Study on the mechanical properties and corrosion resistance of AlxCoFeNiCr1−x high-entropy alloys

Shuai Wang, Yuhong Zhao, Peng Cheng, Qingwei Guo, Xiaotao Xu and Hua Hou
School of Materials Science and Engineering, North University of China, Taiyuan 030051, People’s Republic of China
E-mail: zhaoyuhong@nuc.edu.cn

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Abstract
The AlxCoFeNiCr1−x (0.1 ≤ x ≤ 1) high entropy alloys (HEAs) were prepared by repeated vacuum arc-melting in a Ti-gettered high-purity argon atmosphere to investigate the effects of the different Al/Cr ratio on the microstructure, mechanical properties and corrosion resistance in a 3.5 wt% NaCl solution. The experimental results show that microstructures shift from the FCC-dominated phase to BCC-dominated phase, and phase segregation switches from BCC phase rich in (Al,Ni) to FCC phase rich in (Fe,Ni) with the molar ratio of the Al/Cr increase. It was found that, with the FCC-dominated phase HEAs, the crystal phase changes from primarily single FCC phase (Al0.1CoFeNiCr0.9, Al0.2CoFeNiCr0.8, and Al0.3CoFeNiCr0.7) to a duplex FCC and BCC structure (Al0.4CoFeNiCr0.6, Al0.5CoFeNiCr0.5, and Al0.6CoFeNiCr0.4). With the molar ratio of Al/Cr further increase, The microstructures of the BCC-dominated of HEAs transform from the FCC+BCC/B2 phase (Al0.7CoFeNiCr0.3, Al0.8CoFeNiCr0.2) to FCC + BCC (Al0.9CoFeNiCr0.1) phase, and then to a single BCC structure (Al1.0CoFeNiCr0). The hardness and compression tests analysis reveals that suitable Al/Cr ratio addition to AlxCoFeNiCr1−x HEAs can improve the comprehensive mechanical properties before reaching the highest point at the Al0.8CoFeNiCr0.2 HEAs, while excessive Al/Cr ratio addition will completely sacrifice plasticity, such as Al0.9CoFeNiCr0.1, Al1.0CoFeNiCr0 HEAs. Moreover, the different Al/Cr molar ratio of the AlxCoFeNiCr1−x HEAs on corrosion resistance were discussed. The results showed that, in the single-FCC phase, the corrosion resistance of Al0.1CoFeNiCr0.9, Al0.2CoFeNiCr0.8, Al0.3CoFeNiCr0.7 HEA have gradually decreases, while in a dual phase of AlxCoFeNiCr1−x HEAs, the tendency of corrosion resistance is consistent with the volume fraction of segregation phase. The less the volume fraction of segregation phase of alloys have, the larger the corrosion resistance of it retain, such as Al0.4CoFeNiCr0.6, Al0.5CoFeNiCr0.5, and Al0.6CoFeNiCr0.4. When the volume fraction of segregation phase is close to the matrix phase, it has the worst the corrosion resistance, such as Al0.7CoFeNiCr0.3 HEAs.

1. Introduction
High entropy alloys (HEAs) arise from a new alloy design concept with multi-principal elements, which more than 5 and less than 13 kinds of element components are added, and the atomic content of each element ranges between 5% and 35%. Since the HEAs was proposed, it has attracted extensive attention from scholars. Hundreds of HEAs have been reported by researchers as yet [1]. Contrasted with the other HEAs’ systems, the AlCoCrFeNi HEAs’ system is more mature and excellent performance in many aspects, such as high hardness, strength and ductility, corrosion resistance and superior thermal stability, etc [2, 3], which makes them potential materials for requests demanding high temperature, high wear and corrosion resistance. However, it can well widely applied, which is due to the effects of performance, cost and density. Moreover, by investigating the addition of single element content in AlCoCrFeNi HEAs, it can be found that the increasing Al contents, which is a stabilizer to AlxCoFeNiCr1−x, prompt the formation of BCC/B2 phase [4], adding Fe element can result in
strengthening BCC/B2 phase while reducing cost [5], and raising Co element can enhanced solid solution effect under high temperature conditions etc [6]. With the further study of AlCoCrFeNi HEAs, the effect of the different molar ratio of these elements on microstructures and mechanical properties have drawn greater attention of scholars in recent years. Harihar et al [7] reported that, as the molar ratio of Al/Ni increase, the lattice structure of Al4FeCoCrNi12−x (0.3, 1) was changed from BCC phase to FCC. Borkar et al [8] studied the magnetic properties and microstructure of AlCoFeNiCr1−x (0 ≤ x ≤ 1) HEAs founded that the spinodal decomposition of microstructures, which is mainly composed of BCC/B2 phase, grew rapidly with the addition of Co element. Based on the different ratios of Al, Co, Fe, Ni and Cr elements, several studies such as Al20Co15Cr15Ni70−x, Al2CrCo2−xFeNi10, Al2Cr0.2Fe0.2Ni0.6−xAl0.2Cr0.2Fe2Ni0.6−x [11], Al20−xCr20+0.5xFe20Co20Ni20+0.5x [12], (Al2Co2Cr2Fe24Ni24)100−xCr [13], (AlNiCoFeCr)14 [14], (AlCo2Cr2Fe24Ni24)100−xCr [15] have been extensively developed in the current literature. Although much of these papers, which associated with the different molar ratio of these elements in AlCoCrFeNi HEAs, had been reported, the influence of changing Al/ Cr molar ratio on mechanical behavior are not researched. By comparing the previous studied, it may be found that the addition of decent Al can strengthen the overall mechanical properties of these alloys, the excess Al can cause plastic degradation [16–18], and the addition of an appropriate amount of Cr is beneficial to improve plasticity of alloys [8, 15, 16]. Thence, investigating the different molar ratio of Al/Cr is significant for AlCoCrFeNi HEAs.

It is universally acknowledged that Co and Ni elements are indispensable for studying the high temperature resistance of alloys, and Al and Cr are vital to exploring the corrosion resistance of alloys [17]. The density and cost of AlCoFeNiCr HEAs is widely higher due to these alloys are rich in Fe, Co, Ni and Cr, and poor in Al, which limits their practical application. It is also known that Cr, Ni, Co and Ti elements can create the improvement of corrosion resistance in corrosive solution, while Al Cu and Mn weaken this performance [18, 19]. Because Al and Cr elements can easily induce dense oxide films on alloy surface, the different molar ratio of Al/Cr are inclined to exhibit large corrosion differences. In order to drop the cost of preparing materials, lower the alloy density, and compensate for the defect of present AlCoCrFeNi HEAs' system under the maintaining excellent comprehensive performance, the microstructure evolution and corrosion resistance of AlxCoFeNiCr1−x HEAs are investigated by changing the molar ratio of Al/Cr.

2. Experimental procedures

According to the different molar ratio of Al/Cr, the AlxCoFeNiCr1−x (abbreviated as Al0.1Cr0.9, Al0.2Cr0.8, Al0.3Cr0.7, Al0.4Cr0.6, Al0.5Cr0.5, Al0.6Cr0.4, Al0.7Cr0.3, Al0.8Cr0.2, Al0.9Cr0.1, Al1Cr0) were prepared by Non-consumable vacuum arc melting in a Ti-gettered high-purity argon atmosphere. Each alloy, with a total mass of 50 g and metal granules of Co, Cr, Fe, Ni, Al with purities of higher than 99.5%, was repeatedly smelted 5 times to ensure chemical homogeneity.

The crystal phases was identified by XRD (XRD, Rigaku D/max-rB) with a Cu Kα radiation and a scanning rate of 2θ step size of 0.02 from 20° to 100°. The microstructure and chemical composition distribution of the polished standard pattern were analyzed using SEM (Hitachi SU5000) equipped with energy dispersive spectroscopy (EDS). Based on two-dimensional micrograph, the volume fraction of crystal phase was analyzed by XRD, Rigaku D/Max-RB equipped with Cu Kα radiation and a scanning range of 2θ step size of 0.02 from 20° to 100°. The microstructure and chemical composition distribution of the polished standard pattern were analyzed using SEM (Hitachi SU5000) equipped with energy dispersive spectroscopy (EDS). Based on two-dimensional micrograph, the volume fraction of crystal phase was analyzed by Image J software [5]. Hardness of standard polishing pattern was determined using an HV-1000 under a loading pressure of 1000 N for 10 s and the average hardness of 25 points were taken in order to ensure the accuracy of the experiment. The height and diameter ratio of standard compression pattern was generally controlled between 1.3 and 2 so as to avoid losing stability in the compression tests, which the cylindrical samples for compression were cut as Φ3 × 6 mm cast rods [20]. The rod-shaped samples at room temperature were tested using an Instron 3382 measuring system with loading rate of 0.1 mm min−1 to acquiring the compression parameters of compressive yield strength, compressive fracture strength and compressive deformation ratio.

The electrochemical performance and potentiodynamic polarization Tafel’ curve of Al0.1CoFeNiCr1−x HEAs was measured using a three-electrode electrochemical workstation (CHI660E) in a 3.5% NaCl solution at room temperature. The three- electrode system was used including the reference electrode is a saturated calomel solution, working electrode is a specimens, and the auxiliary electrode made of a large-area Pt sheet was adopted to cut back the interference of ohmic impedance on the results of test [20–22]. The open circuit voltage (OCP) was first measured so that the electrochemical system was stabilized before each test beginning. The polarization curve of each sample was scanned at a rate of 3 mV s−1 from an initial potential of −1 V to a final potential of 0.4 V.
3. Results and discussion

3.1. XRD analysis

The XRD diffraction pattern of the AlxCoFeNiCr1−x high-entropy alloys are shown in figures 1(a) and (b). With the molar ratios of Al/Cr increase, the crystalline phase of AlxCoFeNiCr1−x HEAs has undergone a complex evolution. The Al0.1Cr0.9, Al0.2Cr0.8, and Al0.3Cr0.7 only have a single FCC phase structure and the Al1.0Cr0 has a single BCC phase structure, while the Al0.4Cr0.6, Al0.5Cr0.5, Al0.6Cr0.4, and Al0.9Cr0.1 consists of dual BCC + FCC phase and the Al0.7Cr0.3, Al0.8Cr0.2 are composed of FCC + BCC/B2 phase. Generally, the diffraction peak of XRD are intense, narrow and sharp representing the degree of crystallization of alloy. The diffraction peak at a larger angle tends to be weak because the increased scanning angle enhances the diffuse reflection in distortion of the lattice of AlCoCrFeNi HEAs [20]. In Al0.4Cr0.6 and Al0.9Cr0.1, the diffraction peaks intensity of XRD of both BCC and FCC phase are not better identified because of low crystalline of the second phase, while the dual-phase structure can be confirmed in figures 2(d) and 2(i). Through analysis of the diffraction peaks of AlxCoCrFeCr1−x HEAs, we can find that, as the molar ratio of Al/Cr increases, the diffraction peaks intensity of BCC/B2 phase enhance meaning the FCC phase' diffraction peaks intensity decline, revealing the volume of the FCC phase is gradually squeezed by the BCC phase, which the result can be proved by seeing from figure 3. In addition, it can be also found from figure 1 that the diffraction peaks of BCC and FCC phases have a slightly shift to a small angle, which can be ascribed to the addition of a large radius of Al promoting the distortion of lattice parameters.

3.2. Microstructure analysis

The backscattered electron morphologies and scanning electron microscopy of AlxCoFeNiCr1−x HEAs are shown in figure 2 and the chemical compositions of crystalline phase was listed in table 1 to discuss the evolution of crystal phase of these alloys as the molar ratio of Al/Cr increases. we can clearly see from figure 2 that the same columnar dendrites single FCC phase exists among the microstructure of Al0.1Cr0.9, Al0.2Cr0.8 and Al0.3Cr0.7, whereas the obvious discrepancy of chemical compositions are presented in these alloys. It can be inferred that although lattice parameters of FCC phase increase with addition of the larger radius of Al, the lattice parameters of dual FCC + BCC phase do not reach the limitation [23]. Shown in table 2 are the chemical mixing enthalpy of elements of the AlxCoFeNiCr1−x HEAs. As already known, the Al–Ni elements could easily cause repulsion by the others because of far more negative mixing enthalpy and lead to form the segregation phase rich in (Al, Ni) [20, 24]. The addition of the molar ratios of Al/Cr, increasing separation intensity of crystal phase, enhance the total mixing enthalpy of AlxCoFeNiCr1−x HEAs. If the thermodynamic entropy has less influences on the total Gibbs free energy than the mixing enthalpy, the segregation of Al–Ni elements will reaches a standard degree, readily causing formation of intermetallic compounds [25].

The analytical results from table 1 show that Al and Ni seem to distribute preferentially in BCC/B2 regions, whereas Cr and Fe likely appear to enter into the FCC regions. In Al0.4Cr0.6, the disordered BCC phase rich in (Al–Ni) previously segregated from the FCC-dominated forming dendritic and dropletshape, which are uniformly distributed in the (Fe, Cr)-rich matrix FCC phase. Although the difference of microscopic morphology exists in Al0.4Cr0.6, the concentrations of constituent elements of both microstructures are consistent in table 1 because of the segregation and growth of precipitated phase during high temperature
furnace cooling process. In addition, we can also find from figure 3 that, with the molar ratio of Al/Cr increase, the enhancing elemental segregation intensity bring about the volume fraction of the (Al, Ni)-rich disordered BCC phase transforming from 1.28% (Al0.4Cr0.6) to 23.38% (Al0.6Cr0.4). By analyzing the table 1 and figure 2, it is learned that the formation of segregated disordered BCC phase requires a large amount to satisfy the energy fluctuations required for the solid phase transition. Therefore, due to the larger interfacial energy between the FCC phase boundary and the dendrites exist in alloy, the increasing of the segregation disordered BCC phase
seem to gather in crystal boundaries and dendrite zones distributing on (Al, Ni)-rich columnar dendritic-FCC matrix phase under a non-consumable vacuum arc melting high cooling process, as shown in figures 2(e) and (f). The number of disordered BCC phase nucleated and the number of disordered BCC phase grew on the matrix FCC phase so that the more grain boundaries appear in the FCC matrix so that the volume fraction of the BCC phase rapidly increase in Al$_{0.6}$Cr$_{0.4}$. Additionally, it can be seen from table 1 that the content of Al and Ni at the boundary of columnar dendritic FCC is significantly higher than that of interdendritic suggesting the grain boundary energy at the edge of the columnar dendritic FCC phase is higher than that of its interdendritic so as to the segregation of disordered BCC phase preferentially precipitates from the boundary of the columnar FCC phase.

Figures 2(f) and (g) shows the SEM images of Al$_{0.6}$Cr$_{0.4}$, Al$_{0.7}$Cr$_{0.3}$HEAs respectively. It can be found that the dominant phase of alloy have slightly change from (Fe, Cr)-rich FCC phase to (Fe, Cr)-rich BCC/B2 phase. By comparing figure 2(f) with figure 2(g), It can be pushed that due to the effects of such repulsion between elements are mutual, the Fe, Cr, elements have highly segregated by repulsion of Al, Ni when the Al content is much higher than the Cr content in Al$_{0.7}$Cr$_{0.3}$. According to figure 2 and table 1, because Fe and Cr elements are repulsed by other elements, a number of (Al, Ni)-rich weave-like FCC precipitate near the grain boundary of matrix disordered BCC phase. The precipitation of (Fe, Cr)-rich FCC phase fencing and (Al, Ni)-rich wrapping the BCC/B2 phase and the BCC phase with dendritic shape is coherently distributed inside FCC phase, while the distribution of relationship of BCC/B2 and weave-like FCC morphology was not reported in previous studies.
Table 1. Phase composition analysis results of AlxCoFeNiCr1-x HEAs by EDS (at%).

| Alloys     | Phases                  | Chemical composition/(at%) |
|------------|-------------------------|----------------------------|
| Al0.1CoFeNiCr0.9 | FCC                     | 1.35 | 24.54 | 23.79 | 23.15 | 27.15 |
| Al0.2CoFeNiCr0.8 | FCC                     | 3.45 | 25.97 | 24.59 | 23.36 | 21.63 |
| Al0.3CoFeNiCr0.7 | FCC                     | 5.11 | 25.93 | 24.89 | 24.88 | 19.20 |
| Al0.4CoFeNiCr0.6 | FCC/DR (Disordered BCC) | 4.24 | 29.52 | 23.70 | 25.14 | 17.90 |
| Al0.5CoFeNiCr0.5 | FCC/DR (Disordered BCC) | 4.04 | 29.44 | 23.14 | 27.86 | 15.53 |
| Al0.6CoFeNiCr0.4 | FCC/ID (BCC)            | 7.86 | 21.16 | 36.18 | 24.12 | 10.68 |
| Al0.7CoFeNiCr0.3 | FCC/DR (Disordered BCC) | 14.21 | 20.85 | 33.31 | 23.60 | 8.07  |
| Al0.8CoFeNiCr0.2 | FCC/ID (BCC)            | 8.65 | 23.77 | 32.18 | 26.21 | 9.19  |
| Al0.9CoFeNiCr0.1 | Ordered B2              | 23.21 | 21.84 | 26.22 | 23.31 | 5.42  |
| Al1.0CoFeNiCr0   | Ordered B2              | 11.75 | 25.66 | 26.79 | 28.64 | 7.17  |
| Al0.1CoFeNiCr0.9 | BCC                     | 3.05 | 30.02 | 23.24 | 27.68 | 14.01 |
| Al0.2CoFeNiCr0.8 | BCC                     | 14.21 | 20.85 | 33.31 | 23.60 | 8.07  |
| Al0.3CoFeNiCr0.7 | BCC                     | 11.75 | 25.66 | 26.79 | 28.64 | 7.17  |
| Al0.4CoFeNiCr0.6 | BCC                     | 4.30 | 32.67 | 22.77 | 28.76 | 11.50 |
| Al0.5CoFeNiCr0.5 | BCC                     | 8.65 | 23.77 | 32.18 | 26.21 | 9.19  |
| Al0.6CoFeNiCr0.4 | BCC                     | 4.30 | 32.67 | 22.77 | 28.76 | 11.50 |
| Al0.7CoFeNiCr0.3 | BCC                     | 8.65 | 23.77 | 32.18 | 26.21 | 9.19  |
| Al0.8CoFeNiCr0.2 | BCC                     | 23.21 | 21.84 | 26.22 | 23.31 | 5.42  |
| Al0.9CoFeNiCr0.1 | BCC                     | 11.75 | 25.66 | 26.79 | 28.64 | 7.17  |
| Al1.0CoFeNiCr0   | BCC                     | 12.27 | 33.66 | 29.47 | 24.58 |      |

Table 2. Mixing enthalpies for atomic pairs between elements (kJ mol⁻¹).

| Element         | Al  | Co  | Fe  | Ni  | Cr  |
|-----------------|-----|-----|-----|-----|-----|
| Al(0.143 nm)    | —   | −19 | −11 | −22 | −10 |
| Co(0.125 nm)    | —   | —   | −1  | 0   | −4  |
| Fe(0.127 nm)    | —   | —   | —   | −2  | −1  |
| Ni(0.125 nm)    | —   | —   | —   | —   | −7  |
| Cr(0.128 nm)    | —   | —   | —   | —   | —   |
Additionally, the analysis of chemical composition shows that the content of Al and Ni elements of dendritic-B2 phase is significantly richer than that of matrix BCC phase. Therefore, we can draw a conclusion that B2 phase primarily form and grow inside the FCC phase with the decrease of temperature. It is generally consider that adding of Al element can driving force for BCC/B2 phase formation, and the reduction of Cr element decrease the volume fraction of the FCC phase [26]. As the molar ratio of Al/Cr increase further, the average size of the disorder BCC phase become larger, the resulting reduction of Cr element induce the displaced of the small FCC phase around the grain boundaries of disordered BCC phase in Al0.1Cr0.9. When the mole ratio of Al/Cr = 0.9:0.1, The aggregation of Fe, Cr will inevitably cause the repulsion of Al and Ni inducing a little FCC phase grows along the grain boundaries, as shown in figure 2(i). When the excessive strong BCC stabilizer adding the Al content completely replaces Cr content, the structure of Al1.0Cr0.0 which has a distinct layered structure, becomes entirely BCC phase without any FCC phase in figure 2(j).

In order to further understand the structural transformations and chemical distribution of BCC/B2 and FCC phase, the EDS mapping result of Al0.6Cr0.4, Al0.7Cr0.3, Al0.8Cr0.2 and Al0.9Cr0.1 are detected. we can seen from figure 4 that, by analyzing the volume fraction of BCC phase rapidly raising from 23.38 wt% (Al0.6Cr0.4) to 95.52 wt% (Al0.9Cr0.1), the larger the BCC phase volume fraction of alloy have, the smaller the FCC phase volume fraction of it possess. It is else visible that the dark region rich in Al and Cr is BCC/B2 phase, the white FCC region rich in Fe and Ni is FCC phase, the Co element has no obvious phase separation, and the transformation of FCC-dominated phase and BCC dominated phase between Al0.6Cr0.4 and Al0.7Cr0.3 can be demonstrated in figures 4(a) and (b).

4. Mechanical properties

4.1. Hardness analysis

In order to verify the effect of different molar Al/Cr ratios on mechanical properties of AlxCoFeNiCr1-x HEAs, both hardness and compression experiments were carried out at room-temperature, the hardness value and compressive performance value are recorded in table 3. With the molar ratio of Al/Cr increase, the hardness of AlxCoFeNiCr1-x HEAs rapidly grows from 135.3 HV (Al0.1Cr0.9) to 486.3 HV (Al0.9Cr0.1) and then reduce to 450.4 HV (Al1.0Cr0). The test result also shows that although the Al0.1Cr0.9, Al0.2Cr0.8 and Al0.3Cr0.7 are mainly composed of single-FCC phases, their hardness gradually increases. Compared with the three alloy’ hardness value, it increased from 135.3 HV to 175.3 HV, Which is mainly relevant to the addition of a larger radius of Al element resulting in the lattice strain energy of the FCC phase increases and make the alloys possess higher the solid solution strengthening [27, 28]. It also suggest that the strengthening effect of the Al solid solution is more higher than softening effect of slightly absence of Cr in the single-FCC phase. What’ more, we known that the FCC phase has more closely packed slip directions than BCC phase so that it have better deformation, whereas the the deformation strength is lower than the BCC phase [29, 30]. Thus, the larger quantities of BCC phase in the AlxCoFeNiCr1-x HEAs are useful to promote the alloys’ hardness. when the volume fraction of the BCC phase increases from 1.28% to 95.52%. We can seen from the table 3 that the hardness of Al0.4Cr0.6 ~ Al0.9Cr0.1 with the FCC and BCC/B2 phase increased from 190.4 HV to 486.3 HV. However, Despite the volume fraction of BCC phase slightly higher than Al0.9Cr0.1. The hardness of Al1.0Cr0 alloy is declined from 486.3 HV to
450.4 HV, which be contributed to the disappearance of the second phase strengthening. It can be deduced that alloys with dual-phase or multi-phase structure universally hold more complex microstructure and higher comprehensive performance compared with that of the single-phase alloys.

Plotted in figure 5 are the hardness value, yield strength, and elongation curves of Al<br>CoFeNiCr<sub>1−x</sub> HEAs to further investigate the effect of the different Al/Cr molar ratio on the mechanical properties. The BCC-dominated phase on the growth rate of hardness is much stronger than that of the FCC-dominant phase and the influence of adding Al, leading to produce a large number of BCC phases, enhance the hardness of the alloy in the current studies. The analysis result shows that the hardness of these alloy is mainly affected by the alteration of the dominant phase besides the volume fraction of the BCC phase.

For the purpose of assisting in analyzing the actual density of Al<br>CoFeNiCr<sub>1−x</sub> HEAs, the equations available to calculate these theoretical densities are

\[
p_i = \frac{\sum_{i=1}^{n} C_i A_i}{\sum_{i=1}^{n} p_i}
\]

The C<sub>i</sub>, A<sub>i</sub>, and P<sub>i</sub> are the concentration, atomic weight and density of elements [31]. Shown in table 3 are the result of theoretical densities and actual density of the Al<br>CoFeNiCr<sub>1−x</sub> HEAs. By comparing, the experimental densities is very close to the theoretical densities.

4.2. Compression analysis

Figures 6(a) and (b) clearly presents engineering stress—strain curves and listed in table 4 are the compressive yield strength (σ<sub>y</sub>), compressive fracture strength (σ<sub>max</sub>) and compressive deformation rate (ε<sub>p</sub>) of the Al<br>CoFeNiCr<sub>1−x</sub> HEAs. In order to distinctly understand about the compression properties, figure 5 provided the yield strength values and plastic strain values curves for us to discuss the influence of different ratio of Al/Cr on compressive behavior of these alloys. As shown in figure 6(a), the FCC-dominant alloy with a excellent plasticity, have reached a compression rate of 40%, which the significant fracture are not observed and their yield strength increased from 212 MPa to 670 MPa as the molar ratios of Al/Cr changed from Al<sub>0.1</sub>Cr<sub>0.9</sub> to Al<sub>0.6</sub>Cr<sub>0.4</sub>, respectively under the compression test process. In addition, it can be found that the addition of Al element result in FCC phase local solute produces pinning dislocation effect improving friction stress, the yield strength of Al<sub>0.1</sub>Cr<sub>0.9</sub>, Al<sub>0.3</sub>Cr<sub>0.7</sub> and Al<sub>0.4</sub>Cr<sub>0.6</sub> with the single-FCC phase are significantly enhanced [32, 33]. Therefore, the Al<sub>0.7</sub>Cr<sub>0.3</sub> has the highest yield strength (300 MPa), followed by the Al<sub>0.5</sub>Cr<sub>0.5</sub>(237 MPa) and the Al<sub>0.6</sub>Cr<sub>0.4</sub> (212 MPa). In previous experiments, the BCC phase is less prone to deformation under the same pressure compared with the FCC phase. the yield strength of alloy is enhanced by raising the volume fraction of BCC phase. When the alloy has more second phase and more grain boundaries, this mean that deformation is difficult to take place in the alloys. The yield strength of Al<sub>0.4</sub>Cr<sub>0.6</sub> is slightly higher than that of Al<sub>0.3</sub>Cr<sub>0.7</sub> to a small Volume fraction of BCC phase(1.28 wt %). Comparing the microstructure of Al<sub>0.4</sub>Cr<sub>0.6</sub> with Al<sub>0.5</sub>Cr<sub>0.5</sub>, Al<sub>0.6</sub>Cr<sub>0.4</sub> the yield strength is significantly improvement after further BCC phase gather at the between boundary and interdendrites of columnar FCC-dendritic phase. Due to this good tissues, with the FCC phase (volume fraction) increase from 1.28% to 23.38%, The yield strength of both Al<sub>0.4</sub>Cr<sub>0.5</sub> and Al<sub>0.6</sub>Cr<sub>0.4</sub>, reaching a higher value of 560 MPa and 670 MPa respectively, is almost twice times that of Al<sub>0.4</sub>Cr<sub>0.5</sub> as presented in table 3.

Compared with Al<sub>0.6</sub>Cr<sub>0.4</sub>, it is obvious that the yield strength of Al<sub>0.7</sub>Cr<sub>0.3</sub> changed from 670 MPa jumps sharply to 1052 MPa followed by plasticity suddenly decrease from 38.9 wt% to 27.1 wt%. This phenomenon can be caused by B2 phase was observed within the microstructure and dominated phase transform from face-centered cubic FCC structure to a volumetric cubic BCC structure. The B2 phase and BCC dominant phase are considered as a high strength and lower plasticity phase [34], which contribute to the dramatically grow compressive strength and the drop of plasticity of Al<sub>0.7</sub>Cr<sub>0.3</sub>. Moreover, with the ratio of Al/Cr increase to 0.8:0.2, the increase of BCC phase (volume fraction) raising to 69.11 wt% may provide effective obstruct slip deformation to further optimizes the overall mechanical properties. It is measured that the Al<sub>0.4</sub>Cr<sub>0.5</sub> has the maximum yield strength (1294 MPa) and fracture strength (1674 MPa) accompanied by the plasticity reduce to 24.8 wt %.

In Al<sub>0.6</sub>Cr<sub>0.1</sub> alloy, in spite of the volume fraction of BCC phase reach the highest value, the comprehensive mechanical properties of Al<sub>0.6</sub>Cr<sub>0.1</sub>, which the yield strength is 1142 Mpa and plasticity is only 9.8%, is lowest than Al<sub>0.4</sub>Cr<sub>0.2</sub>, which may due to the B2 phase and weave-like FCC structure disappear. The conclusion are drawn that the effect of the B2 on strength is much higher than that of the BCC and the plasticity of both Al<sub>0.7</sub>Cr<sub>0.3</sub> and Al<sub>0.8</sub>Cr<sub>0.2</sub> can be improved by weave-like FCC solid solution. While when the Cr element is completely replaced by the Al element, the Al<sub>1.0</sub>Cr<sub>0.1</sub> with the single BCC phase, seem to be completely brittle before reaching the yielding point, which might be induced by layered structure, as shown in figure 2(j).
Table 3. The values of vickers hardness, compressive mechanical properties and actual density of the Al$_x$CoFeNiCr$_{1-x}$ HEAs.

| Alloys          | Hardness/HV | $\sigma_y$/MPa | $\sigma_{\text{max}}$/MPa | $\varepsilon_p$/% | Theoretical densities($p$)/(g cm$^{-3}$) | Actual density/(g cm$^{-3}$) |
|-----------------|-------------|-----------------|-----------------------------|-------------------|------------------------------------------|-------------------------------|
| Al$_{0.1}$CoFeNiCr$_{0.9}$ | 135.3       | 212             | Not fracture                 | $\geq$40          | 8.01                                     | 8.11                          |
| Al$_{0.2}$CoFeNiCr$_{0.8}$ | 147.2       | 237             | Not fracture                 | $\geq$40          | 7.85                                     | 7.82                          |
| Al$_{0.3}$CoFeNiCr$_{0.7}$ | 175.3       | 300             | Not fracture                 | $\geq$40          | 7.68                                     | 7.65                          |
| Al$_{0.4}$CoFeNiCr$_{0.6}$ | 190.4       | 317             | Not fracture                 | $\geq$40          | 7.52                                     | 7.49                          |
| Al$_{0.5}$CoFeNiCr$_{0.5}$ | 210.6       | 560             | Not fracture                 | $\geq$40          | 7.36                                     | 7.33                          |
| Al$_{0.6}$CoFeNiCr$_{0.4}$ | 250.3       | 670             | 1561                         | 38.9              | 7.20                                     | 7.17                          |
| Al$_{0.7}$CoFeNiCr$_{0.3}$ | 314.1       | 1052            | 1512                         | 27.1              | 7.05                                     | 7.04                          |
| Al$_{0.8}$CoFeNiCr$_{0.2}$ | 386.5       | 1294            | 1674                         | 24.8              | 6.90                                     | 6.88                          |
| Al$_{0.9}$CoFeNiCr$_{0.1}$ | 486.3       | 1142            | 1305                         | 9.8               | 6.76                                     | 6.81                          |
| Al$_{1.0}$CoFeNiCr$_{0}$  | 450.4       | 640             | 668                          | 4.6               | 6.61                                     | 6.64                          |
Through the comparative analysis of the XRD, SEM, EDS, figures 5 and 6 results, the conclusion can be drawn as follows: firstly, the strengths of FCC-dominant alloys are generally lower than that of BCC dominant alloys [35], while the plasticity is opposite. Secondly, the appearance of B2 phase of the alloy with dual FCC + BCC phase structure has a significant effect on improving its strength. Thirdly, the proper proportion of Al/Cr can help to improve comprehensive mechanical properties of AlxCoFeNiCr1−x HEAs, yet excessive addition the ratio of Al/Cr will be counterproductive. Additionally, based on the traditional Tabor equation [36], namely HV = ασy, the relationship between hardness and yield strength of AlxCoFeNiCr1−x HEAs were tested. The result of test show that proportional coefficient α of alloy(x ≤ 0.4) with single-FCC phase is about
0.6 and others \((0.5 \leq x \leq 0.9)\) with FCC + BCC/B2 phase has a coefficient of 0.32, which obey the this equation. Since the volume fraction of the second phase of \(\text{Al}_{0.4}\text{Cr}_{0.6}\) and \(\text{Al}_{0.9}\text{Cr}_{0.1}\) is negligible, the proportional coefficient is closer to the matrix phase. However, the alloy \((x = 1.0)\) with single BCC phase has a mistake in the proportional coefficient \(\alpha\) which is owing to it does not reach the yield point so we don’t discussed in this work.

5. Corrosion resistance analysis

Shown in figures 7(a)–(c) are the corrosion polarization curves of \(\text{Al}_{x}\text{CoFeNiCr}_{1-x}\) HEAs used to characterize the corrosion properties of alloys. The corrosion potential \(E_{\text{corr}}\) and the corrosion current density \(I_{\text{corr}}\) are obtained by Tafel’s linear fitting the polarization kinetic curve, as shown in table 4. When the alloy possess the higher corrosion potential and the lower corrosion current density suggesting that it has the greatest corrosion resistance. In contrast, the higher corrosion tendency will be found. It can be known that \(\text{Al}_{x}\text{CoFeNiCr}_{1-x}\) HEAs have more lower corrosion current density in the range of \(7.297 \times 10^{-6} \sim 1.022 \times 10^{-6}\) A cm\(^{-2}\) revealing an excellent corrosion resistance in the 3.5 wt% NaCl solution at room temperature, as listed in table 4. With the molar ratio of of Al/Cr increase, it is observed that the corrosion current density of \(\text{Al}_{0.1}\text{Cr}_{0.9}, \text{Al}_{0.2}\text{Cr}_{0.8}\) and \(\text{Al}_{0.3}\text{Cr}_{0.7}\) with single-FCC phase have slowly increases, which may be mainly explained that addition of a large radius of Al contribute to the lattice strain inside the FCC phase exacerbating the corrosion. In comparison with the single-FCC HEAs, the \(\text{Al}_{0.4}\text{Cr}_{0.6}\), the \(\text{Al}_{0.5}\text{Cr}_{0.5}\) and the \(\text{Al}_{0.6}\text{Cr}_{0.4}\) are composed of the FCC and BCC phase causing by the elemental segregation appears in FCC, meaning that the self-corrosion potential will be increased so as to these alloys are easily corroded [37]. However, the experimental results of table 4 are inconsistent with the above analysis, which can be ascribed to the fact that the segregation BCC phase make the corrosion resistance of these alloys with FCC + BCC phase improve not only making amount of Al, Ni elements rich in BCC phase, but also promoting more Fe, Cr elements gathered in FCC phase. According to the concept of corrosion-resistant of traditional alloy, it is known that the Al, Co, Fe, Ni and Cr elements retain higher self-corrosion resistance in NaCl solution [37, 38]. Therefore, when the weakening effect of the FCC + BCC phase
on the corrosion resistance is the lowest than the enhancement caused by element enrichment, the corrosion resistance of Al$_{0.4}$Cr$_{0.6}$ is more higher than that of the Al$_{0.1}$Cr$_{0.9}$, Al$_{0.2}$Cr$_{0.8}$ and Al$_{0.3}$Cr$_{0.7}$.

Due to the presence of segregation BCC phase in the matrix FCC phase, it is a common phenomenon that the micro-anode region and micro-cathode region are formed in the electrolyte solution, which could accelerate corrosion in the dual phase alloy. In previous studies, we will find that the BCC phase rich Al(Ni) is attacked by Cl$^-$ ion suggesting that the corrosion resistance of the FCC phase is better than that of the BCC phase in the 3.5 wt% NaCl liquid [38]. Thus, the volume fraction of the FCC phase can affect the corrosion resistance of the FCC + BCC phase. From figure 7(b) and table 4, it is observed that the corrosion current density of the Al$_{0.6}$Cr$_{0.4}$, Al$_{0.5}$Cr$_{0.5}$ and Al$_{0.4}$Cr$_{0.6}$ increase from $1.022 \times 10^{-6}$ A cm$^{-2}$, $2.776 \times 10^{-6}$ A cm$^{-2}$, and then to $3.257 \times 10^{-6}$ A cm$^{-2}$ with the segregation BCC phase (volume fraction) enlarges from 1.28% to 23.38%, which displays a high degree of consistency. It is also noticeable that because the Al element is easier to form passive film than Cr element in order that the passivation area prevent the Al$_{0.6}$Cr$_{0.4}$ HEAs from further corrosion in 3.5 wt% NaCl solution [39], the corrosion potential of Al$_{0.6}$Cr$_{0.4}$ is slightly higher than Al$_{0.5}$Cr$_{0.5}$ when Cl$^-$ ion penetrate unrecyclable passivation, the corrosion rate of Al$_{0.6}$Cr$_{0.4}$ HEAs will further increase.

Shown in figure 7(c) are the corrosion polarization curves of Al$_{0.7}$Cr$_{0.3}$, Al$_{0.8}$Cr$_{0.2}$, Al$_{0.9}$Cr$_{0.1}$, and Al$_{1.0}$Cr$_{0}$ HEAs. It is illustrous that the Al$_{0.7}$Cr$_{0.3}$ has the worst corrosion resistance with respect to Al$_{0.8}$Cr$_{0.2}$ and Al$_{0.9}$Cr$_{0.1}$ HEAs, which is possible that the volume fraction of the BCC phase is close to that of the FCC phase leading to the corrosion resistance loss. Owning to BCC is readily attacked by Cl$^-$ ion, the segregated FCC phase as the cathode areas can promote matrix BCC phase corrosion.

The smaller the volume fraction of the FCC phase of alloys have, the higher the corrosion resistance of it retain. Thence, the corrosion resistance order of the three alloys are as follows: Al$_{0.0}$Cr$_{0.1}$ > Al$_{0.8}$Cr$_{0.2}$ > Al$_{0.9}$Cr$_{0.3}$. For Al$_{1.0}$Cr$_{0}$, it can be also noticed that the Al$_{1.0}$Cr$_{0}$ possessing the single BCC structure has the largest self-corrosion current density and lowest self-corrosion potential in comparison with Al$_{x}$CoFeNiCr$_{1-x}$ HEAs, indicating that the it has the greatest corrosion tendency. It can be analyzed from the corrosion polarization curves of Al$_{1.0}$Cr$_{0}$ that the BCC phase was penetrated by Cl$^-$ ion before forming a stable passivation film. Additionally, by comparing the the corrosion resistance of the Al$_{0.1}$Cr$_{0.9}$, Al$_{0.2}$Cr$_{0.8}$ and Al$_{0.3}$Cr$_{0.7}$ from the table 4, the corrosion potential of BCC-dominated phase lower than that of FCC-dominated phase in 3.5 wt% NaCl solution can be also found.

6. Conclusions

(1) By increasing the molar ratio of Al/ Cr in the Al$_x$CoFeNiCr$_{1-x}$ HEAs, the volume fraction of the (Al, Ni)-rich BCC phase increases and the (Fe, Cr)-rich FCC phase decrease. When $x = 0.1, 0.2$ and 0.3, these HEAs have only a single FCC structure, while the others, ($x = 0.4, 0.5, 0.6, 0.7, 0.8$ and 0.9), are composed of dual FCC + BCC or FCC + BCC + FCC/B2 phase. When the Cr element is completely replaced by Al element, the FCC phase disappeared and only BCC phase of the Al$_{1.0}$CoFeNiCr$_{0}$ HEAs was observed.

(2) The result of chemical composition reveals that the Al element is primarily assigned to the BCC phase and the Cr element rich in the FCC phase. With the ratio of the Al/ Cr increases, the matrix phase of alloys shifts from the FCC-dominated phase to BCC-dominated phase, and the elemental segregation switches from (Al, Ni)-rich BCC phase to (Fe, Ni)-rich FCC phase improving the strength of alloys accompanied by a decrease in plasticity.

(3) Through the comparative analysis of the mechanical behavior of Al$_x$CoFeNiCr$_{1-x}$ HEAs, it can be found that the appropriate Al addition and Cr reduction are conducive to helping to improve the comprehensive mechanical properties before reaching the optimal ratio of Al/Cr, like Al$_{0.8}$CoFeNiCr$_{0.2}$, while excessive the ratio of the Al/Cr addition can completely sacrificial alloys’ plasticity, such as the Al$_{0.9}$CoFeNiCr$_{0.1}$ and Al$_{1.0}$CoFeNiCr$_{0}$.

(4) The mechanical properties of Al$_x$CoFeNiCr$_{1-x}$ HEAs depend on the molar ratio of Al/Cr. For $x = 0.7$ and $x = 0.8$, the matrix (Al, Ni)-rich BCC phase maintain primary strength, the harder (Al, Ni)-rich B2 further strengthen, and the weave-like (Fe, Cr)-rich weave-like FCC phase provides the excellent plasticity. Due to this optimal microstructure, the better comprehensive mechanical properties is exhibited in Al$_{0.7}$CoFeNiCr$_{0.3}$ and Al$_{0.8}$CoFeNiCr$_{0.2}$.

(5) Whatever the alloys is composed of single-FCC phase or single-BCC phase in 3.5% NaCl solution, the corrosion resistance of Al$_{0.1}$CoFeNiCr$_{0.9}$, Al$_{0.2}$CoFeNiCr$_{0.8}$, Al$_{0.3}$CoFeNiCr$_{0.7}$, and Al$_{1.0}$CoFeNiCr$_{0}$ have gradually decreases with the molar ratio of the Al/Cr increase suggesting that the negative affect of Cr addition on corrosion resistance cannot completely offset by adding Al in 3.5 wt% NaCl solution.
(6) In the dual phase of the Al\textsubscript{x}CoFeNiCr\textsubscript{1-x} HEAs. The tendency of corrosion current density raise from 1.022 × 10\textsuperscript{-6} A cm\textsuperscript{-2} (Al\textsubscript{0.6}CoFeNiCr\textsubscript{0.4}) to 4.078 × 10\textsuperscript{-6} A cm\textsuperscript{-2} (Al\textsubscript{0.4}CoFeNiCr\textsubscript{0.6}) and then to dropping from 4.078 × 10\textsuperscript{-6} A cm\textsuperscript{-2} (Al\textsubscript{0.7}CoFeNiCr\textsubscript{0.3}) to 2.025 × 10\textsuperscript{-6} A cm\textsuperscript{-2} (Al\textsubscript{0.9}CoFeNiCr\textsubscript{0.1}), which is consistent with the volume fraction of segregation phase. The less the volume fraction of segregation phase of alloys have, the larger the corrosion resistance of it achieve. When the volume fraction of segregation phase is close to the matrix phase, it possess the worst corrosion resistance, like Al\textsubscript{0.5}CoFeNiCr\textsubscript{0.5} HEAs.

(7) According to corrosion current density, it can be found that the order of corrosion resistance is Al\textsubscript{0.6}CoFeNiCr\textsubscript{0.4} > Al\textsubscript{0.9}CoFeNiCr\textsubscript{0.1} > Al\textsubscript{0.7}CoFeNiCr\textsubscript{0.3} > Al\textsubscript{0.5}CoFeNiCr\textsubscript{0.2} > Al\textsubscript{0.6}CoFeNiCr\textsubscript{0.5} > Al\textsubscript{0.3}CoFeNiCr\textsubscript{0.7} > Al\textsubscript{0.7}CoFeNiCr\textsubscript{0.3} > Al\textsubscript{0.5}CoFeNiCr\textsubscript{0.1} implying that the corrosion resistance of alloy is related to the element distributions of crystal phase and the microstructure. Therefore, adding the proper molar ratio of Al/ Cr on corrosion resistance is worthy further investigating for us so that AlCoCrNiFe HEAs system is rapidly used for impractical applications.

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ORCID iDs

Shuai Wang @ https://orcid.org/0000-0002-5064-5276

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