Review



# Research Progress on CoCrCuNi High-Entropy Alloys

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Posted Date: 25 September 2024

doi: 10.20944/preprints202409.2005.v1

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*Article*

# Research Progress on CoCrCuNi High-Entropy Alloys

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**Abstract:** High-entropy alloys have attracted widespread attention from researchers worldwide due to their unique microstructure and outstanding mechanical properties, making them a prominent focus in the field of metallurgy. Among the various high-entropy alloys, the CoCrCuNi system was among the first alloys discovered, and it has shown significant progress in development. By employing different preparation and heat treatment processes, researchers have obtained alloys with diverse performances. The addition of various alloying elements or other components can lead to pronounced variations in the properties of CoCrCuNi high-entropy alloys. This work provides a comprehensive review of recent research progress on CoCrCuNi high-entropy alloys both domestically and internationally. It covers the preparation methods, thermodynamic and kinetic simulation calculations, as well as discussions on heat treatment processes and the influence of alloying elements on the microstructure and mechanical properties of CoCrCuNi high-entropy alloys. Finally, the review concludes with a prospective analysis and predictions for the potential applications and future directions in developing novel high-entropy alloys.

**Keywords:** high-entropy alloys; CoCrCuNi system; microstructure; mechanical properties

## 1. Introduction

With the rapid development of industries such as aerospace, power generation, and chemical engineering, the performance requirements for metallic materials have increased significantly. Traditional alloy systems can no longer meet these demanding conditions. As a result, Yeh et al. 1 and Cantor et al.2 independently introduced the concept of high-entropy alloys (HEAs) in 2004. HEAs are homogeneous solid solutions composed of five or more metallic elements. Unlike conventional alloys, in HEAs, each element is present in relatively equal proportions, forming a highly mixed microstructure. These alloys exhibit excellent mechanical properties, such as high hardness, strength, temperature resistance, and wear resistance, making them highly durable under extreme conditions. Additionally, HEAs have outstanding corrosion resistance, effectively protecting against oxidation and other environmental degradation, which makes them suitable for harsh working conditions, such as turbine blades, aircraft fuselages, wings, and missile guidance systems.

The discovery of HEAs has opened vast possibilities in materials design and has become a major research focus in the field of metallic materials3. HEAs are generally categorized into transition metal high-entropy alloys and refractory high-entropy alloys. Transition metal HEAs typically contain at least four elements from a group of nine: Al, Co, Cr, Cu, Fe, Mn, Ni, Ti, and V. Examples include AlCrFeNi[8], CoCrFeNi[9], AlCoFeNi[10], CoCrCuNi[11], and AlCoCrFe[Error! Reference source not found.]. The CoCrCuNi HEA is composed of an FCC structure with Cu-rich and Cu-poor phases, offering excellent ductility[13], which provides additional advantages for practical applications.

To enhance the overall properties of CoCrCuNi alloys, researchers have explored various preparation techniques to investigate their microstructure and mechanical performance. Derimow et

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al. [14] produced CoCrCuNiFe HEAs using vacuum arc melting and mechanical alloying, finding that the microhardness of the alloy prepared by mechanical alloying was about 2.5 times higher than that prepared by vacuum arc melting. Other scholars have reported that CoCrCuFeNi HEAs prepared via mechanical alloying maintain a single FCC structure below an annealing temperature of 800°C, while at higher temperatures, Cu-rich FCC phases precipitate, separating the single FCC structure into two FCC phases[15].

Furthermore, researchers have found that CoCrCuNiAl consists of a BCC + FCC dual-phase solid solution, while CoCrCuNiFe remains a single FCC phase, indicating that Al promotes the formation of the BCC phase, whereas Fe promotes the formation of the FCC phase [4,16]. Studies also show that in CoCrCuNiFe HEAs, Cu reduces grain size and increases hardness, and at high temperatures, a glassy copper oxide layer forms, improving high-temperature wear resistance16.

It is clear that different preparation techniques, heat treatments, and alloying elements significantly affect the microstructure and mechanical properties of CoCrCuNi-based HEAs. To provide researchers with a comprehensive understanding of the development of CoCrCuNi-based HEAs, this paper reviews recent progress in their research, focusing on preparation methods, thermodynamic and kinetic simulations, and the effects of processing and alloying elements on their properties, while also discussing future research directions and development prospects.

## 2. Preparation Methods of CoCrCuNi-Based High-Entropy Alloys

The emergence of HEAs has sparked widespread interest in the field of materials science, offering new possibilities for various applications due to their exceptional properties. However, the performance of HEAs is closely related to their preparation methods, which can result in significant differences in microstructure, crystal structure, and mechanical properties.

Vacuum melting is a commonly used method for fabricating HEAs and is mainly divided into vacuum arc melting and vacuum induction melting. In vacuum arc melting, an electric arc is generated in a high-vacuum environment, melting the surface of the HEA. The energy from the arc helps homogenize the alloy's composition, enhancing its properties. In vacuum induction melting, electromagnetic induction is used to heat and melt the alloy under high-vacuum conditions, providing uniform heating. However, both methods suffer from low production efficiency, making them unsuitable for large-scale production and industrial applications, and are therefore primarily used for basic research in laboratories. Min et al.[18] used vacuum arc melting to prepare CoCrCuFeNi, CoCrFeMnNi, and CoCrCuFeMn HEAs, with CoCrCuFeMn showing the highest compressive yield stress of approximately 805 MPa, though its fracture strain was relatively low at around 30%. Hsu et al.[19] prepared Al0.5CoCrFeCuNi HEAs using vacuum arc melting, achieving a hardness of 208 HV, which is comparable to the hardness of 211 HV for the same alloy prepared by vacuum induction melting. Sheng et al. [20] and Du[21] both used vacuum induction melting to produce Al0.5CoCrFeCuNi HEAs. Sheng found that the alloy consisted of an FCC1 phase and a small amount of FCC2 (Cu-rich) phase, with a tensile strength of around 1.8 GPa. Du's study, on the other hand, showed that the alloy had a single FCC phase, with tensile strengths of 616 MPa and 593 MPa for samples without and with electromagnetic stirring, respectively.

Powder metallurgy is a method that uses metal or non-metal powders as raw materials to create components through pressing and sintering processes. This method offers advantages such as near-net shaping, excellent hot-working performance, low cost, and high efficiency. Furthermore, due to the small particle size of the powders, rapid cooling leads to a more homogeneous microstructure and the formation of fine equiaxed grains, avoiding the compositional segregation and dendritic structures common in casting. As a result, powder metallurgy often yields better mechanical properties compared to traditional cast alloys. Mukanov et al. [22] used powder metallurgy to prepare CoCrCuxFeNi HEAs and studied their microstructure, wear resistance, and corrosion resistance. Nam et al. [23] also investigated CoCrCuFeNi HEAs produced by powder metallurgy, finding that the alloy contained two FCC phases (a CoCrFeNi-rich phase and a Cu-rich phase), with an average grain size of 59 nm. The alloy achieved high values of hardness (525 HV) and yield strength (1920 MPa). Xia et al. [24] noted that the tensile strength and elongation of CoCrCuFeNi

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HEAs initially increased and then decreased with rising sintering temperatures, reaching a peak tensile strength of 655 MPa and elongation of 21.9 % at. 1100 °C.

Mechanical alloying is a process in which powders are processed via ball milling to achieve atomic-level alloying, mixing of elements, formation of homogeneous structures, and improved plasticity. This technique can also alter grain size by controlling ball milling parameters 24, which allows for effective control of mechanical properties. To obtain bulk HEAs, further processes such as high-temperature sintering or spark plasma sintering (SPS) can be employed. Derimow et al. [14] used mechanical alloying and SPS to fabricate CoCrCuNi, CoCrCuNiFe, and CoCrCuNiMn HEAs, achieving microhardness values of 4.42 ± 0.17 GPa, 4.42 ± 0.17 GPa, and 5.12 ± 0.06 GPa, respectively, which are approximately 2.5 times higher than those of as-cast and annealed alloys. Sriharitha et al. 26 used mechanical alloying and SPS to produce $Al_xCoCrCuFeNi$ HEAs, finding that the grain sizes of all phases were significantly smaller than those of traditionally cast alloys, with hardness differences of up to 120 HV. Additionally, they found that the specific hardness of Al5CoCrCuFeNi alloys could reach 160 HV/g $cm^3$. Gómez-Esparza et al.[27] investigated NiCoAlFeCuCr HEAs produced by mechanical alloying and SPS, reporting that the microstructure consisted of FCC and BCC phases, with a nano-hardness of 269 ± 39 HV.

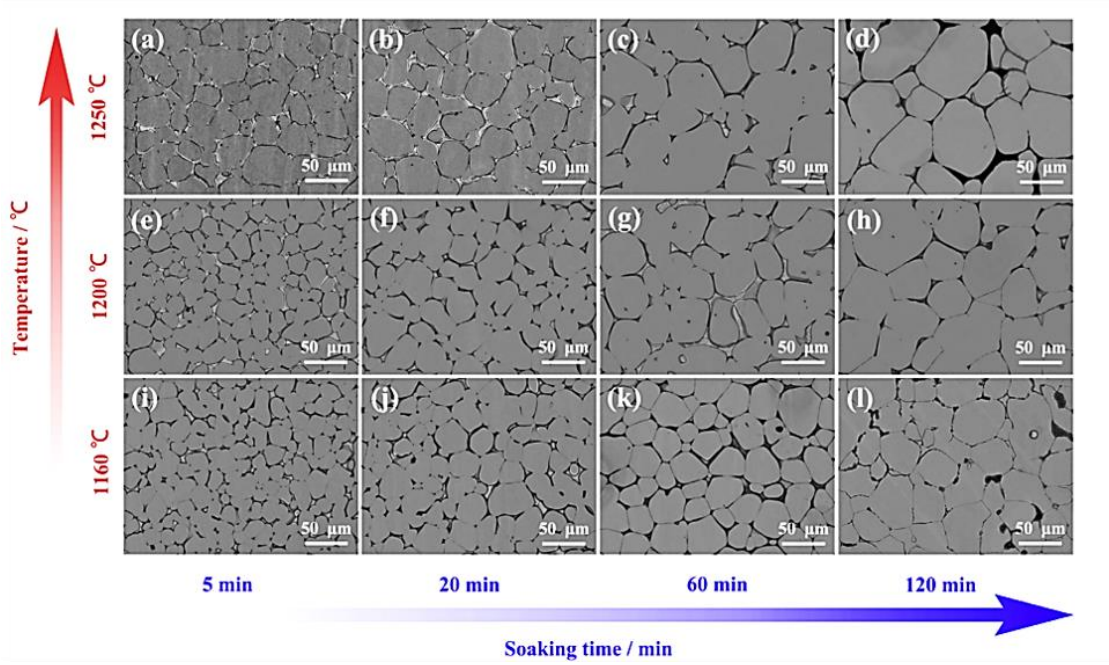


**Figure 1.** he microstructure of CoCrCuFeNi high entropy alloy under isothermal conditions at 1250 °C for 5 minutes (a), 20 minutes (b), 60 minutes (c), and 120 minutes (d); Isothermal 1200 °C: 5 minutes (e), 20 minutes (f), 60 minutes (g), 120 minutes (h); The isothermal temperature is 1160 °C: 5 minutes (i), 20 minutes (j), 60 minutes (k), 120 minutes (l) [27].

Semi-solid processing takes advantage of the unique properties of metals that exist simultaneously in both liquid and solid states within a specific temperature range. In this semi-solid state, metals exhibit relatively low viscosity, making it possible to shape them into specific forms through mechanical or pressure-based methods, such as extrusion, forging, and drawing. Rogal et al.28 employed semi-solid processing to fabricate AlCoCrCuNi HEAs, finding that the yield strength and hardness of the alloy were 1580 MPa and 581 ± 9 HV, respectively, representing an improvement of approximately 20% over the as-cast alloy. Campo et al. [29] investigated the melting behavior and formation of globular structures in semi-solid $CoCrCu_xFeNi$ HEAs. Zhang et al. [30] successfully produced CoCrCuFeNi HEAs using semi-solid processing and found that the grain size and spheroidization degree of the globular grains, as shown in Figure 1, increased with higher isothermal temperatures and extended holding times. When the isothermal temperature and holding time reached 1250 °C and 120 minutes, respectively, the grain size ranged from 45 to 65 μm. The average

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hardness decreased with increasing isothermal temperature and longer holding times, and the relationship between Vickers hardness and average grain size followed the Hall-Petch equation, as illustrated in Figure 2.

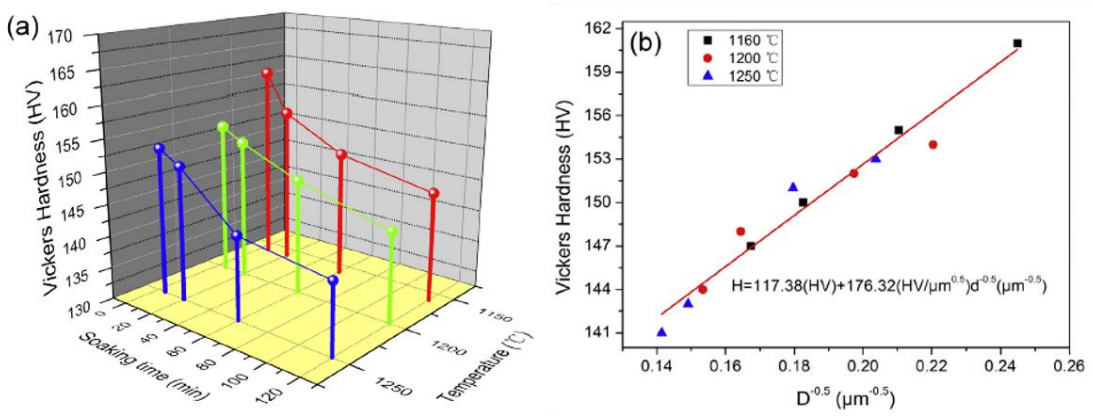


**Figure 2.** The variation of (a) hardness in CoCrCuFeNi high entropy alloy with different isothermal temperatures and holding times; (b) Hardness as a function of average grain size[27].

Laser cladding is an advanced surface modification technique that involves adding a cladding material to the substrate surface and irradiating it with a high-energy laser beam. This process can be completed in a very short time, resulting in a cladding layer with special physical, chemical, or mechanical properties, thereby improving the surface performance of the substrate. Wang et al. 31 studied equimolar AlCoCrCuFeNi HEA fabricated by selective laser melting and found that the alloy exhibited high Vickers hardness, reaching up to 710.4 HV, but had high crack sensitivity. Verma et al. [32] used laser cladding to prepare CoCrCuFeNi and CoCrFeNi HEA coatings on DMR 249A steel. Their research showed that the hardness of CoCrFeNi (415 ± 11 HV) was higher than that of CoCrCuFeNi (310 ± 8 HV). This was due to the larger grain size of the CoCrCuFeNi coating (with average grain length and width of 178 ± 58 μm and 22 ± 12 μm, respectively), and the presence of a second Cu-rich FCC phase, which is relatively soft, further reducing the hardness of the coating. Xu et al. [33] prepared a CoCrCu0.4FeNi HEA coating on Q235 steel using laser cladding and analyzed its microstructure and properties. The microstructure of the coating was mainly composed of dendrites and interdendritic regions, where the dendrites were FCC1, a Cu-rich and Cr-poor phase, while the interdendritic regions were FCC2, a Cr-rich and Cu-poor phase, with a small amount of Cr-rich nanophase also present. The microhardness of the coating ranged from 280 to 300 $HV_{0.2}$, which was approximately 2.5 times higher than the substrate hardness (110 to 130 $HV_{0.2}$).

Magnetron sputtering is one of the methods for fabricating alloy films. Conducted in a vacuum environment, it involves applying a high voltage between the cathode (target) and anode (chamber walls), which generates a magnetically confined glow discharge. High-speed electrons collide with argon molecules, ionizing the argon gas. The argon ions are then accelerated toward the cathode target, bombarding its surface, causing target atoms to be sputtered onto the substrate to form a thin film. The resulting films exhibit high purity and density, although the utilization rate of the target material is not ideal, and this technique does not perform well with ferromagnetic materials. Dolique et al. [34] used magnetron sputtering to fabricate AlCoCrCuFeNi HEA films and investigated the complex structure/composition relationships and thermal stability. Li et al. [35] studied the mechanical properties of CoCrCuFeNi films deposited on Kapton substrates using magnetron sputtering. The results indicated that the residual stress evolved from compressive to tensile with increasing film thickness, showing typical grain growth characteristics. Cai[36] prepared CoCrCuFeNi single-layer films with thicknesses of 30, 20, and 10 nm using magnetron sputtering. It was found that the thinner the film, the more BCC phase precipitated after annealing at 550°C. The dual-phase HEA film consisted of uniform equiaxed grains with an average size of about 40 nm. Compared to the single-phase FCC HEA, the dual-phase HEA exhibited higher hardness, reaching up to 10.4 GPa.

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## 3. Thermodynamic and Kinetic Simulation of CoCrCuNi-Based High-Entropy Alloys

The structure and properties of HEAs are closely interconnected, making structure prediction and kinetic calculations critical in their development. Studying phase formation rules through experimental trial and error can be highly time-consuming, especially under extreme conditions such as ultrahigh temperatures and pressures. Characterizing the crystal structure of materials through experimental methods becomes particularly challenging in such scenarios. As a result, computational simulations play a pivotal role in predicting crystal structures and calculating material properties.

Wang et al.[37], using first-principles density functional theory (DFT) combined with pseudopotential plane-wave and generalized gradient approximation (GGA) methods, modeled the effect of Al content on the $Al_xCoCrCuFeNi$ HEA using the virtual crystal approximation (VCA) method. As the Al content increased, the formation enthalpy of the alloy increased, while its density decreased. When x = 1, the lattice constant was at its minimum; when x = 2, the alloy met thermodynamic stability criteria. Based on Poisson's ratio, Al2CoCrCuFeNi was classified as a ductile material, while the others were brittle. Under these conditions, the high-entropy alloy could be considered thermodynamically stable. In another study, Wang et al.[38] calculated the structural properties, elastic moduli, and pressure-induced phase transformations of the AlCoCrCuFeNi HEA under varying pressures. The results indicated that with increasing pressure, the lattice constant gradually decreased while the density increased. The AlCoCrCuFeNi alloy satisfied mechanical stability criteria only in the FCC structure, not in the BCC structure. Additionally, the bulk modulus increased with pressure. The alloy was predicted to undergo a phase transition from FCC to BCC at approximately 21,820 GPa.

Cheng et al.[39] conducted a detailed investigation on the phonon behavior of CrFeCoNiCu high-entropy alloys (HEAs) across different temperatures using molecular dynamics (MD) simulations and first-principles calculations. Their study particularly focused on the impact of anharmonic phonon scattering on the structural stability and thermal conductivity of the alloy. It was found that below 800°C, the CrFeCoNiCu alloy maintains stability as a single FCC solid solution phase. However, after annealing above 800°C, the alloy separates into a dual-phase structure consisting of Cu-rich and Cu-poor FCC phases. The study also demonstrated that the higher the Cu content, the more prone the alloy is to phase separation from a single FCC phase into a dual-phase structure. This phase separation becomes most prominent when the Cu content reaches 50-60 at%. Additionally, the contribution of phonons with different wavelengths to lattice thermal conductivity was analyzed. The results revealed that, at 300K, long-wavelength phonons contribute approximately 10 times more to thermal conductivity than short-wavelength phonons. Although the anharmonic scattering of short-wavelength longitudinal acoustic (LA) phonons is stronger, the higher group velocity of long-wavelength phonons makes them the primary contributors to heat conduction. This study quantified the anharmonic phonon behavior of CrFeCoNiCu HEAs over a wide temperature range from 10K to 1200K, shedding light on its effects on structural stability and thermal performance. These findings provide theoretical support for the design and optimization of HEAs in thermoelectric materials and thermal management applications.

Liu[40] conducted molecular dynamics simulations to study the impact of phase separation on the mechanical behavior of CoCrCuFeNi HEAs. The findings revealed that Cu-rich grains significantly reduced the alloy's strength. The larger the volume fraction of Cu-rich grains, the more pronounced the strength reduction. This was attributed to the lower critical shear stress required to activate slip systems in Cu-rich grains compared to Cu-poor grains, making the former more prone to plastic deformation. Additionally, uneven stress distribution was observed, with Cu-poor grains experiencing much higher stress than Cu-rich grains.

Liu et al. [41] further investigated the relationship between mechanical properties and the onset temperature of plasticity in CoCrCuFeNi HEAs using molecular dynamics simulations. The study showed that the mechanical properties of the alloy were highly sensitive to temperature: both the elastic modulus and ultimate tensile strength increased as temperature decreased, as illustrated in Figure 3. As the temperature increased, atomic thermal motion intensified, reducing the bond energy between atoms. This suggests that the alloy's lattice deforms with increasing temperature, leading to

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a decrease in elastic modulus. However, compared to Cu, the alloy exhibited superior heat resistance in terms of elastic modulus and ultimate tensile strength.

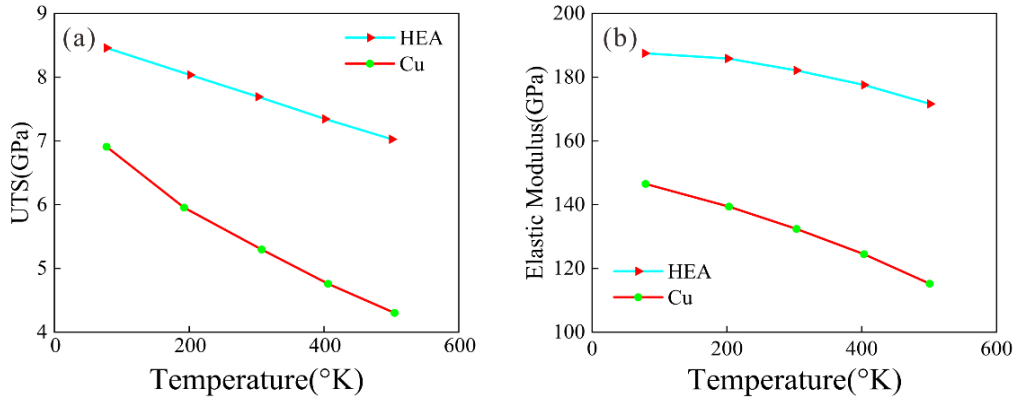


**Figure 3.** (a) Effect of temperature on ultimate tensile strength; (b) The influence of temperature on elastic modulus[41].

Liu et al.[42] investigated the uniaxial tensile and compressive deformation behavior of CoCrCuFeNi HEA with different orientations at various temperatures using molecular dynamics simulations. The results indicated that temperature-induced softening occurred in all simulated conditions. The mechanical properties of CoCrCuFeNi HEAs exhibited orientation dependency and tension-compression asymmetry. Furthermore, twin boundary (TB)-assisted dislocation multiplication was observed. Compared to low-entropy materials and theoretical calculations, alloying was found to reduce the stacking fault energy (SFE) of the crystal, thereby enhancing its ability to undergo twinning deformation.

Tran[43] employed molecular dynamics simulations to study the deformation mechanisms and tensile properties of nanocrystalline CoCrCuFeNi HEAs. The study revealed that stacking faults formed during tensile deformation, and the crystal structure primarily transformed from FCC to HCP. Tensile strength decreased as grain size decreased. As the Cu content decreased, the formation of stacking faults became more frequent, and grain elongation occurred in regions prone to abrupt deformation. Cu-rich samples exhibited significantly lower tensile strength and dislocation density compared to CoCrFeNi-rich samples.

Qi et al. [44] used molecular dynamics simulations to explore the effect of Cu on the mechanical properties of CoCrFeNi-based HEAs. The results showed that the addition of Cu reduced the stacking fault energy, thereby increasing the hardness of the alloy. Additionally, the presence of Cu promoted the amorphization of the alloy.

These findings suggest that computational simulations can be used to design ideal CoCrCuNi-based HEAs with high lattice structure stability and heat resistance, making them promising candidates for high-temperature structural materials. Furthermore, the mechanical properties of HEAs can be enhanced through compositional adjustments and microstructural control.

## 4. Effects of Heat Treatment Processes on CoCrCuNi-Based High-Entropy Alloys

HEAs often exhibit remarkable advantages in certain properties, and their microstructure, elemental distribution, and phase composition can be significantly altered through appropriate processing techniques. These changes can, in turn, affect the mechanical, physical, and chemical properties of the alloys. Yi et al. 45 investigated the microstructure and mechanical properties of CoCrCuNiTi HEA under different annealing conditions. The study revealed that both as-cast and annealed alloys were composed of a complex multiphase structure, as shown in Figure 4, including two BCC phases, one FCC phase, one cubic Laves phase, and an HCP-structured $Ni_3Ti$ phase. The two BCC phases accounted for approximately 50% of the total volume, while the remaining three phases were distributed throughout the other regions. The alloy demonstrated excellent ultimate compressive strength under both casting and annealing conditions (as-cast: 2.53 GPa, annealed: 2.22

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GPa). The hardness of the as-cast alloy was relatively high, reaching 694 HV, while the annealed alloy exhibited improved plastic strain, achieving 7.8%.

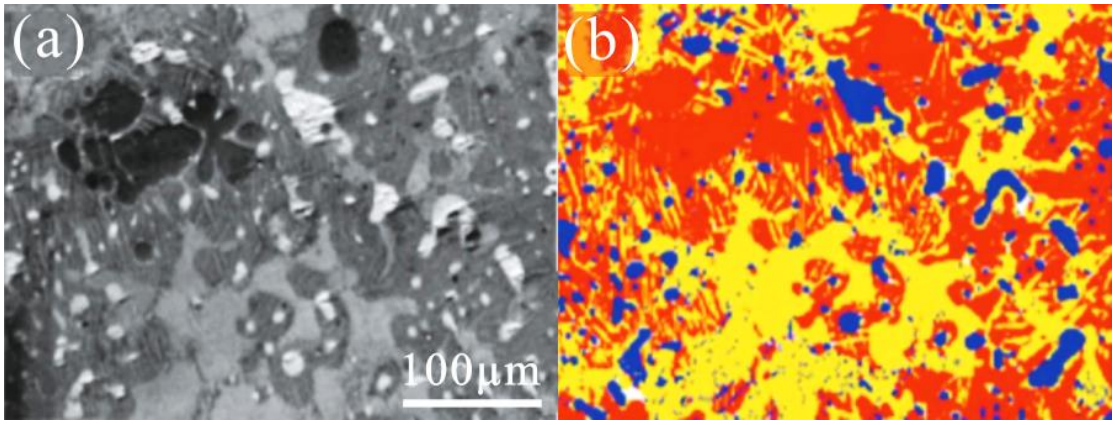


**Figure 4.** (a) SEM of the microstructure of high entropy alloy CoCrCuNiTi; (b) Phase diagram of high entropy alloy (a): BCC (red), FCC (blue), HCP (yellow) [45].

Liu et al.[Error! Reference source not found.] investigated the annealing microstructure and hardness variation of AlFeCuCoNiCrTi$_x$ high-entropy alloys. The experiments revealed that with the increase in annealing temperature, the phase composition of the Ti$_0$ alloy gradually transitioned from an original FCC+BCC structure to an FCC1+FCC2+BCC structure after 636 ℃. The hardness increased slightly at 636 ℃, decreased significantly between 636 and 1112 ℃, and then remained relatively stable beyond 1112 ℃. For the Ti0.5 alloy, annealing had little effect on its phase composition, maintaining an FCC+BCC1+BCC2 structure throughout. Its hardness increased slightly at 607 ℃, decreased significantly between 607 and 1092 ℃, and remained stable after 1092 ℃. For the Ti$_1$ alloy, an Fe$_2$Ti-type Laves phase precipitated when the annealing temperature reached 800 ℃, contributing to an increase in hardness. When the annealing temperature reached 1200 ℃, the hardness increased to 51 HRC.

Singh et al. 46 studied the phase composition of equiatomic AlCoCrCuFeNi high-entropy alloys produced by sputter quenching and traditional casting. The experiments found that only a single BCC phase formed in the alloy produced by sputter quenching, which differs from the traditionally cast alloy, which contained one BCC phase and two FCC phases (FCC1 and FCC2). More detailed TEM studies revealed that the sputter-quenched alloy contained a partially ordered BCC phase with nanometer-sized domain structures. In the as-cast alloy, the interdendritic regions consisted of a BCC phase and FCC phase with lamellar B2 Cu-rich regions, and the rhombic L1$_2$ Cu-rich phase (FCC2). The dendritic regions contained the L1$_2$ Cu-rich phase (FCC1). Detailed local composition analysis using atom probe tomography showed that the alloying elements in the Ni-Al-rich and Cr-Fe-rich phases were not randomly distributed but segregated during solidification due to the large mixing enthalpy differences among elements like Al-Ni, Cu-Ni, and Fe-Cr in high-entropy alloys.

Lin et al. 48 explored the relationship between phase transformation and hardness in as-cast and heat-treated FeCoNiCrCu0.5 high-entropy alloys. The study showed that both the as-cast and annealed alloys exhibited an FCC phase, with the TEM results shown in Figure 5. A combination of SEM and TEM indicated that when the temperature was below 350 ℃, the Cu-rich phase appeared as a continuous precipitate. As the annealing temperature increased, both continuous and discontinuous precipitates coexisted. When the annealing temperature reached 1350 ℃, the Cu-rich phase transitioned into short and needle-like precipitates. The hardness of the as-cast alloy was 174 HV, and it decreased gradually with increasing annealing temperature. The reduction in hardness was attributed to the weakening of atomic bonding, mainly due to the positive mixing enthalpy between Cu and Co/Ni.

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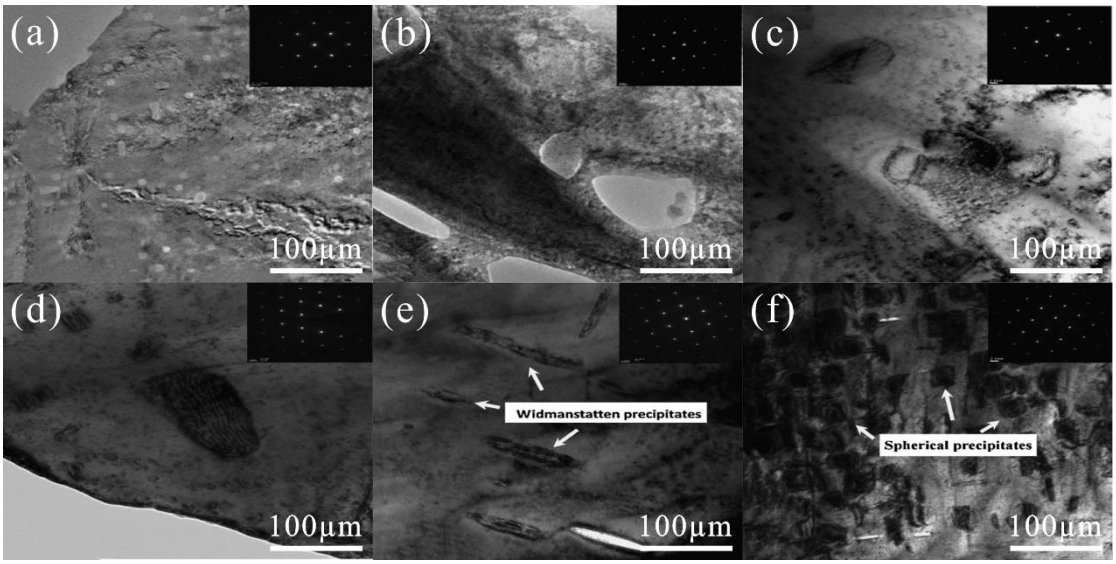


**Figure 5.** TEM microstructure of FeCoNiCrCu0.5 alloy under different heat treatment temperatures, with the inset showing the SAED pattern: (a) as cast; (b) 650 ℃; (c) 950 ℃; (d) 1100 ℃; (e) 1250 ℃; (f) 1350 ℃ annealing temperature [48].

Deng et al. 49 investigated the effects of strong magnetic field-assisted heat treatment on the microstructure and properties of AlCoCrCuFeNi high-entropy alloys. The study found that applying a magnetic field refined the FeCoCr-rich phase and suppressed large-scale Cu-rich phase segregation. As a result, the alloy's maximum compressive strength increased from 1482 MPa to 1795 MPa, and elongation improved from 23% to 27%. These changes were mainly attributed to the reduction in the BCC phase volume fraction, decreased Cu-rich segregation, and grain refinement under the influence of the strong magnetic field, leading to enhanced mechanical performance.

Wen et al. 50 studied the microstructure and properties of AlCoCrCuFeNi high-entropy alloys after aging at temperatures ranging from 500 to 1000 ℃. The results showed that, initially, with increasing aging temperature, the alloy's BCC and FCC phase structures remained unchanged, with yield and compressive strengths of approximately 1750 MPa and 2100 MPa, respectively. However, when the temperature exceeded 645 ℃, the BCC phase in the microstructure gradually transformed into the FCC phase, leading to a reduction in yield strength and an increase in elongation. At 1000 ℃ aging, the alloy's elongation reached as high as 27%.

Wang et al. [51] reported a detailed structural characterization and tensile testing of $CoCrFeNiMn_{0.75}Cu_{0.25}$ high-entropy alloys after cold rolling and subsequent annealing at various temperatures. They found that the annealing temperature significantly influences the microstructural evolution and mechanical properties of the alloy. Specifically, as the annealing temperature increases, the recrystallized grains within the alloy coarsen, accompanied by various transformations in grain boundary types and the precipitation of phases. The mechanical testing results indicate that adjusting the annealing temperature can optimize the strength-ductility balance of the alloy, with the best overall mechanical performance achieved at 1173 K, where the yield strength reached 767 MPa, tensile strength was 1128 MPa, and elongation was 18.3%. The study reveals that at higher annealing temperatures, the primary strengthening mechanism is grain boundary strengthening, whereas at lower temperatures, strengthening arises from a combination of precipitates, dislocations, and high-angle grain boundaries. This research provides valuable theoretical support for the optimization design of high-entropy alloys by exploring the impact of different annealing temperatures on the microstructure and its relationship with mechanical properties.

## 5. Effects of Alloying Elements on the Microstructure and Properties of CoCrCuNi-Based High-Entropy Alloys

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To further study and improve the performance of HEAs based on the CoCrCuNi system, many researchers have introduced various alloying elements to modify the alloy's microstructure. Hsu et al.52 investigated the effects of adding Fe, Ag, and Au on the microstructure and hardness of AlCoCrCuNi equimolar high-entropy alloys. The experiments showed that the microstructure of the Fe-added alloy was similar to that of the as-cast alloy, both exhibiting dendritic structures with a microhardness of approximately 420 HV. After adding Ag, the alloy's microstructure was divided into two layers: a golden layer composed of Ag-rich dendrites and inter-dendritic Ag-Cu phases, and a silver layer mainly consisting of dendrites rich in Al, Co, Cr, and Ni, with a small amount of Cu in the inter-dendritic regions. Consequently, there was a significant difference in microhardness between the two layers, with values of 104 HV and 451 HV, respectively. This was attributed to the eutectic Ag-Cu phases in the former and the lower Cu content in the latter. Additionally, it was found that Au mixed well with the other five elements, suggesting that Au acts as a mixing agent between Cu and the other elements. Moreover, it was determined that for effective mixing in six-element alloys, the maximum positive enthalpy between atomic pairs should not exceed approximately 10 kJ/mol. When designing HEAs, the interaction of the mixing enthalpy between elements should be considered.

Chandrakar et al. 53 studied the effects of Si addition on the phase transformation and mechanical properties of AlCoCrCuFeNiSix alloy systems. XRD analysis revealed that the AlCoCrCuFeNiSix HEA powders underwent dual-phase evolution of FCC and BCC during mechanical alloying. As the Si content increased, the proportion of the BCC phase significantly increased. After spark plasma sintering, the $AlCoCrCuFeNiSi_x$ (x=0.9) HEA was composed of a BCC phase, along with minor FCC and $\sigma$ phases. Furthermore, the evolution of the $\sigma$ phase may have resulted from the partial transformation of the BCC phase formed during mechanical alloying. The alloy's hardness and wear resistance improved with increasing Si content, primarily due to the evolution of the BCC and σ phases and solid-solution strengthening. Additionally, since Si has a smaller atomic radius than other elements in the alloy, it reduced dislocation mobility and increased strain energy, thereby enhancing the solid-solution strengthening effect. Kumar et al.54 found that when the Si content reached 0.3, the AlCoCrCuFeNi HEA produced by spark plasma sintering exhibited FCC and BCC dual phases. At a Si content of 0.6, the σ phase appeared alongside the BCC and FCC phases. With increasing Si content, the alloy's hardness and wear resistance improved due to the formation of BCC and σ phases in conjunction with solid-solution strengthening. These results indicate that adding the non-metallic element Si is an effective way to enhance the strength and hardness of HEAs.

Qin et al.55 reported the effects of Nb on the microstructure evolution and mechanical properties of CoCrCuFeNi HEAs. The results showed that as the Nb content increased from 0% to 16%, the alloy's compressive yield strength increased from 338 MPa to 1322 MPa, while the fracture strain decreased from 60% to 8.1%. This demonstrates that Nb has a positive effect on improving the mechanical properties of HEAs, primarily because the addition of Nb promotes the transformation of the FCC phase into the Laves phase. As the Nb content increased, the volume fraction of the Laves phase rose from 0% to 58.2%. The increase in the hard Laves phase fraction was the key factor contributing to the enhanced strength, while the decrease in the valence electron concentration (VEC) due to Nb addition favored the formation of the Laves phase. Therefore, Nb is promising for strengthening CoCrCuFeNi HEAs, which are known for their excellent ductility but relatively low strength. Similarly, Cheng et al.56 found that Nb had a significant effect on the microstructure and mechanical properties of CoCrCuFeNi HEA coatings. The study showed that the addition of Nb resulted in the formation of an ordered Laves phase and an FCC solid solution. Nb addition increased the alloy's microhardness (H), elastic modulus (E), hardness-to-elastic modulus ratio (H/E), and resistance to plastic deformation ($H^3/E^2$). Furthermore, under the same wear test conditions, the wear resistance of Nb-containing coatings was approximately 1.5 times higher than that of Nb-free coatings. In a 6 N HCl solution, Nb-containing coatings exhibited the lowest corrosion current density and overall corrosion rate. These results indicate that Nb-doped HEA coatings possess excellent mechanical properties, providing valuable guidance for expanding their industrial applications.

 doi:10.20944/preprints202409.2005.v

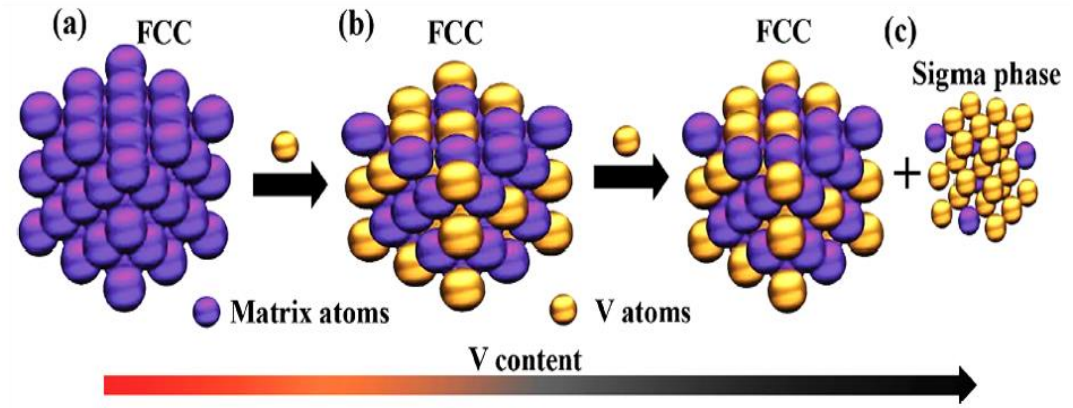


Sun et al. 57 investigated the effects of Ti content on the microstructure of AlFeCuCoNiCrTix HEAs. The results showed that AlFeCuCoNiCr exhibited a typical dendritic structure, which changed significantly with the addition of Ti. When the Ti content was x=0.5 and x=1, the alloy displayed a eutectic structure. Thus, Ti can promote the transition from a dendritic to a eutectic microstructure. A comparison of alloys with different Ti contents revealed that Ti first refined and then coarsened the grain structure. The as-cast alloy was a supersaturated solid solution that underwent spinodal decomposition during cooling to reduce energy, resulting in the formation of nano-precipitates and amorphous phases due to the effects of slow diffusion and severe lattice distortion. The AlFeCuCoNiCrTix alloys were composed of BCC and FCC phases, along with a small amount of intermetallic compounds, and exhibited an ordered lattice structure. With the addition of Ti, the phase structure gradually transitioned from FCC+BCC to FCC+BCC1+BCC2, with BCC2 becoming the dominant phase. All three alloys exhibited compositional segregation, especially of Cu. The addition of Ti promoted Cu segregation due to the high mixing enthalpy between Ti and Cu.

Zhuang et al.58studied the effect of Cr on the microstructure and mechanical properties of FeCoNiCuAl HEAs. The experiments demonstrated that both FeCoNiCuAl and FeCoNiCuAlCr alloys were primarily composed of simple FCC+BCC solid solution phases, with the BCC phase being the primary Cu-poor phase. Cr segregated in the dendritic regions of the alloy, enhancing Cu segregation in the interdendritic regions, but it had no significant effect on the lattice constant of the BCC phase. The maximum compressive strength and true strain of FeCoNiCuAl and FeCoNiCuAlCr alloys were 1.33 GPa, 9%, and 1.51 GPa, 16.0%, respectively. It is evident that adding Cr can significantly improve both the compressive strength and ductility of the alloy.

Zhu et al. 59 reported the influence of Mo on the microstructure and properties of AlCoCrCuFeNi HEAs. The study found that as-cast AlCoCrCuFeNiMox alloys exhibited simple BCC and FCC solid solution structures. When the Mo content was 0 and 0.2, the alloy exhibited a typical as-cast dendritic and interdendritic microstructure. When the Mo content exceeded 0.2, the alloy's microstructure transitioned to a eutectic structure. With increasing Mo content, the alloy's strength increased significantly, while its ductility decreased to varying degrees. When the Mo content reached 0.8, the maximum yield strength of the alloy was 1920 MPa, and at a Mo content of 0.6, the maximum compressive strength reached 2820 MPa. The strength improvement was attributed to the solid-solution strengthening effect of Mo and the formation of lamellar phases in the eutectic structure. Additionally, the addition of Mo suppressed low-temperature phase transformations and increased the high-temperature phase transition temperature, which benefited the alloy's high-temperature performance. Consequently, the alloy is expected to exhibit excellent high-temperature properties.

Qin et al.60 studied the effects of V on the microstructure and mechanical properties of CoCrCuFeNi HEAs. The results showed that the addition of V promoted the precipitation of the σ phase from the FCC phase in $(CoCrCuFeNi)_{100-x}V_x$ HEAs, thereby improving the alloy's strength, as shown in Figure 6. When the V content increased from 0% to 16%, the volume fraction of the σ phase increased from 0% to 12%, and the compressive yield strength of $(CoCrCuFeNi)_{100-x}V_x$ HEAs increased from 300 MPa to 613 MPa. However, the compressive fracture strain decreased from 50% to 28%.



 doi:10.20944/preprints202409.2005.v



**Figure 6.** Atomic diffusion model for sigma phase formation in $(CoCrCuFeNi)_{100-x}V_x$ HEAs[60].

Yang et al. [61] found that the addition of carbon significantly influences the microstructure and properties of $Al_2CoCrCuFeNi$ high-entropy alloys. The $Al_2CoCrCuFeNiC_{0.02}$ alloy consists of an ordered BCC (B2) phase and an FCC phase, with hardness increasing by approximately 58% upon carbon addition. The interstitial carbon atoms did not form intermetallic compounds with other alloying elements under the high-entropy effect; instead, they formed a simple solid solution. The high hardness values of the alloy can be attributed to the combined effects of lattice frictional stress, solid-solution strengthening, dislocation strengthening, and grain boundary strengthening.

Wang et al.[62] investigated the influence of the Ni/Cr ratio on the phase structure, microstructure, and mechanical properties of $Ni_xCoCuFeCr_{2-x}$ (x = 1.0, 1.2, 1.5, 1.8) HEAs. The study revealed that as the Ni/Cr ratio increases, the σ phase gradually diminishes, and the FCC2 phase nearly disappears, leading to a more homogeneous microstructure within the alloy. In terms of mechanical properties, while the yield strength ($\sigma_y$) does not exhibit a monotonic trend, it reaches a maximum value in the $Ni_{1.2}CoCuFeCr_{0.8}$ alloy, which simultaneously shows the minimum plasticity ($\varepsilon_p$). This behavior is attributed to the role of the σ phase as a secondary strengthening phase. Furthermore, the solid-solution strengthening effect of Cu becomes significant in the $Ni_{1.5}CoCuFeCr0.5\}$ alloy, partially offsetting the strength reduction caused by the decrease of the σ phase, resulting in optimal overall mechanical performance for this alloy. Additionally, the densification process of the alloy powders was analyzed using differential scanning calorimetry (DSC) and spark plasma sintering (SPS) techniques, revealing that the FCC2 phase may have acted similarly to a brazing material during the SPS process, facilitating densification. Wang et al. [63] also found that both the yield strength and compressive strength of the alloy exhibit a variation pattern from low to high and then back to low, with the AlCoCrCuFeNi alloy demonstrating the best overall performance. As the Ni content increases, the compression fracture transitions from cleavage fracture to quasi-cleavage fracture and ultimately to ductile fracture, indicating a gradual transformation of the alloy from a brittle material to a ductile one.

In summary, alloying elements can significantly alter the crystal structure, atomic arrangement, compositional uniformity, and mechanical properties of high-entropy alloys. The CoCrCuNi-based high-entropy alloys can mitigate Cu segregation and adjust the ratio of FCC to BCC phases by varying the content of alloying elements. Alloy design must comprehensively consider these factors to achieve the desired properties, such as enhanced strength, hardness, corrosion resistance, and high-temperature stability.

## 6. Conclusion and Outlook

This paper focuses on the preparation processes and computational simulations of CoCrCuNi-based high-entropy alloys (HEAs), exploring the effects of various treatments such as annealing and aging, as well as the addition of different alloying elements on the microstructure and mechanical properties of the alloys. With the advancement of computational materials science, it is now possible to provide theoretical references through computational simulations and to predict alloy phase compositions via phase diagram calculations. However, current understanding of the phase compositions and crystal structures of high-entropy alloys primarily relies on their as-cast states, and there is still a lack of in-depth discussion regarding their equilibrium states and phase diagrams. Future research in this field will require more integration of experimental and computational efforts to reduce trial-and-error time and costs.

Most existing studies on CoCrCuNi-based HEAs focus on their as-cast structures, which often exhibit various casting defects. Therefore, altering the alloy's microstructure and properties through heat treatment processes such as annealing, quenching, and solution aging is one of the key future research directions. Additionally, enhancing the mechanical properties—such as strength, hardness, and ductility—by modulating the composition of the FCC and BCC phases or forming intermetallic compounds through the addition of various alloying elements remains a critical focus for future studies.

 doi:10.20944/preprints202409.2005.v



Research on CoCrCuNi-based HEAs is ongoing, and more investigations are expected to clarify the relationship between their microstructures and properties, thereby further improving their overall mechanical performance. The application fields of high-entropy alloys are set to expand, including aerospace, automotive, energy, and biomedical sectors, as their properties meet the demands across multiple applications. In summary, CoCrCuNi represents one of the most characteristic alloy systems in high-entropy alloys, and its unique properties offer promising prospects in various applications. Through deeper research and exploration, its potential can be further unlocked, paving the way for broader applications. With continuous technological advancements, it is anticipated that high-entropy alloys will showcase even more exceptional properties and potential in the future.

**Acknowledgements:** F.F. Wu acknowledges the National Natural Science Foundation of China (Grant No 51971106) and the Programs for Innovation Groups in Liaoning Universities and Liaoning Distinguished Professor. X.M. Zhao acknowledges the Youth Project of Liaoning Provincial Department of Education (LJKQZ20222270).

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