear resistance in air and 5% HCl, 5~30 times that of the casting; the wear rate and friction coefficient in 5% HCl was lower than in air due to the lubrication and cooling effect provided by the HCl solution, as shown in Fig. 10. In air, the wear mechanism of the casting was abrasive wear accompanied by plastic deformation, while that of the borated alloy was a combination of abrasive wear and polishing action; in 5% HCl, the wear mechanism of the casting was abrasive wear accompanied by pitting wear, while that of the borated alloy was abrasive wear accompanied by oxidative wear and pitting wear.

3.2.4. Sulfurizing and siliconizing treatment

The sulfide layer prepared on the surface of the high entropy alloy has good friction and wear reduction properties and the lubricating film formed by its sulfide boundary allows the friction system to operate for a

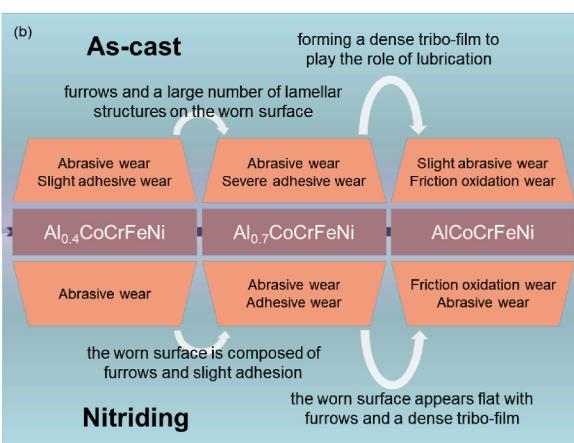
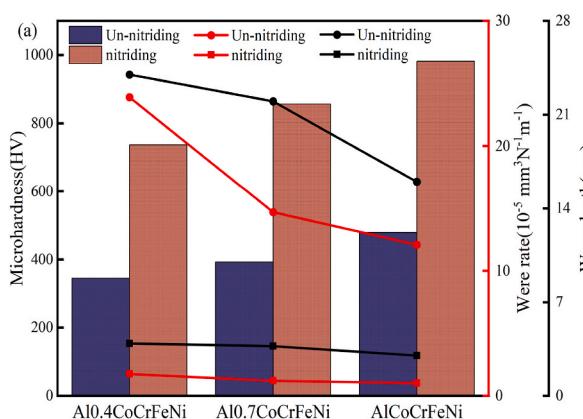


Fig. 9. (a) Microhardness, wear rates and wear depth of Al_xCoCrFeNi HEA coatings with as-cast and nitriding. (b) Mechanism of Al_xCoCrFeNi HEA coatings with as-cast and nitriding.

Table 2

Comparison of wear properties and wear mechanism between as-cast alloy and nitriding alloy under different environmental conditions.

High entropy alloy	Microhardness (HV)	Wear rate ($10^{-4} \text{mm}^3/(\text{N}\cdot\text{m})$)			Friction coefficient			Wear mechanism		
		Air	Water	Rain	Air	Water	Rain	Air	Water	Rain
As-cast AlCoCrFeNi [60]	522	1.8	1.6	0.7	0.429	0.301	0.302	Abrasive wear	Abrasive wear	Abrasive wear, corrosive wear
Nitriding AlCoCrFeNi [60]	720	0.39	0.32	0.28	0.512	0.681	0.213	Abrasive wear, adhesive wear	Abrasive wear, adhesive wear	Corrosion wear
As-cast Al _{1.3} CoCuFeNi ₂ [61]	340	12	1.2	2.3	0.62	0.42	0.36	Abrasive wear, delamination wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear
Nitriding Al _{1.3} CoCuFeNi ₂ [61]	587	10	105	1	0.51	0.42	0.19	Adhesion wear, abrasive wear	Abrasive wear, adhesive wear	Abrasive wear, abrasive wear
As-cast Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 5 N [62]	5.91	5.25	6.61	0.21	0.38	0.15	Abrasive wear, delamination wear, oxidation wear	Abrasive wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, delamination wear
As-cast Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 7 N [62]	7.31	3.89	7.3	0.42	0.41	0.14	Abrasive wear, delamination wear, oxidation wear	Abrasive wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, delamination wear
As-cast Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 10 N [62]	9.85	4.35	5.9	0.38	0.49	0.10	Abrasive wear, delamination wear, oxidation wear	Abrasive wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, delamination wear
As-cast Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 12 N [62]	7.01	6.69	4.65	0.18	0.38	0.20	Abrasive wear, delamination wear, oxidation wear	Abrasive wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, delamination wear
Nitriding Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 5 N [62]	1.60	2.02	0.4	0.6	0.5	0.4	Delamination wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, corrosive wear, adhesive wear	Abrasive wear, adhesive wear
Nitriding Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 7 N [62]	3.85	3.35	0.43	0.6	0.32	0.39	Delamination wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, corrosive wear, adhesive wear.	Abrasive wear, adhesive wear.
Nitriding Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 10 N [62]	4.73	4.04	0.58	0.58	0.42	0.38	Delamination wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, corrosive wear, adhesive wear.	Abrasive wear, adhesive wear.
Nitriding Ni ₄₅ (FeCoCr) ₄₀ (AlTi) ₁₅ A load of 12 N [62]	2.66	4.26	2.8	0.58	0.31	0.38	Delamination wear, adhesive wear	Abrasive wear, delamination wear	Abrasive wear, corrosive wear, adhesive wear.	Abrasive wear, adhesive wear.

longer period of time at a lower coefficient of friction. And the dense and continuous oxide film in the silicon infiltrated layer can avoid direct contact between the high entropy alloy and its counterpart, reducing the friction coefficient to some extent.

Cui et al. [69] subjected CoCrFeNiSi_{0.4} HEA and CoCrFeMoNi HEA to ionic sulfide treatment to form 4 μm FeS and FeS/MoS₂ lubricating films on the surface, respectively. The results showed that the ionic sulfide treatment can significantly improve the friction reduction performance of the high entropy alloys. After the sulfide treatment, the friction coefficients of CoCrFeNiSi_{0.4} HEA and CoCrFeMoNi HEA were reduced respectively from 0.6 to 0.47 to 0.152 and 0.12, as shown in Fig. 11. Due to the synergistic effect of FeS and FeS/MoS₂ lubricating phases, the multiple sulfide films had a lower coefficient of friction and wear than the single sulfide films, as shown in Fig. 12, and the CoCrFeMoNi HEA had better friction reduction and wear resistance than the CoCrFeNiSi_{0.4} HEA.

The frictional properties of CoCrFeMoNi HEA were further investigated [70]. It was found that the friction coefficient was reduced from 0.47 for the untreated alloy to 0.15 for the sulfide alloy, and the wear was reduced from 15.1 mg to 4.25 mg. The wear resistance of the sulfide alloy was significantly better than that of the untreated alloy. During the friction process, a mixed mechanism of abrasive wear, fatigue wear and adhesive wear occurred in the untreated alloy, and the wear mechanism in the sulfide alloy was slight adhesive wear and friction wear. Yan et al. [71] used a solid silicon infiltration technique to prepare a multi-silicide coating on the surface of WMoNbCrTi HEA, forming a continuous and dense composite oxide film with a thickness of about 1 μm. The silicon infiltrated alloy only gained 0.44 mg/cm² in weight for 100 h of oxidation at 1200 °C, effectively improving the resistance to high temperature oxidation of the refractory high-entropy alloy.

In addition, low temperature nitrogen carburizing can supersaturate

the gap of solid solution, harden the surface of alloy and improve the wear resistance of alloy. Lindner et al. [72] used low temperature nitrogen carburizing method to harden the surface of CrMnFeCoNi HEA and CrFeCoNi HEA, and found that in the state of nitrogen carburizing, the austenite phase formed by the interstitial solution of nitrogen and carbon can significantly improve the microhardness and wear resistance of the alloy. Therefore the hardness and wear resistance of FCC high entropy alloy can be improved by low temperature nitrogen carburizing.

3.2.5. Metal infiltration treatment

In addition to the thermochemical processes of infiltrating non-metallic elements such as nitriding, carburizing and boriding, there are also thermochemical processes of infiltrating other metals such as aluminizing and chromizing that can improve the surface hardness and wear resistance of high-entropy alloys.

Sun et al. [46,73] carried out solid aluminizing treatment on AlCo-FeCrNi HEA. The results showed that the hardness of the aluminide infiltration layer was much higher than that of the substrate, and there was a reduction in the wear of the infiltrated aluminum alloy under small loads compared to the original alloy, meaning the aluminizing treatment could improve the wear resistance of the alloy under certain conditions. A number of studies have shown [74–78] that the addition of aluminum or aluminizing treatment to high entropy alloys can also enhance the high temperature oxidation resistance of high entropy alloys.

Qiao et al. [79] treated Al0.3CoCrFeNi HEA using the double glow chromium infiltration technique and found that the overall hardness of the material increased from 181 HV to 300 HV, the yield strength increased from 51.20 MPa to 93.85 MPa and the tensile strength also increased after metal infiltration. The increase in hardness made the surface less susceptible to plastic deformation, adhesion and adhesive

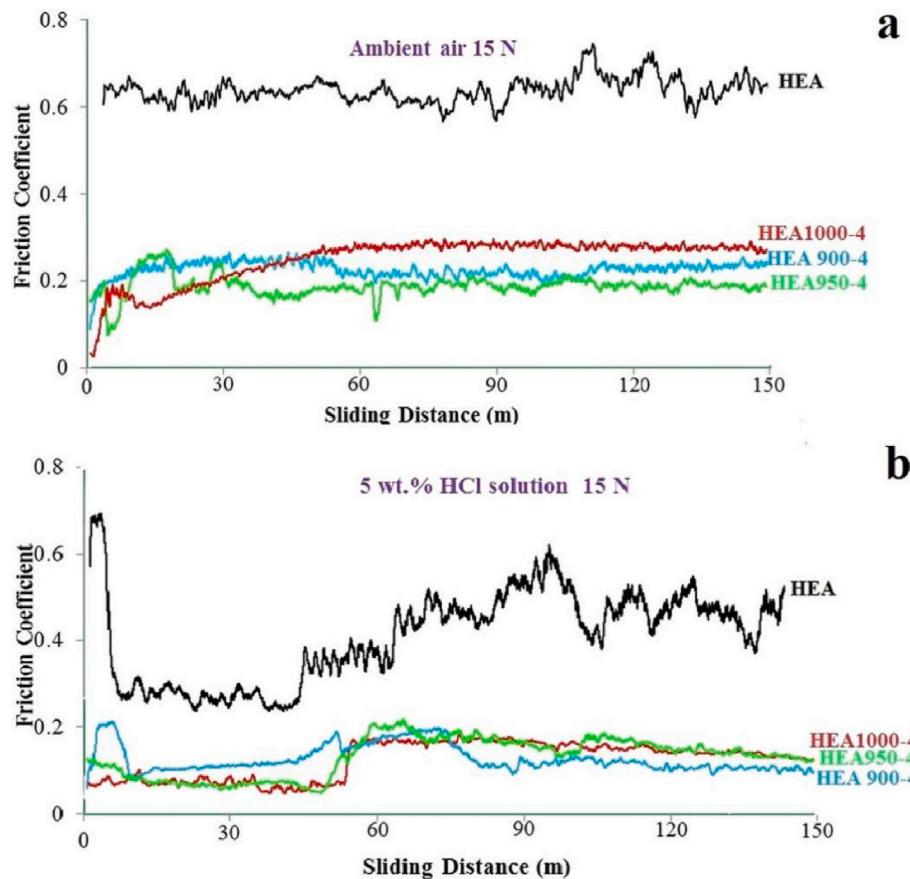


Fig. 10. Friction coefficients of untreated and boriding in different environments: (a) air, (b) HCl solution.

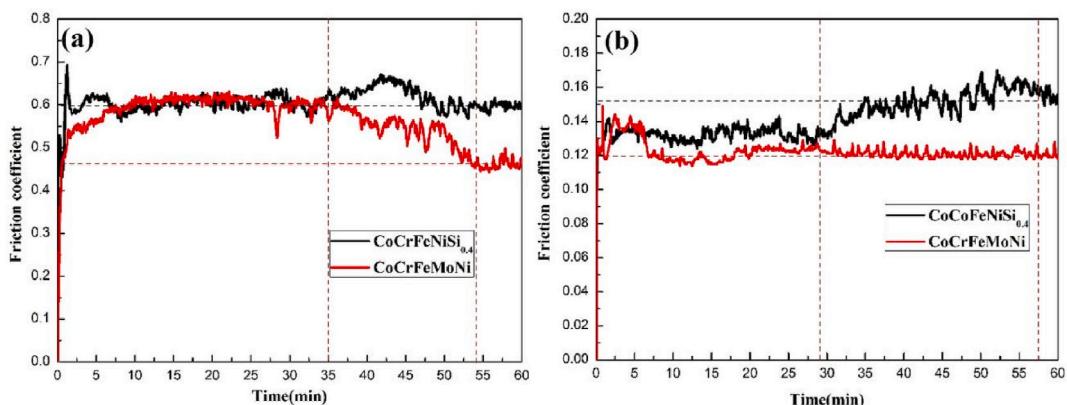


Fig. 11. Friction coefficient-time curves of CoCrFeNbSi_{0.4} HEA and CoCrFeMoNi HEA (a) before and (b) after sulfurization.

detachment, and the average friction coefficient decreased from 0.436 to 0.279, improving the wear resistance of the material surface. Han et al. [80] prepared WTaTiVCr HEA layer on pure tungsten by the double glow plasma infiltration metal technique. The results showed that the surface hardness of HEA layer can reach 1300 HV due to the strengthening mechanism of solid solution strengthening and lattice distortion. The wear mechanism was abrasive wear, the average friction coefficient was 0.447, and the wear rate was $3.43 \times 10^{-8} \text{ mm}^3/(\text{N}\cdot\text{mm})$.

3.3. Surface heat treatment and heat treatment strengthening

Surface heat treatment can produce a surface layer with a high hardness and a good internal stress distribution, increase the workpiece

wear resistance and fatigue resistance, and keep the surface layer's chemical makeup unaltered. Heat treatment surface modification technology may strengthen the surface performance of parts, increase the service performance of materials and components, and help materials and essential components achieve their maximum performance [48,81].

Han et al. [82] treated AlCoCrFeNi–Cu composite coating with induction heating. After induction heating, the porosity of the coating was reduced by 41.29 %, resulting in more uniform microstructure distribution and microhardness up to 510 HV. The wear mode of the composite coating gradually changed from abrasive wear to fatigue wear, and the wear amount was 0.69 mg, which showed good friction and wear performance. Lin et al. [83] placed FeCoCrNiAl HEA coating at different temperatures for vacuum heat treatment. After vacuum heat

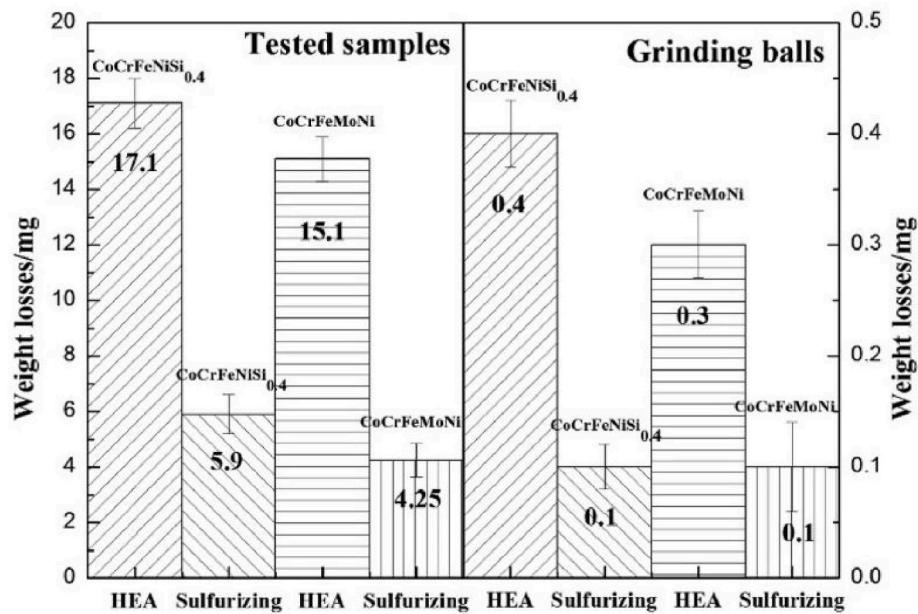


Fig. 12. Wear weightlessness of high entropy alloy and corresponding grinding balls before and after vulcanization.

treatment, the degree of alloying, hardness and wear quality of the coating were increased. The wear mechanism of the original coating was adhesive wear, while the wear mechanism after vacuum heat treatment was adhesive wear and abrasive wear.

Zhang et al. [84] used high temperature furnace heating method to heat treat CoCrFeNiSi_{2.0} HEA coating at different temperatures. The results showed that the high entropy alloy with high temperature heat treatment showed excellent comprehensiveness, and the highest microhardness reached 1347.4 HV due to the dispersion strengthening of Cr₂Si and Ni₄Si. When the heat treatment temperature was 800 °C, the friction coefficient of the alloy was 0.19, the wear amount is 9.9 mg, and the wear mechanism was abrasive wear and adhesive wear. Laser remelting technology can not only improve the surface hardness of high entropy alloy, but also improve the surface high entropy and densification of sintered materials, so as to jointly improve the surface wear resistance [85].

Lin et al. [83] performed laser remelting of FeCoCrNiAl HEA coating. The results showed that after laser remelting, the elements were uniformly distributed, the entropy is significantly increased, and the

average hardness of the coating was 682 HV. Because the high entropy effect limited the formation of intermetallic compounds and reduces the possibility of abrasive wear, the wear mode of coating was mainly adhesive wear. Miao et al. [86] used laser remelting and aging heat treatment to perform surface modification on AlCoCrFeNi_{2.1} eutectic high entropy alloy (EHEA). The intensification process was shown in Fig. 13. After laser remelting, the alloy formed FCC and BCC phase supersaturated solid solution structure. The nano-precipitates were reintroduced into FCC and BCC phases after further aging treatment. Due to the decrease of lamellar spacing between FCC and BCC and the coherent precipitation of nanoparticles, the surface microhardness of EHEA increased by 59 %, and the average friction coefficient and wear rate decreased by 26 % and 68 %, respectively. The EHEA wear process was mainly determined by abrasive wear.

Qi et al. [87] modified the surface of NiCoCrFeAlTi EHEA by laser remelting and aging treatment. The grain was obviously refined after laser remelting. After aging treatment, γ' nanoprecipitation was precipitated. The average surface microhardness of Ni₂₉C_o₃₀Cr₁₀Fe₁₀Al₁₈Ti₃ alloy was the highest, which was about 620 HV.

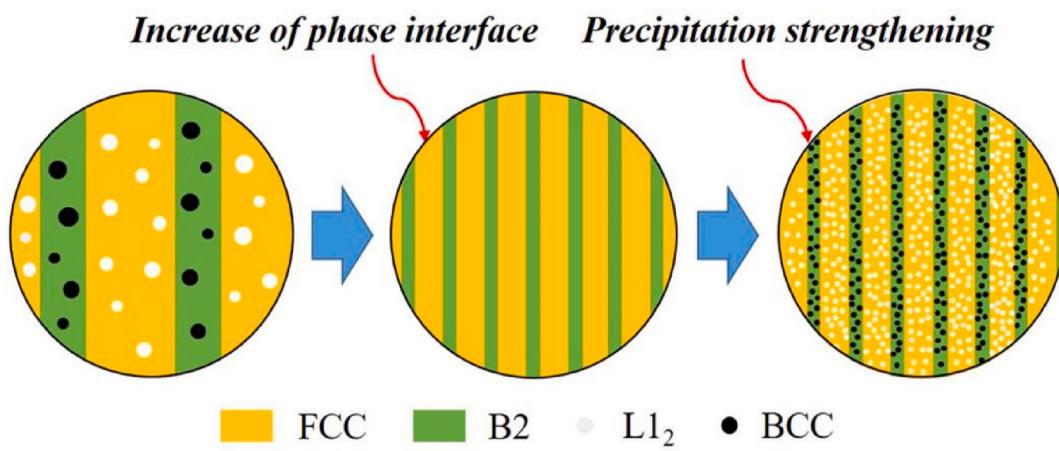


Fig. 13. Schematic diagram of AlCoCrFeNi_{2.1} EHEA surface modification process.

Zhang et al. [88] prepared CoCrFeMnNi HEA with reverse gradient grain structure through cold rolling process and laser surface heat treatment technology. It had been shown that the strength and plasticity of the alloy can be improved by hetero-deformation strengthening and strain hardening caused by the inverse gradient grain structure, seeing section 3.1 for details. Laser surface heat treatment technology can significantly improve the strength and plasticity of metal materials without thickness limitation. The key to produce graded structural materials was to enlarge the microhardness difference and reduce the thermal stress.

As a heat treatment technology, annealing can refine grains, eliminate internal stresses and defects, and improve microstructure. Sha et al. [89] annealed AlCoCrFeNiTi_{0.5} HEA coating to form a solid solution with a bcc structure and a typical fine junction decomposition structure. Due to solute strengthening and precipitation strengthening, the average Vickers hardness of annealed samples was increased by 73.5 %, wear width was reduced by 50 %, and mass loss was reduced by 92.5 % compared with as-cast samples. Wu et al. [67] annealed Al_{0.1}CoCrFeNi HEA. The main wear mechanisms of the annealed alloy in air were adhesive wear and plastic ploughing; in water the wear mechanisms were mainly abrasive and delamination wear with slight adhesive wear. The wear rates of annealed alloys in dry water and deionized water were approximately $1.86 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$ and $1.37 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$ respectively.

Chen et al. [90] annealed Al_{0.6}CoCrFeNi HEA at 850 °C for 4 h. The results showed that after heat treatment, σ-CrFe hard phase was precipitated in the alloy, and the hardness increased from 278 HV to 480 HV. The wear resistance of annealed alloy at 600 °C was nearly 3 times that of GCr15 steel, and the wear mechanism changed from abrasive wear to mild oxidation wear and layered wear. Ren et al. [91] CoCrNiCu HEA was thermally annealed. It was found that a large number of Cu-rich second phase precipitates in the matrix, and the nano-precipitates changed into lamellar structures during the sliding

process, as showed in Fig. 14. Due to the formation of the self-organized non-uniform layer structure and the oxide friction layer on the surface, the wear rate of the annealed alloy was reduced by 48.2 % and 57.4 % compared with that of the as-cast alloy under the dry sliding wear tests of 5 N and 10 N, respectively. The wear mechanism of as-cast alloy was abrasive wear and adhesive wear, while the wear mechanism of annealed alloy was abrasive wear and oxidation wear. Tian et al. [92] studied the effect of annealing temperature on the performance of FeCoCrNiMo_{0.1} HEA. With the increase of annealing temperature, the deformation twins changed to annealing twins, and the wear mechanism changed from plastic deformation, adhesive wear and abrasive wear to plastic deformation and adhesive wear. The yield strength, wear resistance and friction coefficient of the alloy were increased.

In conclusion, the friction and wear properties of high entropy alloy can be improved by adjusting the properties of high entropy alloy through heat treatment. At the same time, in order to prevent the adverse change of wear mechanism, during the heat treatment process, we should avoid: the uneven distribution of hardness caused by the enrichment phase segregation [93], the reduction of dislocation density after recrystallization [94], the homogenization tendency of alloying elements [95], and the Ostwald curing phenomenon of precipitates [96]. Liu et al. [96] found that the Ostwald curing behavior of AlNi precipitated phase existed in AlCoCrFeNiTi_{0.8} HEA coating. As shown in Fig. 15, the precipitation of AlNi with relatively small size disappeared and the remaining AlNi phase coarsened, resulting in the wear resistance of the coating decreased.

3.4. Surface laser cladding treatment

Laser cladding technology uses a laser beam of high energy degree to fuse the clad material with a thin layer of the substrate surface to form a layer of alloy with better properties on the surface of the substrate [97].

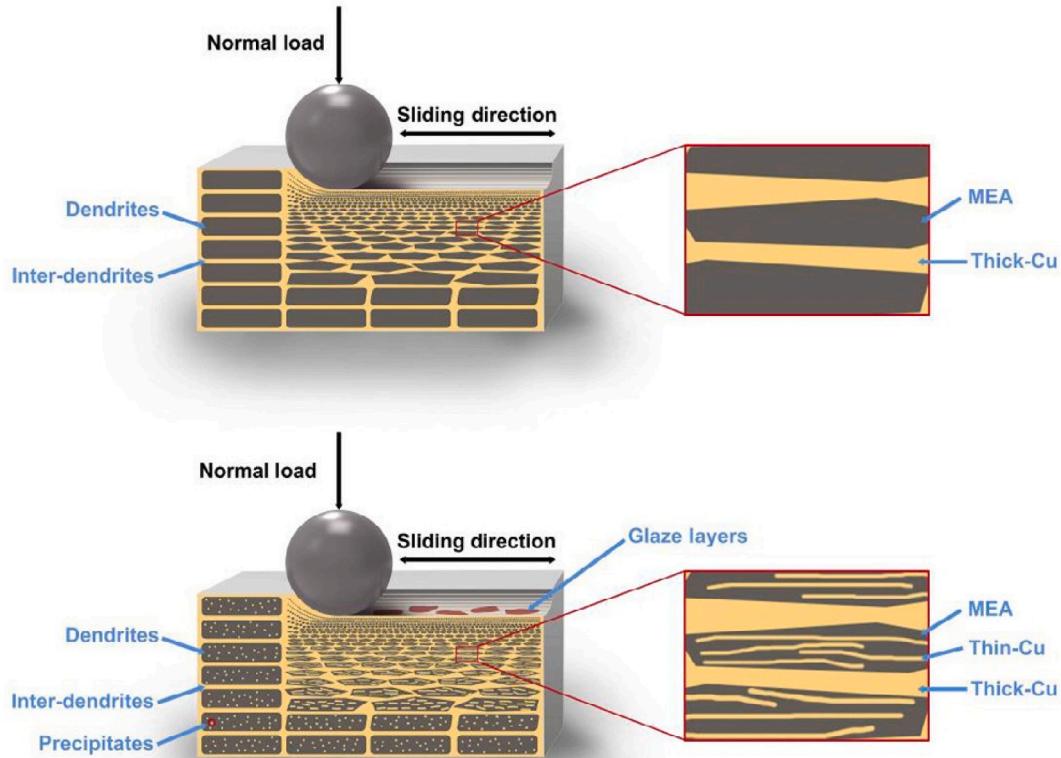


Fig. 14. Mechanisms for the enhanced wear-resistance in the metastable CoCrNiCu HEA.

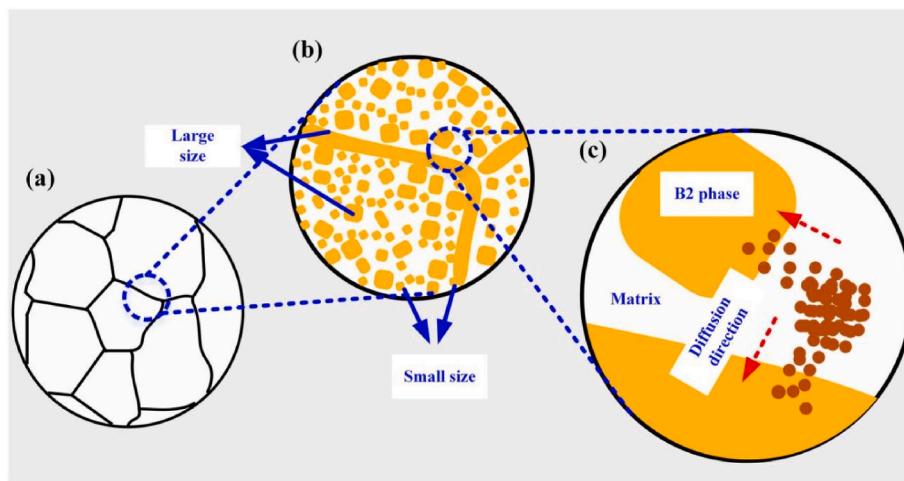


Fig. 15. Schematic diagram of Ostwald curing of AlNi precipitate phase in the AlCoCrFeNiTi_{0.8} HEA coating.

Laser melting is a common process for coating preparation and surface modification to ensure the excellent properties of the alloy material and also to achieve a good metallurgical bond between the coating and the substrate for the purpose of enhancing the surface wear resistance [98–100]. The typical strengthening phases that commonly affect the frictional wear properties of the HEA after laser melting treatment are shown in Table 3 and Table 4.

Huang et al. [110] and Cai et al. [111] respectively prepared the TiV80CrAlSi and NiCrCoTiVAL laser cladding coatings on the surface of the Ti6Al4V titanium alloy substrate. The results showed that the silicide phase or titanium phase produced in the laser cladding layer improved the wear resistance of the substrate. Liang et al. [112] prepared the AlCrFeNi₂W_{0.2}Nb_{2.0} HEA coating on 304 stainless steel plates by laser cladding technology. The microhardness of the AlCrFeNi₂W_{0.2}Nb_{2.0} HEA coating was about 4.5 times that of the substrate. Compared with 304 stainless steel, the average friction coefficient values of Nb_{0.5}, Nb_{1.0}, Nb_{1.5} and Nb_{2.0} were reduced by 94 %, 83 %, 59 % and 75 %. As shown in Fig. 16, due to the anti-wear effect of the oxide film, the coating of the AlCrFeNi₂W_{0.2}Nb_x ($x = 1.5, 2.0$) HEA under the same loading condition had better wear resistance than that of 304 stainless steel.

Shu et al. [113] used laser cladding technology to prepare an amorphous coating of the FeCrCoNiSiB HEA on Q235 steel. The upper layer of the coating was an amorphous/crystalline layer, and the dendritic layer was the bottom layer. Because the amorphous phase of the upper layer can promote the formation of a new wear mechanism and its high hardness, the wear rate of the upper layer was 25.9 % lower than that of the bottom layer, and the upper layer had better high-temperature wear resistance. Jia et al. [114] prepared a coating of the CoCrNbNiW HEA on the surface of 45 steel by laser cladding technology. The existence of Nb and Cr-rich FCC phase and dispersed un-melted W particles in the coating made the overall wear resistance of the CoCrNbNiW HEA higher than that of the substrate. The average microhardness of the substrate was about 0.36 times that of the coating, and the wear amount and wear rate was 0.26 times and 0.23 times that of the substrate, respectively. Peng et al. [115] prepared the FeCoCrNi-WC (spherical and irregular) HEA composite coating by plasma cladding technology. The results showed that both the FeCoCrNi-WC (spherical and irregular) HEA coatings showed well wear resistance. From Fig. 17(a and b), the spherical WC particles and a mass of M6C carbides were the chief anti-wear skeleton and provided

Table 3

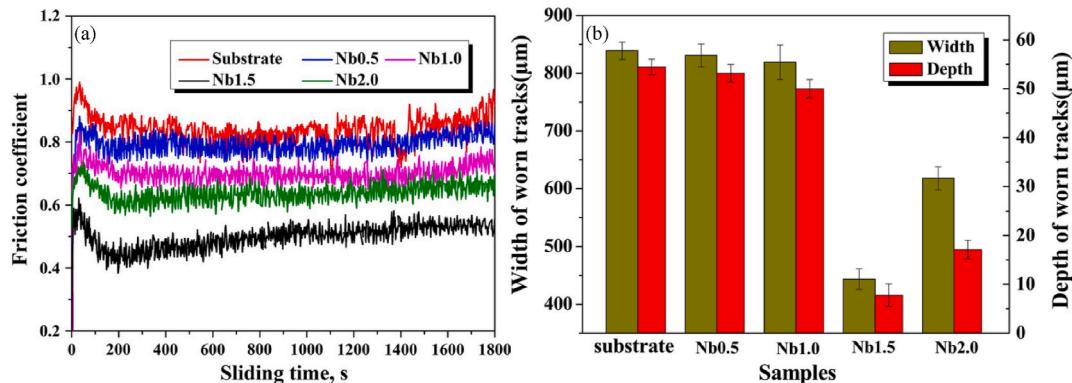
Typical strengthening phases affecting the friction and wear properties of the HEA after laser cladding treatment on the surface.

Coating	Matrix	Strengthening phase	M/C-Hardness (HV)	M/C-Friction coefficients	M/C-wear properties	
					M/C-volume wear (mm ³)	M/C-volume wear (mg)
AlCoCrFeNi [101]	45 steel	WC	<200/500	0.9/0.8		3.18/2.19
AlCoCrFeNi + WC [101]	45 steel	WC	<200/600	0.9/0.6		3.18/1.35
MoFe _{1.5} CrTiWAlNb _{1.5} [102]	M2 tool steel	MC, C14 Laves	320/810	0.87/0.72	0.4314/>0.4314	
MoFe _{1.5} CrTiWAlNb ₂ [102]	M2 tool steel	MC, C14 Laves	320/851	0.87/0.68	0.4314/>0.4314	
MoFe _{1.5} CrTiWAlNb _{2.5} [102]	M2 tool steel	MC, C14 Laves	320/873	0.87/0.58	0.4314/>0.4314	
MoFe _{1.5} CrTiWAlNb ₃ [102]	M2 tool steel	MC, C14 Laves	320/910	0.87/0.52	0.4314/0.045	
MoFeCrTiWAlNb [103]	M2 tool steel	(Nb, Ti) C, Fe ₂ Nb-Laves	330/820	0.78/0.5–0.6	0.354/0.16	
AlCoCrFeMoVTi [104]	Ti6Al4V	(Co, Cr, Fe) phase, Al ₂ O ₃	340/1120	0.52/0.31	1.4.91 × 10 ⁻³ /0.179 × 10 ⁻³	
FeMoCoNiCrTi _{0.25} [105]	T10A steel	NiCrFe, NiCrCoMo, TiFe		0.4/0.26		
FeMoCoNiCrTi _{0.5} [105]	T10A steel	NiCrFe, NiCrCoMo, TiFe		0.4/0.25		
FeMoCoNiCrTi _{0.75} [105]	T10A steel	NiCrFe, NiCrCoMo, TiFe		0.4/0.23		
FeMoCoNiCrTi ₁ [105]	T10A steel	NiCrFe, NiCrCoMo, TiFe		0.4/0.20		

Table 4

Main strengthening phases and tribological characteristics of the HEA coating with different elements.

Coating	Matrix	Strengthening phase	Hardness (HV)	Friction coefficients	Wear properties	
					Volume wear (mm ³)	Wear rate (mm ³ /(N·m))
CoCrFeMnNi [106]	45 steel	M23C6	183.2	0.44	0.54	
CoCrFeMnNiCo _{0.03} [106]	45 steel	M23C6	>183.2	0.47	0.40	
CoCrFeMnNiCo _{0.06} [106]	45 steel	M23C6	>183.2	0.36	0.14	
CoCrFeMnNiCo _{0.09} [106]	45 steel	M23C6	223.48	0.22	0.46	
CoCrFeMnNiCo _{0.12} [106]	45 steel/98	M23C6	<223.48	0.40	0.21	
CoCrFeMnNiCo _{0.15} [106]	45 steel	M23C6	195.04	0.13	0.30	
AlCoCrFeNi [107]	304 stainless steel	TiC	594.7			0.38 × 10 ⁻⁴
AlCoCrFeNi+2%TiC [107]	304 stainless steel	TiC	683.2			0.22 × 10 ⁻⁴
CoCrFeMnNi [108]	45 steel	TiC	183.2	0.44	0.54	
CoCrFeMnNiTi _{0.25} [108]	45 steel	TiC	>183.2	0.47	0.40	
CoCrFeMnNiTi _{0.50} [108]	45 steel	TiC	>183.2	0.36	0.14	
CoCrFeMnNiTi _{0.75} [108]	45 steel	TiC	223.48	0.22	0.46	
CoCrFeMnNiTi _{1.00} [108]	45 steel	TiC	<223.48	0.40	0.21	
CoCrFeMnNiCo _{0.15} [108]	45 steel	TiC	195.04	0.13	0.30	
CoCrNiMnMn [109]	Q235 steel	(Co, Fe) ₂ Ti-Laves	165.33	0.6997		
CoCrNiMnTi _{0.25} [109]	Q235 steel	(Co, Fe) ₂ Ti-Laves	>165.33	0.6447		
CoCrNiMnTi _{0.50} [109]	Q235 steel	(Co, Fe) ₂ Ti-Laves	>165.33	0.6222		
CoCrNiMnTi _{0.75} [109]	Q235 steel	(Co, Fe) ₂ Ti-Laves	>165.33	0.5923		
CoCrNiMnTi _{1.00} [109]	Q235 steel	(Co, Fe) ₂ Ti-Laves	523.73	0.6268		

**Fig. 16.** Friction coefficient (a) and the depth and width of wear marks (b) of the AlCrFeNi₂W_{0.2}Nb_{2.0} HEA coating and 304 stainless steel.

protection to the HEA substrate, which improved the wear resistance of the coating. In the irregular WC coating, the small, uniformly dispersed M6C carbide abrasive grains resisted the ploughing effect of broken debris and prevented the formation of three-body abrasive wear, whereas in the HEA matrix, the well-developed M6C fishbone carbide phase acted as a tiny abrasive, as shown in Fig. 17(c and d). Cai et al. [116] combined laser cladding and laser remelting technology to prepare the NiCrCoTiV HEA coating on the surface of Ti-6Al-4V alloy. The effect of solid solution strengthening and grain boundary strengthening resulted in a HEA coating with a much higher microhardness than the Ti6Al4V matrix and exhibited better wear properties.

3.5. Surface plasma treatment

Plasma is an aggregation state with high material energy, and plasma technology modification is a plasma application of heterogeneous gas-solid reaction. Plasma technology is used to process the surface of metal materials to form coating or diffusion, so as to improve the surface properties of materials [117–119].

Zhang et al. [120] prepared the AlCoCrFeNi HEA using spark plasma sintering technology. The results showed that the wear rate reached the highest at 200 °C, and decreased with the increasing of the test temperature, reaching the lowest at 800 °C, with the wear rate lower than 2×10^{-5} mm³/(N·m), showing good wear resistance. Ji et al. [121] prepared the Al_xCrFeCoNiCu HEA coating on carbon steel substrate by spark plasma sintering technology. It was found that among all the

components, the Al₃CrFeCoNiCu HEA had the lowest mass loss and the Al₂CrFeCoNiCu HEA had the lowest sliding friction coefficient. Yadav et al. [122] prepared two kinds of (CuCrFeTiZn)_{100-x}Pb_x and (AlCr-FeMnV)_{100-x}Bi_x HEAs in argon atmosphere by spark plasma sintering technology. The experiment explored that the increase or decrease of Pb content had no obvious relationship with the friction coefficient, while the addition of Bi made the friction coefficient decrease greatly, but with the increase of Pb and Bi, the wear rate decreased by 21 % and 25 %. Lobel et al. [123] prepared the AlCoCrFeNiTi HEA by spark plasma sintering and added different contents of solid lubricant MoS₂. The results indicated that the addition of MoS₂ at room temperature effectively reduced the depth of those wear marks. Compared with the matrix, with the addition of MoS₂, the sliding friction coefficient increased first and then decreased, which improved the wear resistance to some extent.

Plasma spraying is considered an economical and efficient surface engineering technology, which can deposit any materials on substrates and improve the life of solid materials under harsh working conditions [124]. Tian et al. [125] prepared the AlCoCrFeNiTi/Ni60 coating on 316 stainless steel by plasma spraying. The wear resistance of the coating was studied by a cutting disc friction tester. It is found that Ni60 as a reinforcing agent can improve the wear resistance of the coating. Liu et al. [126] used plasma spraying technology to prepare the FeCoCrAlNi HEA wear-resistant coating on the surface of 304 stainless steel. The average microhardness of the coating was 411 HV, almost 1.5 times that of the substrate. Compared to the matrix, the coating showed significantly lower wear rates of 54 % and 84 % at room temperature and high

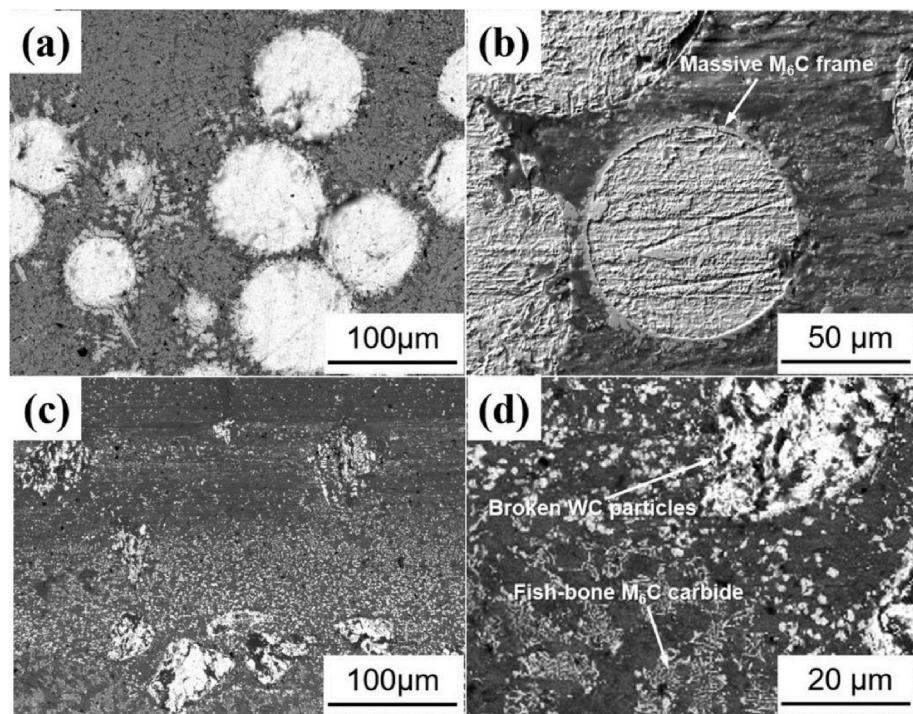


Fig. 17. WC coating wear surface: (a) and (b) spherical, (c) and (d) irregular WC coating.

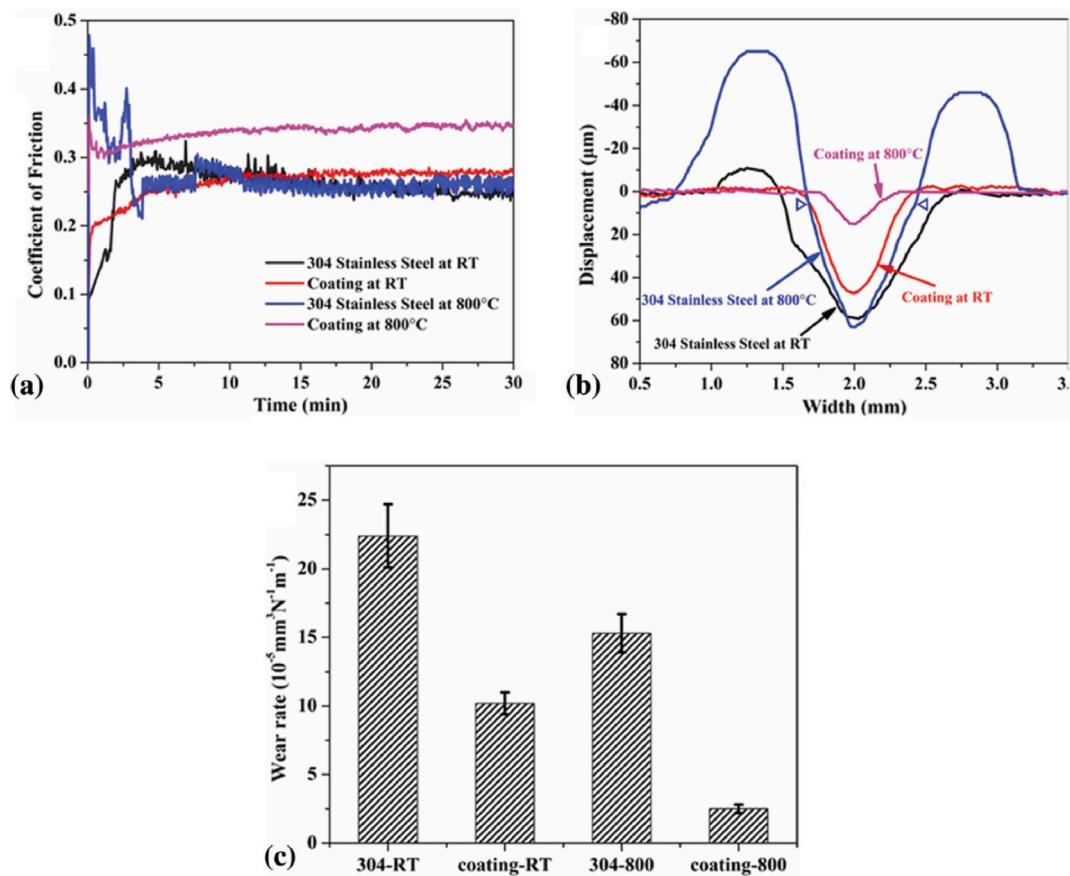


Fig. 18. Friction coefficient (a), surface profile (b) and wear rate (c) of the 304 stainless steel substrate and the FeCoCrAlNi HEA coating at room temperature and 800 °C.

temperature. The HEA coating showed better wear resistance at room temperature and 800 °C, as shown in Fig. 18. The wear mechanism of the FeCoCrAlNi HEA coating was oxidative wear and light abrasive wear at room temperature, with increased adhesive wear and oxide film formation at high temperatures.

Jin et al. [127] used plasma spraying and laser remelting technology to prepare the FeCoCrNiAl_{0.5}Si_x HEA coating on Q235 steel surface. Based on the results of sliding wear tests, it was found that as the Si content increased, the coefficient of friction, wear depth and mass loss decreased, thus improving the wear resistance of the coating. Zhang et al. [128] prepared the AlCoCrFeNiV_x HEA coating on Q235 steel substrate by plasma spraying and laser remelting, and tested the hardness and wear resistance of the coating. The results showed that the surface of the coating with low V would form oxide such as Fe₂O₃, which would adhere to the worn surface to form an oxide film, which could isolate the contact between tungsten nitride pellets and the sample, thus playing a protective role. As the increase of V, the hardness became higher and higher, and many fine and dispersed acicular CrV phases appeared in the alloy structure. The joint action of these two factors improved the wear resistance of the alloy, which made the wear surface of the alloy flatter and the furrow shallower.

3.6. Surface electron beam treatment

The surface electron beam makes use of the heat source of the high-energy electron beam to make the surface temperature of materials rise rapidly, and the surface composition and microstructure change, thus improving the surface properties of materials. After electron beam surface modification, the hardness and wear resistance of the material can be improved, and it has the advantages and technical characteristics of small deformation, strong controllability, cleanliness, no pollution and a high energy utilization rate [129,130].

Koval et al. [131] prepared the (Zn–Cu–Al+12Cr18Ni10Ti) coating on 45 steel by vacuum ion plasma method, and then obtained the

coating by pulsed electron beam irradiation. The research results showed that during electron beam treatment and cooling of multi-element coating, the stress state decreases, and the microhardness of coating material increases due to dislocation hardening. Moreover, due to the reduction of defects and residual stress caused by irradiation annealing, the roughness was reduced and the structural state was changed, the wear resistance of the multi-component coating was improved, and its friction coefficient was significantly reduced. Peng et al. [132] prepared the CoCrFeNiMo_{0.2} HEA by vacuum arc melting method, and treated its surface with a high current pulsed electron beam. After high current pulsed electron beam irradiation, the surface microhardness and wear resistance of the cast CrFeCoNiMo_{0.2} were improved by diffuse strengthening of the hard reinforced σ-phase and the fine grain strengthening of FCC superfine grains. After 35 pulse irradiations, the alloy attained the maximum microhardness (392.9 HV) and the lowest wear rate ($0.92 \times 10^{-4} \text{ mm}^3/(\text{N}\cdot\text{m})$), which the wear mechanism was abrasive wear accompanied by oxidative wear.

Yu et al. [133] used electron beam melting technology to prepare the AlCrTiNbMo HEA coatings to improve the low surface hardness and wear resistance of Ti600. Because of the solid solution strengthening effect as well as the diffusion-enhanced second phase strengthening which enhanced the hardness of the coating, the microhardness of the coating was approximately 2.2 times that of the matrix and exhibited better wear resistance. The wear surface of the coating contained only shear surface marks and the wear mechanism was micro-cutting; whereas the surface of Ti600 had severe fracture and delamination and was a mixture of microchipping and micro-cutting wear. Shu et al. [134] prepared the CoCrCuFeNiV_x HEA by powder metallurgy and electron beam remelting. It was found that with the increase of V, the FCC solid solution phase became more stable, and the hardness of the CoCrCuFeNiV_x HEA increased from 301.5 HV to 408.4 HV. The wear resistance was dramatically increased and the wear rate was decreased by 80.6 %, because of the hardened layer and oxide film formed on the surface. The wear mechanism of the CoCrCuFeNiV_x HEA was abrasive

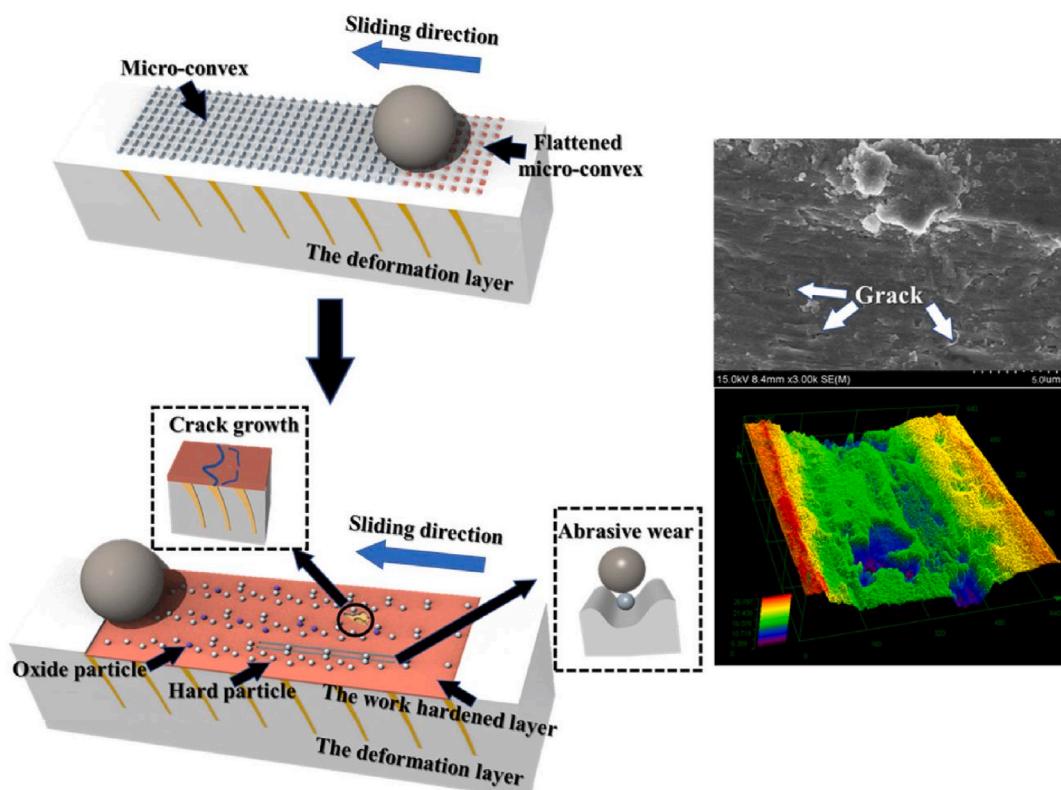


Fig. 19. Wear mechanism of the CoCrCuFeNiV_x HEA under 131 mA current.

wear, adhesive wear and oxidation wear, and its wear mechanism was shown in Fig. 19.

3.7. Vapor deposition technology

Vapor deposition is the technology of condensing the vapor of the material to be deposited on the base material in a vacuum to produce micron-sized films, mainly including physical vapor deposition and chemical vapor deposition, and physical-chemical vapor deposition developed on this basis, among which physical vapor deposition technology mainly includes evaporation coating, sputtering coating and ion coating [135,136].

Chai et al. [137] prepared Ti(C, N)-based cermet with the CoCr-FeMnNi HEA bonding phase by chemical vapor deposition, and found that the CoCrFeMnNi HEA bonding phase can refine the grains of Ti(C, N)-based cermet. Because of the small grain size, high hardness and strong adhesion of the bonded cermet, the coating had good wear resistance, and the wear forms were abrasive wear, adhesive wear and oxidation wear. Chang et al. [138] synthesized the CrVTiNbZr HEA and the nitrogen-containing CrVTiNbZr(N) coating by high ionization cathode arc deposition method. Compared with the HEA, the high H^3/E^2 value and multi-layer structure improved the energy dissipation related to frictional contact, so the wear rate and average friction coefficient of the CrVTiNbZr(N) were significantly reduced and the wear resistance was enhanced. At the same time, owing to the formation of a multi-layer metal nitride structure and the solid solution strengthening of multi-component elements, the coating achieved the lowest wear rate and showed high wear resistance. Chen et al. [139] successfully prepared the FeCrNiCuAlSn_{0.5} HEA coating on the surface of 45 steel by thermit reaction/spray deposition technology. The coating had higher hardness and better bearing capacity compared with the substrate. Meanwhile, the added Sn played a good role in reducing wear and lubrication, making the friction coefficient at a relatively stable level, with a wear rate of $(7.24 \pm 0.52) \times 10^{-6} \text{ mm}^3/(\text{N}\cdot\text{m})$. But the friction coefficient of the 45 steel is 0.75, and the wear rate was $(1.45 \pm 0.35) \times$

$10^{-6} \text{ mm}^3/(\text{N}\cdot\text{m})$. The coating showed better wear resistance than the matrix.

Lee et al. [140] deposited the $(\text{CrNbSiTiZr})\text{C}_x\text{N}_y$ HEA carbon-nitride films by radio frequency magnetron sputtering under different CH_4 and N_2 fluxes. It was found that the uncoated WC-Co substrate had a friction coefficient of 0.77, while most carbide and carbon-nitride films was between 0.13 and 0.23. The $(\text{CrNbSiTiZr})\text{C}_x\text{N}_y$ film with 71 % carbon content had the lowest friction coefficient (0.13) and wear rate ($0.4 \times 10^{-7} \text{ mm}^3/(\text{N}\cdot\text{m})$); the $(\text{CrNbSiTiZr})\text{C}_x\text{N}_y$ film with 48.2 % nitrogen content had the highest hardness value, oxidation resistance, coefficient of friction and wear rate. The films contained carbon-carbon bonds in the amorphous graphite region and their amorphous carbon phase exerted a good lubricating effect, resulting in relatively low wear rates and friction coefficients. Li et al. [141] prepared the $\text{TaW}_{25}\text{Ti}_{16}\text{V}_{18}\text{Cr}_{17}$ HEA film by double-target co-sputtering deposition method, in which the $\text{Ta}_{24}\text{W}_{25}\text{Ti}_{16}\text{V}_{18}\text{Cr}_{17}$ film showed the best mechanical properties with hardness and modulus of 27.61 GPa and 274.42 GPa, respectively. At the same time, the average friction coefficient and wear rate of the $\text{Ta}_{24}\text{W}_{25}\text{Ti}_{16}\text{V}_{18}\text{Cr}_{17}$ film were 0.34 and $5.01 \times 10^{-9} \text{ mm}^3/(\text{N}\cdot\text{mm})$, respectively, due to the high hardness and modulus of the film itself, as well as the surface morphology and regular pit-like structure, showing excellent film-based adhesion and wear resistance. Sha et al. [142] deposited the $(\text{FeMnNiCoCr})\text{N}_x$ coating on an M2 steel substrate by DC magnetron sputtering system. It was found that as the nitrogen content increased, substrate exposure decreased, oxide formation was reduced and the coatings showed an excellent combination of excellent hardness, wear resistance and fairly good scratch response, as shown in Fig. 20. Compared with the nitrogen flow rate of 4 sccm, the coating at a nitrogen flow rate of 25 sccm showed a relatively smooth and clean wear surface with little evidence of any substrate exposure or black oxide. Liu et al. [143] used magnetron sputtering technology to prepare the AlV-TiCrW HEA film on superalloy substrate, and discovered the unreported high temperature solid wetting phase AlV_3O_9 . The crystal surface with the preferred orientation formed can make the film show a low friction coefficient (0.15) and a low wear rate at 800 °C. Chen et al. [144]

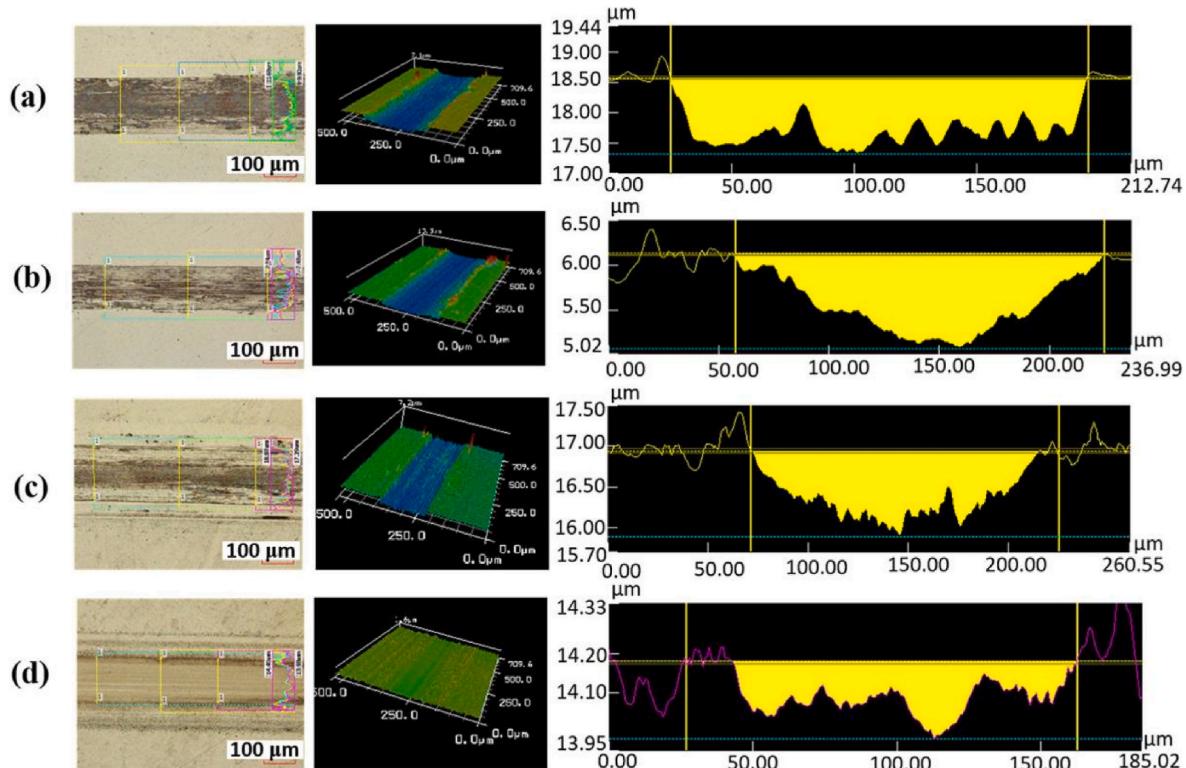


Fig. 20. 2D and 3D laser optical images of wear tracks and corresponding profiles at different nitrogen flow rates: (a) 4 sccm; (b) 8 sccm; (c) 15 sccm; (d) 25 sccm.

prepared the TiNbWN films on 304 stainless steel substrates by magnetron sputtering. The nanocrystalline tungsten and titanium oxides (W_nO_{3n-2} , TiO_2 , and γ - TiO_x) formed in the films at 750 °C can act as solid lubricants to reduce interfacial adhesion between the films and sliders, while enhancing the hardness of the films and helping to reduce wear losses.

3.8. Ion implantation technology

Ion implantation is an important means of material modification. It changes the tribological characteristics of the substrate by high-energy ion beam bombardment and implantation into the surface of the substrate at high speed, to obtain a highly supersaturated solid solution, metastable phase and amorphous structure. The hard phase formed between the implanted ions and the matrix is uniformly dispersed on the metal surface, which can strengthen the hardness and wear resistance of the metal surface and improve the metal properties such as oxidation resistance, corrosion resistance and fatigue resistance [145–147].

Pogrebniak et al. [148] prepared nitride coating of the $(TiHfZrNbVTa)_N$ HEA by cathodic arc vapor deposition. The hardness of the coating was increased by negative gold ion implantation, and the wear resistance of the $(TiHfZrNbVTa)_N$ coating was improved. The friction coefficient decreased from 0.75 to 0.243 in the first 320s, and then increased to 0.78 in the next 1000s. Pogrebniak et al. [149] used vacuum arc evaporation technology to study the $(TiHfZrNbVTa)_N$ coating before and after ion implantation under different deposition conditions. After ion implantation, a multi-phase structure composed of amorphous, nanocrystalline and initial nanostructure phases was formed in the surface layer of the coating, the surface roughness was reduced, and the coating had good wear resistance. Jenczyk et al. [150] used ion implantation and nanoindentation to study the $AlCoCrFeNiTi_{0.2}$ HEA. The results showed that ion implantation would lead to phase transformation and hardening of the surface layer, and induce new high entropy or medium entropy ceramics. The wear resistance and the hardness of the $AlCoCrFeNiTi_{0.2}$ HEA increased with the increase of ion implantation, and the wear resistance of ion-implanted samples was better than that of non-implanted samples. As shown in Fig. 21, the samples with the highest ion implantation had the shallowest scratches and the highest wear resistance, while the non-implanted samples had the deepest scratches.

4. Conclusion

The unique and rich structure of the high entropy alloy allows for superior tribological properties. Surface treatment is essential for the wear resistance of well performance metallic materials, and its effect on the organization and tribological properties of high entropy alloys is

mainly manifested as follows.

- i) dislocation strengthening and fine grain strengthening by thermomechanical treatment;
- ii) solid solution strengthening and second phase strengthening of interstitial solid solution and second phase precipitation;
- iii) forming a surface cladding with high hardness/strength on the surface;
- iv) formation of surface micro-heterogeneous surface structure and other strengthening behaviors.

Currently, the technology most frequently utilized for surface strengthening is laser cladding. The preparation of HEA coating using laser cladding technology results in a good metallurgical bond between the coating and substrate and serves the goal of increasing surface wear resistance. However, we also encounter many problems when studying the friction and wear properties of high entropy alloys: Firstly, due to the rich design range of components and the unique characteristics of high entropy alloys, the research on wear mechanism in friction environment is not systematic enough. Secondly, the wear form is affected by a variety of factors, such as the wear resistance of the elements themselves, whether new phases will be formed between the elements, the proportion of elements, etc., How to design and optimize the wear-resistant element ratio of high-entropy alloys is also important. Furthermore, high entropy alloys, bulk amorphous and composite materials are known as the three most promising hotspots in the next few decades. People have high hopes for the new type of high entropy alloy, expecting that it can balance tribological properties and other properties at the same time. How to regulate the key morphology, distribution and interface is also a problem. Finally, how to make full use of the excellent friction and wear properties of high entropy alloy to benefit people's well-being in actual production and life remains to be further studied.

5. Future outlook

With the continuous in-depth study of scholars at home and abroad, the excellent friction and wear properties of high entropy alloys have been recognized, which can greatly expand the field of engineering materials development, and provide more options and ideas for materials workers to use high entropy alloys.

Although there are many researchers in the study, but the study of high entropy alloy properties is far from enough; The future development direction of friction and wear properties of high entropy alloy can be summarized as follows.

- 1) The most comprehensive and in-depth research on the wear resistance of the HEA is mainly focused on 3d transition metal high

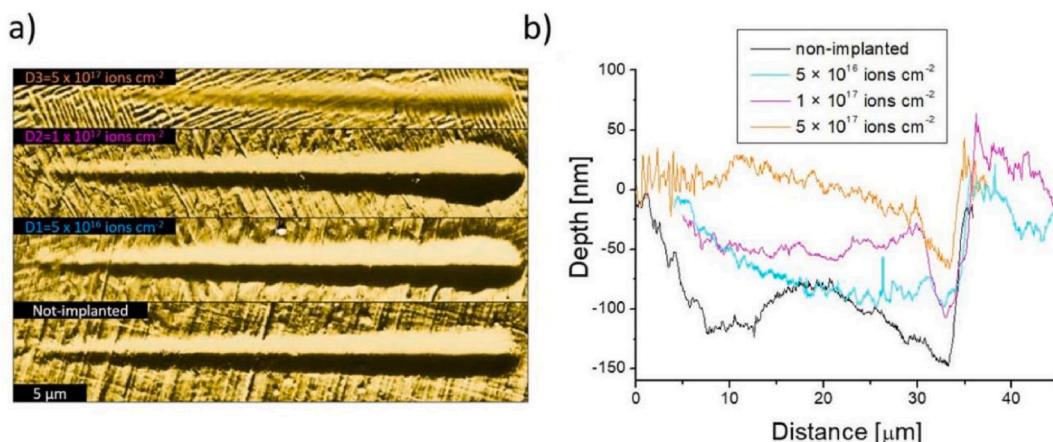


Fig. 21. The $AlCoCrFeNiTi_{0.2}$ Scratch test: (a) the morphology of the wear track and (b) the profile of the representative wear track.

- entropy alloys, and there is still a gap in the research on RHEA and lightweight HEA, especially the latter need to carry out research on the frictional wear properties. The study of friction and wear mechanism is of great significance to enrich the tribological theory and expand the field of anti-wear lubricant materials.
- 2) Frictional wear has always been a major challenge in the application of engineering materials, and surface strengthening technology can effectively improve the anti-frictional wear performance and significantly increase the service life, indicating a strong potential for development. Therefore, combining the HEA with new surface strengthening technologies, surface composite strengthening technologies and surface nano-reinforcement technologies can be a focus of future research in the HEA tribology, and research in this direction is of great significance to the engineering utility.
- 3) For the research work on tribology of the HEA, many scholars have devoted themselves to improving the performance through surface modification techniques, but the current research still needs to further optimize the formulations and processes and continue to seek effective control of friction and wear in different applications. Using the wide composition space and unique structural characteristics, the engineering application value of the HEA under harsh conditions can be improved, such as achieving stable lubrication and anti-wear under extreme working conditions and ensuring anti-wear under specific functional roles, in order to make the anti-wear performance of the HEA close to the range with practical application significance.
- 4) In the tribological performance research, researchers have combined the traditional alloy surface modification methods and methods accumulated during the research process, through different strengthening means to analyze and test the friction and wear resistance of the HEA materials, for excellent performance of the HEA material system, we still need to clarify its strengthening mechanism, the formation of a common mechanism system. At the same time, the development of the HEA tribological properties should focus on breaking through the performance limits of traditional anti-wear and lubricating alloys, solving the bottleneck problems of traditional alloys, and obtaining tribological properties better than those of traditional alloys.
- 5) Almost no model can currently accurately forecast the friction and wear at high temperatures, and the fundamental understanding of the tribological process of the HEA at high temperature is insufficient. Due to the difficulty of friction and wear process experiments and modeling, new technologies and techniques must be developed in order to address high-temperature tribology issues. Fast wear estimation using a machine learning framework might offer an answer.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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