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Correlation Between Structure, Microstructure, and Magnetic Properties of AlCoCrFeNi High-Entropy Alloy

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Abstract: This study explores the crystal structure, microstructure and magnetic phase evolution of the AlCoCrFeNi high-entropy alloy (HEA), highlighting its potential for applications requiring tailored magnetic properties across diverse temperatures. Electron microscopy and X-ray diffraction revealed that the as-cast alloy's microstructure comprises equiaxed grains with branching dendrites, showing compositional variations between interdendritic regions enriched in Al and Ni. Temperature-induced phase transformations were observed above room temperature, transitioning from body centered cubic (BCC) phases (A2 and B2) to a predominant FCC phase at higher temperatures, followed by recrystallization of the A2 phase upon cooling. Magnetization measurements showed a drop near 380 K, suggesting the Curie temperature of BCC phases, a peak at 830 K attributed to optimal magnetic alignment in the FCC phase, and a sharp decline at 950 K marking the transition to a paramagnetic state. Magnetic moment calculations provided insights into magnetic alignment dynamics, while low-temperature analysis highlighted the alloy's magnetically soft nature, dominated by ferromagnetic contributions from the A2 phase. These findings underscore the strong interdependence of microstructural features and magnetic behavior, offering a foundation for optimizing HEAs for temperature-sensitive scientific and industrial applications.

Keywords: high entropy alloys; field cooling-zero field cooling; microstructure



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1. Introduction

High-entropy alloys (HEAs) represent a novel class of materials characterized by their multi-component composition, where each element is present in roughly equal atomic proportions. Unlike traditional alloys, which are typically based on one or two elements, HEAs consist of five or more elements. This distinctive feature leads to high configurational entropy, which stabilizes solid solution phases (such as face cubic centered, FCC, or body cubic centered, BCC) and imparts a range of exceptional combinations of mechanical strength, ductility, thermal stability, and corrosion resistance [1–5].

Recently, interest in HEAs has extended beyond mechanical performance to include their magnetic properties, especially in transition-metal-based systems such as AlCoCrFeNi [2,6,7]. These materials are increasingly investigated as potential candidates for soft magnetic applications, magnetic sensors, and even spintronic devices [8–10]. The

underlying mechanisms driving magnetic ordering in HEAs, however, remain a subject of ongoing debate. Some researchers attribute soft-magnetic behavior and low coercivity to reduced magnetocrystalline anisotropy due to severe lattice distortion and atomic-scale disorder [6,7,11]. Others emphasize the role of local chemical environments and d-band electron interactions in determining spin alignment and magnetic phase transitions [12].

Moreover, both intrinsic properties (e.g., magnetic moment, Curie temperature) and extrinsic factors (e.g., grain size, phase distribution, synthesis method) significantly affect the magnetic performance of HEAs [7,10,13–15]. For example, Ma et al. [10] demonstrated that nanoscale control in HEA fibers can lead to excellent magnetic softness and mechanical flexibility, while Wang et al. [15] showed that Al addition and nanocrystallinity can tune coercivity and phase composition. A comprehension of the mechanisms underlying these structural alterations will enable the modification of the characteristics of these systems for the development of novel functional materials.

The random distribution of multiple elements can lead to complex magnetic interactions and unique magnetic behaviors. Their ability to maintain stable magnetic states under varying conditions can enhance the performance and reliability of magnetic storage devices [8,11,14]. On the other hand, HEAs with high spin polarization are promising for spintronic applications; for instance, one study explores how permeability modulation in HEAs can enhance spin arrangements, thereby improving the efficiency of water oxidation reactions. The research highlights the potential of HEAs in spintronic applications due to their strong d-d Coulomb interactions and excellent soft-magnetic properties [9].

Despite the promising attributes of equiatomic alloys, there remain significant gaps in our understanding of their physical properties. The structural and microstructural characteristics of equiatomic alloys are of paramount importance as they directly influence their mechanical performance and overall stability. X-ray diffraction (XRD) and Rietveld refinement techniques are essential for determining the crystal structure and phase composition of these alloys at different temperatures. This paper aims to provide a comprehensive investigation of the structural, microstructural and magnetic properties of equiatomic Al-CoCrFeNi alloys. By employing a combination of experimental techniques and theoretical modeling, we seek to elucidate the underlying mechanisms that govern these properties and explore their potential applications.

2. Results and Discussion

2.1. Microstructure of as Cast Alloy

Figure 1a shows a representative backscattered electron image for the as cast Al-CoCrFeNi high-entropy alloy, obtained via field emission scanning electron microscopy (FE-SEM) at low magnification. With a higher atomic number, there is a greater emission of backscattered electrons, therefore, areas with heavier elements appear brighter in the image. Images taken at room temperature show a microstructure composed of equiaxed grains of about 80 μm ; inside the grains, intricate, branching dendrites are present. The dendrites span $\sim 70 \mu\text{m}$ in length, and the arm thickness measures $\sim 20 \mu\text{m}$. Lower secondary electron (LEI) micrographs of the dendrites, red area, reveals the presence of nanometric precipitates (Figure 1b), whereas the interdendritic area, depicted in blue, displays a periodic labyrinth-like pattern (Figure 1c), similar to those reported by other authors [7,16,17].

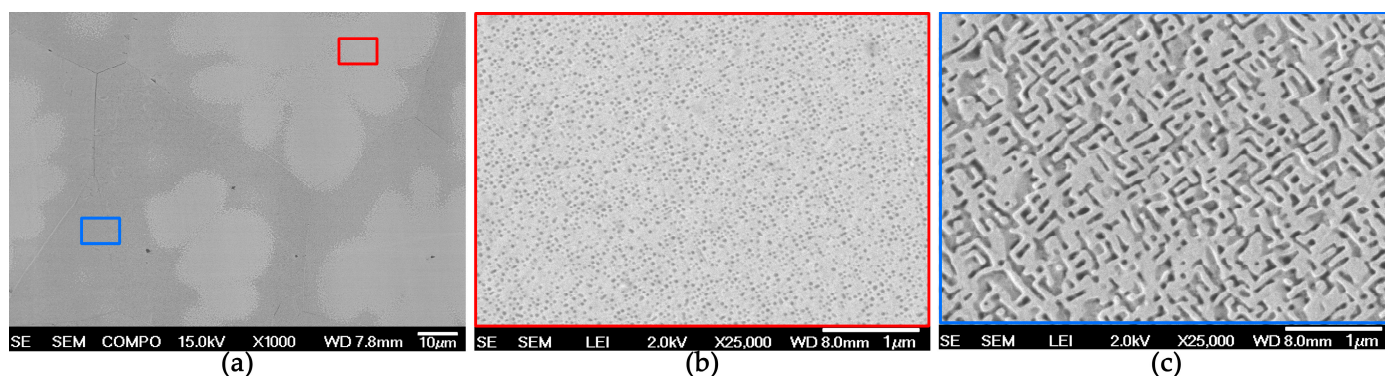


Figure 1. (a) Backscattered electrons micrograph of a representative area of the as cast HEA, (b) lower secondary electron (LEI) of the dendrites, (c) LEI micrograph of the interdendritic area.

A chemical composition analysis using energy-dispersive X-ray spectroscopy (EDS) reveals compositional fluctuations between the two regions. Multiple analyses were conducted in each zone to obtain average atomic percentage values, as presented in Table 1, while ensuring consistent experimental conditions: magnification ($2000\times$), accelerating voltage (15 kV), analyzed areas ($3 \times 3 \mu\text{m}$), and data acquisition time (30 s). The interdendritic area shows enrichment in Al and Ni, accompanied by a significant decrease in Cr and, to a lesser extent, Fe. In contrast, the region containing nanometric precipitates retains a composition similar to that of the overall alloy.

Table 1. Chemical composition analysis by X-ray spectroscopy (EDS).

Area	Al (at. %)	Co (at. %)	Cr (at. %)	Fe (at. %)	Ni (at. %)
Overall	32 ± 0.7	18 ± 1.3	10 ± 0.8	16 ± 0.6	24 ± 2
Dendrites	31 ± 1	18 ± 1.5	14 ± 0.6	16 ± 0.5	21 ± 2.4
Interdendritic area	35 ± 0.3	18 ± 0.5	5 ± 0.5	14 ± 0.4	27 ± 1

Additionally, elemental mapping by EDS confirmed the distribution of elements in the two distinct microstructures. One is rich in Al and Ni but has a low content of Cr and Fe, while the other exhibits a higher concentration of Cr and Fe with a lower proportion of Al and Ni. In both regions, Co is uniformly distributed.

X-ray diffraction patterns of the as cast sample were measured at different temperatures up to 973 K and are shown in Figure 2; patterns for 773 K, 873 K, and 973 K temperatures are graphically excluded since no structural changes were identified. The patterns were measured in vacuum, using a heating ramp of 10 K/min with a waiting time of 5 min upon reaching the desired temperature before starting the measurement. For the as cast sample measured at room temperature (named AC), two body centered cubic (BCC) phases were identified, namely ordered B2 (space group 221, Pm-3m) and a disordered A2 (space group 229, Im-3m). After increasing the temperature to 443 K, the presence of both BCC phases is reduced (see reflection intensities decreasing) and the formation of a face cubic centered (FCC) phase (space group 225, Fm-3m) takes place. When the temperature reached 673 K, A2 and B2 phases disappeared completely, leaving only the FCC phase. This phase remained until the maximum applied temperature (973 K).

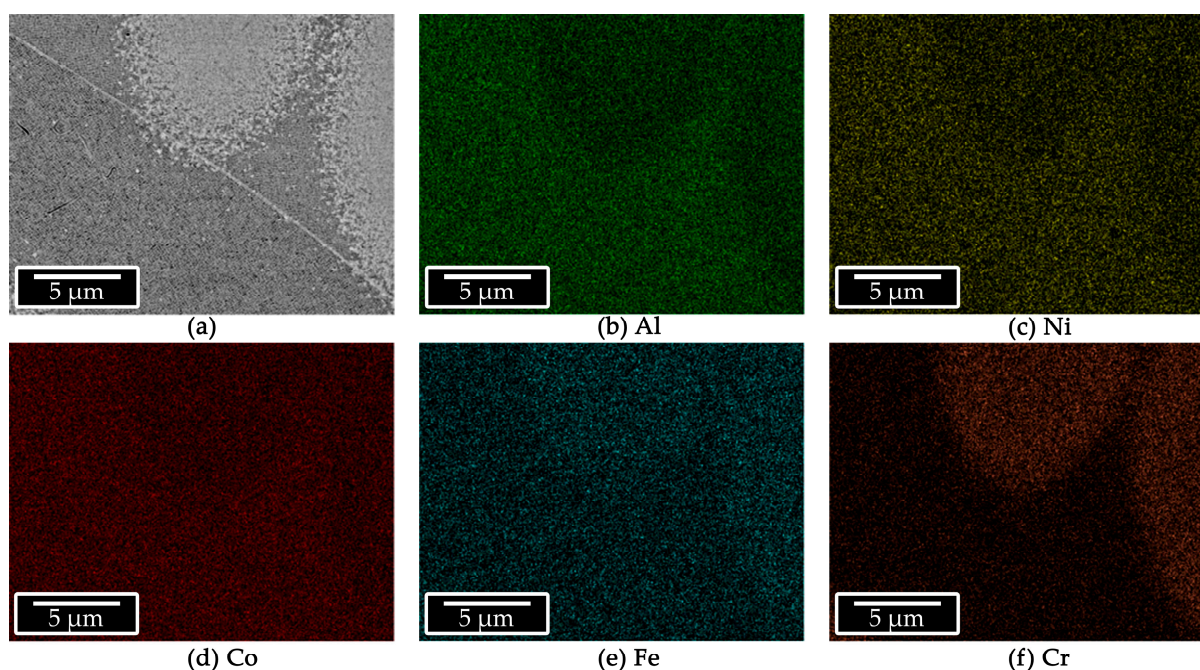


Figure 2. EDS elemental mapping of the HEA as cast alloy. (a) Backscattered electrons micrograph of a representative area; (b–f) EDS maps of Al, Ni, Co, Fe, and Cr.

This phases evolution occurred due an increase in the kinetic energy of the atoms within the crystal structure, in other words, the atoms are vibrating with a higher intensity in comparison with the vibrations produced at room temperature. If the atomic vibrations are strong enough, the forces that maintain the atomic bonds in the crystal structure can be overcome allowing them to reorganize into other crystalline structures [18]. Finally, the sample was cooled to room temperature and measured again (named F), and the X-ray pattern indicated that the FCC phase disappeared and a pure A2 phase reformed. A change in the (211) peak intensity can be noticed for the final measurement; this could be an indication of an apparent preferential orientation in that direction.

The texture coefficient (TC) is a commonly used parameter for quantitatively evaluating the preferred crystal orientation. The reflection intensities from each XRD pattern contain information related to the preferential growth of phases in polycrystalline material. For crystal planes (hkl), TC can be defined as follows:

$$TC_{(hkl)} = \frac{\frac{I_{(hkl)}}{I_0(hkl)}}{n^{-1} \sum_n \frac{I_{(hkl)}}{I_0(hkl)}}, \quad (1)$$

where $I_{(hkl)}$ is the measured relative intensity obtained from XRD, $I_0(hkl)$ is the standard intensity taken from ICSD 102751, and n is the number of diffraction peaks considered [19,20]. For the AC sample, TC calculated along (211) reflection is 1.01, indicating that the sample comprises randomly oriented grains similar to the information reflected by the ICSD. After all the heating/cooling process, for the same reflection $TC = 2.08$, which is more than a 200% improvement for the texture coefficient and denotes that (211) crystal plane indeed is the preferred orientation.

The phase weight fraction and lattice parameters of the indexed phases were determined using Rietveld refinement with the peak-profile pseudo-Voigt (pV) function in Fullprof Suite [21]. The refinement strategy followed in this work is the same used in previous works [22], which helps to reduce convergence errors in the Rietveld refinement method. The models used as initial solution for the refinement were ICSD 102751,

ICSD 608802 and ICSD 41506 for the A2, B2 and FCC phases, respectively [23–25]. Also, the volume-average crystallite size and microstrains values were calculated via Thompson Cox Hastings method (TCH-pV function) implemented in Fullprof Suite that is useful to determinate the microstrains and volume-average apparent crystallite size contribution on the peaks broadening. In the pV function, a total peak broadening and a shape parameter are approximated to indicate the contributions from Gaussian and Lorentzian line-shape fit, while the TCH-pV function acts in the inverse way (approximating the contributions from Gaussian and Lorentzian broadening to calculate the total peak broadening and the shape parameter) [26]. Additionally, according to the Fullprof Suite Manual (Res = 1), the instrumental resolution function, for a constant wavelength λ , was obtained by measuring the peak profiles of a well crystallized Si powder diffraction pattern.

The TCH-pV function is defined by two functions that approximate the peak shape (Gaussian and Lorentzian). Each of them gives a contribution to the broadening of the peak caused by the microstrains and volume-average apparent size which are defined as a change to the full width at half maximum (FWHM). Moreover, the broadening contributions can be calculated by two different approximations (isotropic and anisotropic). The isotropic approximation establishes that each reflection in the XRD pattern has an equal FWHM value, i.e., an average among all the reflections. In other hand, the anisotropic approximation suppose that each reflection has a different FWHM value (which is more accurate to experimental measurements). Hence, the anisotropic approximation was used in the actual work and the broadening equations can be defined in their reduced form as:

$$FWHM^2 \left(\text{Gaussian} - \frac{\text{strain}}{\text{size}} \right) = H_G^2 = \left[(1 - \xi)^2 DST^2 \right] \tan^2 \theta + \frac{I_G}{\cos^2 \theta}, \quad (2)$$

$$FWHM \left(\text{Lorentzian} - \frac{\text{strain}}{\text{size}} \right) = H_L = [\xi DST] \tan \theta + \frac{[F(S_Z)]}{\cos \theta}, \quad (3)$$

where ξ is the Lorentzian mixing parameter (represents the Lorentzian weight of the strain-broadened profile), DST is the anisotropic model function of the strain, I_G is the Gaussian mixing parameter and $F(S_Z)$ is the anisotropic model function of the apparent crystallite size. In the DST function there are fifteen terms, of which only S_{400} and S_{220} take values due to the symmetry constraints for the Laue class m-3m [27,28]. On other hand, $F(S_Z)$ function, for symmetry constraints, contains five spherical harmonics symmetrized, and only K_{00} , K_{41} , K_{61} and K_{81} take values [29,30]. The Lorentzian and Gaussian mixing parameters, ξ and I_G , are to improve the quality of the fit and, therefore, to obtain more accurate values of the average crystallite size and microstrains, respectively [26,28]. The complete TCH-pV function discussion for cubic crystalline structures has already been explained elsewhere [31]. Volume average crystallite size and microstrains were calculated only for the phase with the highest weight fraction at each pattern. The standard deviations of the global average strain ($DA_{(S)}$) and apparent crystallite size ($DA_{(C)}$), derived from the reciprocal lattice, are considered indicators of anisotropy.

The Rietveld refinement for the as-cast alloy, and for the 443 K, 673 K, and 298 K (F) temperatures are also shown in Figure 3. Moreover, calculated structural and microstructural parameters, the weight fraction of identified phases and agreement factors of the refinement are shown in Table 2.

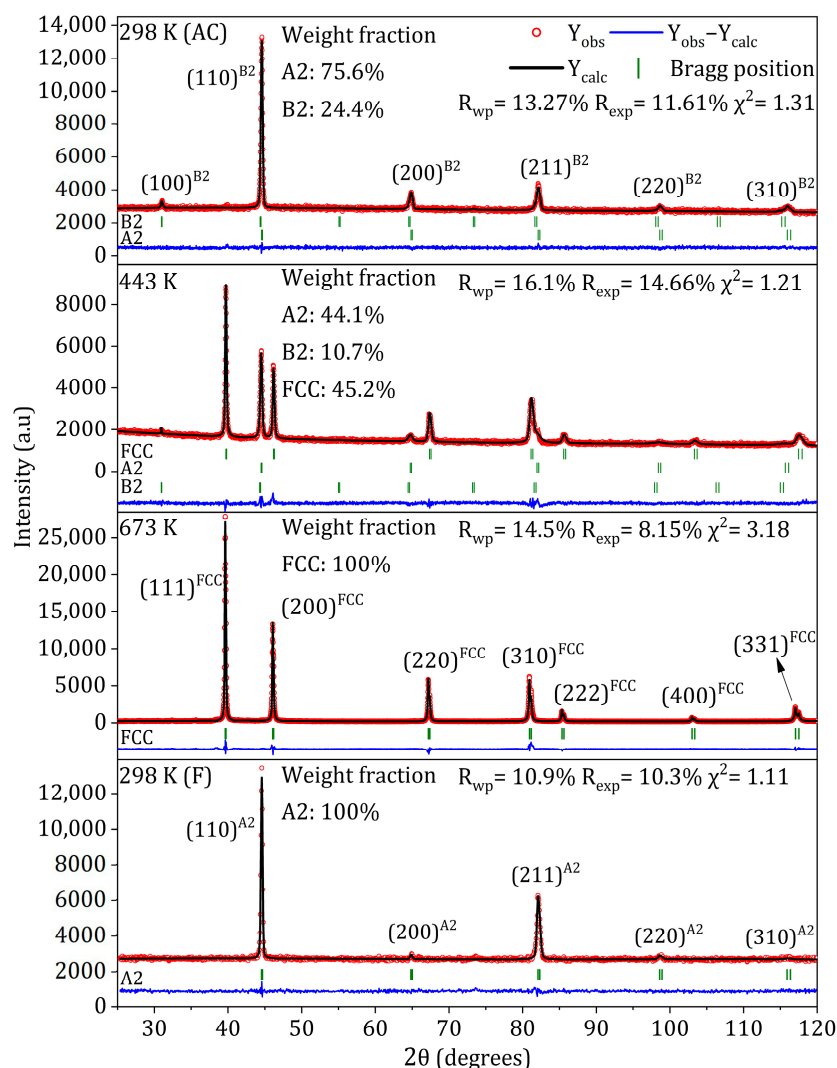


Figure 3. XRD patterns and structural refinements for the as-cast alloy, and for the 443 K, 673 K, and 298 K (F) temperatures; the different observed cubic structures are identified.

For the as-cast alloy, there is a higher weight fraction for the A2 phase (75.6%) against B2 phase (24.4%); as the temperature increased and up to 443 K, both phases reduced and the FCC phase became predominant in the system (45.2%). As we mentioned before, the system at higher temperatures is completely characterized by FCC phase. The lattice parameters in all phases, as expected, increase due to cell distortion suffered at heating, reaching a maximum at 973 K. Regarding the size of the crystallites, they first decreased when the temperature increased to 443 K, due the A2 phase being reduced by the formation of the FCC phase. After this temperature, analogously to the lattice parameters, crystallite size increased with temperature increment. After cooling to room temperature, there is only A2 phase present, the crystallite size value decreased to a 33.50 nm, lower than the initial value in the as-cast state (45.86 nm), this behavior can be attributed to the recrystallization of A2 phase from FCC phase.

An easy way to understand or visualize DA (regardless of whether it is crystallite size or microstrains), is when $DA = 0$: the shape of the average crystallite or the microstrains is a perfect sphere, i.e., homogeneous in all directions, unlike when it has a high value, the shape is more heterogeneous in different directions. An example in this case is for the microstrains for $443 \text{ K} \leq \text{Temperature} \leq 973 \text{ K}$ range, where the microstrains value is reduced due the crystallite size increment and, hence $DA_{(S)} \approx 0$, this indicates that the microstrains value has been reduced so much that the apparent influence in which it affects

the crystal lattice can be defined as homogeneous (in the same way and intensity in all the directions). In other hand, the $DA_{(C)}$ decreased when the temperature increment from 298 K to 443 K, this is attributed to the growth of the new FCC phase which has a lower crystallite size (32.65 nm) than the A2 phase in 298 K. If the temperature rise from 443 K to 973 K, the crystallite size of the new predominant FCC phase is positively influenced. Moreover, in this range of values, any variation in size is correlated as a huge quantification in $DA_{(C)}$ reaching a maximum of 505.26 at 973 K in comparison with the lowest value of 87.32 at 443 K.

Table 2. Calculated lattice parameters, weight fraction for each phase, average-volume crystallite size and microstrains for all XRD patterns.

Temperature	298 K (AC) *	443 K	673 K	773 K	873 K	973 K	298 K (F) *
Structure parameters							
a (nm)							
A2	2.8767 (3)	2.8804 (3)	—	—	—	—	2.87362 (13)
B2	2.87310 (19)	2.8934 (4)	—	—	—	—	—
FCC	—	3.92928 (15)	3.93671 (10)	3.94105 (10)	3.94518 (12)	3.94939 (12)	—
Weight fraction							
A2	0.756 (3)	0.441 (7)	—	—	—	—	1.00
B2	0.244 (2)	0.107 (9)	—	—	—	—	—
FCC	—	0.452 (9)	1.00	1.00	1.00	1.00	—
Microstructure parameters							
Crystallite size (nm)	40.82	32.65	80.16	127.24	198.24	231.11	33.50
$DA_{(C)}$	227.38	87.32	151.49	190.70	399.48	505.26	267.88
Maximum strain (10^{-4})	21.81	36.51	11.57	6.03	4.28	3.63	36.05
$DA_{(S)}$	11.58	1.39	0.80	0.49	0.31	0.17	5.69
Agreement factors							
R_{wp} (%)	13.27	16.1	14.5	16.2	13.4	11.8	10.9
R_{exp} (%)	11.61	14.66	8.15	8.88	6.45	5.44	10.3
χ^2	1.31	1.21	3.18	3.33	4.32	4.68	1.11

* For 298 K (AC) and 298 K (F) temperatures, the crystallite, and microstrains were calculated for A2 phase, while for the rest of temperatures, were calculated for FCC phase.

In Figure 4, the spherical harmonics projections of the average apparent crystallite size, using the Fourier program GFourier incorporated in Fullprof Suite software, v. 8.20, for 298 K (AC) and 298 K (F) temperatures in the crystallographic plane (001), are shown.

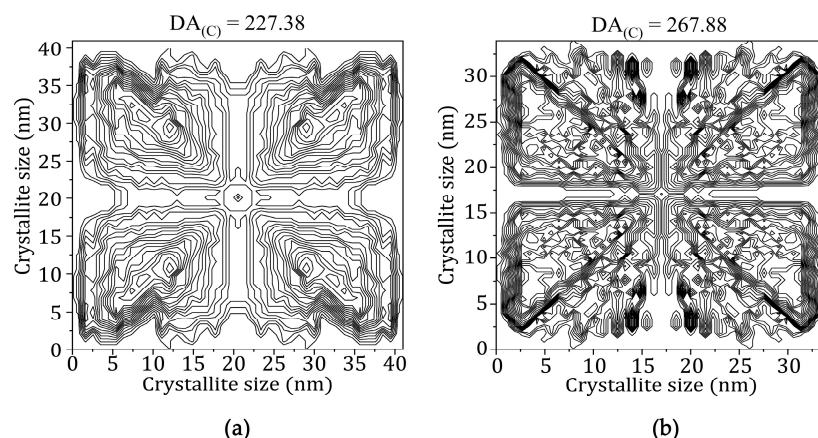


Figure 4. The spherical harmonics projections of three-dimensional bodies representing average apparent size for (a), 298 K (AC) and (b) 298 K (F) temperatures in crystallographic plane (001).

An important fact is that for Laue class $m\bar{3}m$ all spherical harmonics projections on crystal axes are equal. The crystallite size reduction (from 40.82 nm to 33.50 nm) is due to the reformation of A2 phase from pure FCC phase, the general shape in both figures is pretty similar, but the enhancement in DA(C) to 267.88 is attributed to the texture development (Figure 4b) which can be seen in the black lines agglomerated (a crystallite surface relief) in the [110] and [210] directions in the 2D plane, which agrees with the calculated TC.

2.2. Magnetic Properties

Several magnetization curves were measured at different temperatures below RT, with a maximum applied magnetic field of 3 MA/m (Figure 5a). At room temperature the magnetization at the highest applied field was 442 kA/m, while at 3 K the maximum magnetization was 540 kA/m. The sample is magnetically soft, with very low remanence and coercivity. Similar studies have shown that the magnetic properties of HEAs can vary depending on their microstructure and composition, but generally they exhibit soft magnetic behavior [7,12]. As expected, the saturation magnetization decreases with increasing temperature. The shape of the hysteresis loops suggest that the sample behaves as superparamagnetic.

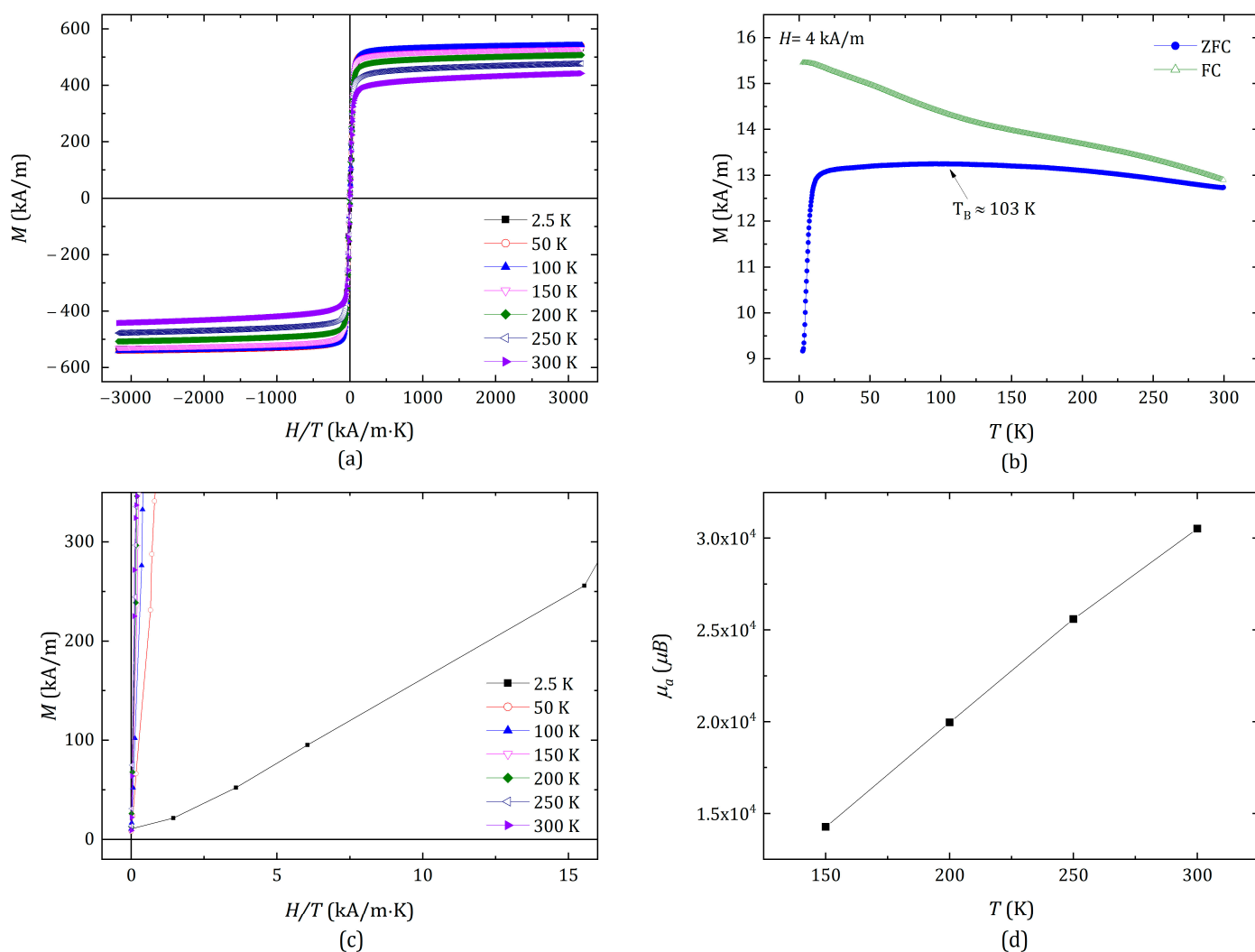


Figure 5. (a) Magnetization (M) vs. magnetic field (H) of the HEA measured from 2.5 to 300 K; (b) M vs. H/T plots corresponding to the Langevin expression; (c) apparent magnetic moment calculated from the fitting of the Langevin function to the M vs. H/T plots; (d) M vs. T plots measured in the FC and ZFC regimen, from 2.5 to 300 K; blocking temperature (T_B) is indicated.

In Figure 5b, field cooling (FC) and zero field cooling (ZFC) plots at temperatures below RT are shown. In ZFC, the initial low magnetization indicates random alignment of magnetic domains at low temperatures. The rapid increase to a peak reflects the alignment of magnetic moments as thermal energy allows them to overcome energy barriers, reaching optimal alignment where M is maximum, around 103 K; this could be considered the blocking temperature (T_B). The gradual decrease after T_B suggests that higher temperatures disrupt magnetic alignment due to increased thermal agitation. For FC, during cooling magnetic domains aligned with the direction of the applied magnetic field. Then, as the temperature was increased, the magnetization decreased because the thermal energy that disrupts the alignment of magnetic moments was increased.

Anhyseretic curves of magnetic materials that exhibit superparamagnetic characteristics, can be described as a superposition of a set of Langevin functions (Equation (4)).

$$M(H, T) = M_s \left(\coth \frac{\mu H}{k_B T} - \frac{k_B T}{\mu H} \right), \quad (4)$$

where μ is the mean moment of the magnetic domains, k_B is the Boltzmann constant, and M_s is the saturation of magnetization [32].

However, the standard scaling law, which is expected when plotting the magnetization as a function of the H/T ratio (Figure 5c), is only approximately observed; this could be an indication of more complex interactions beyond simple thermal activation. Straight lines at temperatures from 150 K and higher can be observed, corresponding to the Langevin expression. All the lines show identical slope, which is consistent with the superparamagnetic nature of the sample in this temperature range. However, the difference in the slope of the curves measured at temperatures lower than the blocking temperature is noticeable and consistent with ferromagnetic behavior.

The fitting of Equation (4) to the anhyseretic superparamagnetic curves yielded magnetic moments, as shown in Figure 5d. These moments exhibit an unusual linear increase with temperature. While this trend might suggest an increase in saturation magnetization, this is not physically accurate, as M_s typically decreases with temperature. The extracted moment values, therefore, do not represent actual domain moments and are instead referred to as apparent moments (μ_a). Previous research suggests that in granular systems where direct particle contact is minimal, short-range exchange interactions are weak, leaving long-range dipolar interactions as the dominant force [33].

Although dipolar interactions can influence the collective alignment of moments, they are insufficient to sustain long-range magnetic order. This interaction framework may contribute to the deviations from the expected scaling behavior and the unexpected temperature dependence of μ_a . Furthermore, fitting the coercivity suppression law yielded a suppression exponent of $\delta \sim 0.35\text{--}0.45$, aligning with theoretical values for dipolar-interaction dominated systems [34,35].

Additionally, dendritic connectivity plays a critical role in stabilizing domain interactions through extended pinning networks. The presence of nanometric precipitates within dendrites acts as localized pinning centers, restricting domain wall mobility and influencing hysteresis loop shape. This suggests that, while thermal activation drives the moment scaling behavior, residual anisotropy effects persist beyond the blocking temperature, contributing to the stabilization of collective dipolar interactions even in the anhyseretic regime.

The evolution of the magnetic phases of the sample over a temperature range from RT to 995 K can be observed in the M vs. T plot (Figure 6a), measured under an applied magnetic field of 4 kA/m. Additionally, the first derivative of magnetization with temperature (dM/dT) is shown, providing further insight into the temperature-induced transitions. A

notable decrease in magnetization around 380 K suggests that thermal agitation weakens the magnetic alignment within the BCC phases (A2 and B2). Previous studies have reported Curie temperatures (T_C) for BCC high-entropy alloys near this value [12,36], supporting the interpretation that this transition is associated with the loss of ferromagnetic ordering.

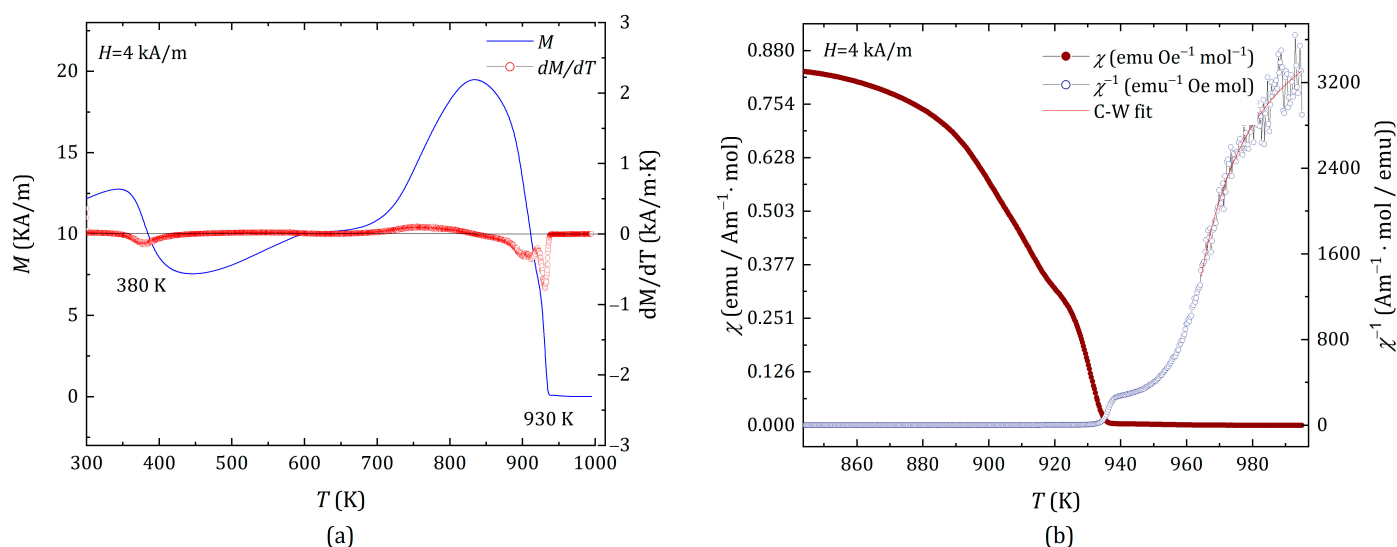


Figure 6. (a) Magnetization (M) vs. temperature (T) plot of the HEA measured from 300 to 1000 K. The derivative of M with T is also shown; (b) M vs. T plot measured from 2.5 to 300 K, blocking temperature (T_B) is indicated.

An increment of magnetization can be noted after 443 K. This can be attributed to the structural variations that happened. From our X-ray diffraction analysis was confirmed that at 443 K, the ordered B2 phase disappears, while the disordered A2 phase remains, accompanied by the formation of the FCC phase. Typically, the ordered B2 phase exhibits lower magnetic moments and can be paramagnetic or weakly ferromagnetic. This means it contributes less to the overall magnetization of the material, while the disordered A2 phase usually exhibits higher magnetic moments and strong ferromagnetic behavior, contributing significantly to the overall magnetization.

As temperature keeps increasing, the FCC phase stabilizes, and by 673 K and above, it remains the dominant structural phase. The increase of magnetization above 673 K, to a maximum at 830 K can no longer be explained by structural changes alone, since FCC structure remains up to the maximum temperature of measurement.

While the magnetic transitions observed in this study align with prior phase evolution models, compositional gradients may also play a crucial role in modifying local magnetization behavior. At elevated temperatures, vacancy formation increases, facilitating atomic redistribution, particularly for elements such as Fe, Co, and Ni. This diffusion influences local magnetic interactions, enhancing short-range exchange coupling and leading to a temporary increase in magnetization. Studies on FCC Fe-Ni alloys indicate that magne-tochemical effects govern vacancy formation energy, thereby affecting atomic mobility and driving compositional fluctuations within the material [37]. These redistributions can locally reinforce ferromagnetic coupling, contributing to the observed peak in magnetization at 830 K. Given that the FCC structure remains stable throughout this temperature range, structural transformations alone cannot fully account for this trend, suggesting that vacancy-mediated atomic reorganization plays a key role in the enhanced moment alignment. Experimental and theoretical investigations further support the impact of vacancy-driven magnetization shifts, demonstrating that high-temperature vacancy behav-

ior is closely linked to charge transfer mechanisms and variations in exchange coupling within complex alloys [38].

As temperature approaches 930 K, a sharp decline in magnetization signals the onset of the paramagnetic state, marking the experimentally observed Curie temperature (T_C). To further investigate the transition to the paramagnetic state, we analyzed high-temperature susceptibility using the modified Curie–Weiss law [39] in the temperature range 964–1000 K:

$$\chi = \chi_0 + \frac{C}{T - \theta_{CW}}, \quad (5)$$

where χ is the magnetic susceptibility, C is the Curie constant, θ_{CW} is the Curie–Weiss temperature and χ_0 is the temperature independent susceptibility, which causes the fit of the $1/\chi$ vs. T plot (Figure 6b) to deviate from a linear fit. The fitted θ_{CW} was 958 K, which could be considered the paramagnetic Curie temperature and aligns well with the observed Curie temperature of 930 K.

The effective magnetic moment, an intrinsic magnetic property of the material, can be derived from the Curie–Weiss law by the relation:

$$\mu_{eff} = \sqrt{\frac{3k_B C}{N_A}}, \quad (6)$$

where k_B and N_A are the Boltzmann constant and the Avogadro's number, respectively [40]. The effective magnetic moment was found to be $1.35 \mu_B$, a value consistent with similar high-entropy alloys [41].

Finally, the component of μ_{eff} along the applied magnetic field, namely μ_H , can be derived from the Langevin fitting through the equation:

$$\mu_H = \sigma_0 \frac{A}{N_A}, \quad (7)$$

where σ_0 is the saturation magnetization per unit mass and A is the molar weight of the sample [42]. The calculated μ_H was $0.646 \mu_B$ at 2.5 K and $0.517 \mu_B$ at 300 K; these values reflect the progressive weakening of exchange interactions as thermal agitation increases, leading to a diminished ability of individual moments to align with the applied field. Assuming that only BCC structures are found below room temperature, and since these calculations were performed within this temperature range, they reflect the magnetization behavior of the A2/B2 phases and should not be directly extrapolated to the FCC phase.

The calculated effective magnetic moment represents an intrinsic property of the alloy, providing valuable insight into its magnetic behavior. While these findings are specific to the composition studied, the methodology used can be broadly applied to other high-entropy alloys (HEAs), helping researchers assess phase-magnetic correlations in different systems. The wide temperature range explored in this work enhances our understanding of moment evolution, offering a framework that may inspire further investigations into anisotropy suppression and interaction-driven phenomena in complex alloys. These findings highlight the complex interplay between phase stability, dipolar interactions, and exchange coupling, shaping the temperature evolution of magnetization. Future experimental studies incorporating neutron scattering or first-principles calculations could help refine the role of moment scaling mechanisms in high-entropy alloys. While synthesis effects such as phase stability and microstructural evolution were not the primary focus, future studies incorporating these variables could refine and expand the applicability of this approach, making it an even more powerful tool for materials characterization. Furthermore, this methodology offers a foundation for optimizing HEAs for temperature-sensitive sci-

tific and industrial applications, enabling the design of advanced materials with tailored magnetic properties.

3. Materials and Methods

Ingots of the AlCoCrFeNi alloy were arc melted using high-purity elements (>99.9%) in a water-cooled copper crucible under an argon atmosphere. The process was conducted using a Series 5SA single-arc furnace from Centorr Vacuum Industries (Nashua, NH, USA). While this furnace does not have a built-in temperature measurement system, a separate experiment using an infrared temperature detector confirmed that the typical melting temperature reached approximately 2000 K. To ensure homogeneity, the ingots were flipped and re-melted five times. Each melting cycle lasted about 30 s, allowing sufficient mixing of the elements. Following the final melting step, the alloy was left to cool within the same copper crucible for approximately 180 s, until it reached room temperature.

Specimens were cut, grinded and polished to obtain a smooth surface and X-ray diffraction was performed using a PANalytical X'Pert PRO MPD diffractometer with an X'Celerator detector, using Cu K α radiation. Patterns were measured for all samples from 25–120° with a step of 0.017° and 120 s per step in Bragg–Brentano geometry. Rietveld refinement and the microstructure study was carried out with the Fullprof Software [21].

Scanning electron microscopy images of oxalic acid etched samples were obtained with a JEOL JSM-7401F field emission microscope (JEOL, Akishima, Tokyo, Japan) and the composition was measured by energy-dispersive X-ray spectroscopy (EDAX, Warrendale, PA, USA).

Magnetic properties were measured with a Quantum Design Physical Properties Measurement System, endowed with a vibrating sample magnetometer (VSM) option with a resolution of 1×10^{-6} emu. Magnetization curves were measured from temperatures from 2.5 to 300 K, with a maximum applied magnetic field of 3 kA/m. Zero field cooling (ZFC) and field cooling (FC) measurements of M vs. T were carried out as follows: for ZFC, the sample was cooled from room temperature to 3 K without an external magnetic field. Once at the lowest temperature, a magnetic field of 4 kA/m is applied, and the magnetization is measured as the temperature increases. ZFC curve starts lower, increases rapidly to a peak and then decreases gradually. For FC, a constant magnetic field of 4 kA/m was applied as the sample was cooled from room temperature to 3 K, then, the temperature was increased while measuring M . From RT up to 995 K, M was measured vs. T with an applied magnetic field of 4 kA/m.

4. Conclusions

The AlCoCrFeNi high-entropy alloy exhibits a strong interplay between structural, microstructural, and magnetic properties across temperatures. The as-cast microstructure consists of equiaxed grains ($\sim 80 \mu\text{m}$), dendrites ($\sim 70 \mu\text{m}$ in length), and interdendritic nanoprecipitates, which influence phase stability and exchange interactions.

As temperature increases, the alloy transitions from dual BCC phases (A2/B2) to FCC, which dominates at high temperatures. Upon cooling, the A2 phase recrystallizes, reducing crystallite size ($\sim 33.50 \text{ nm}$) and promoting texture along (211) planes. Magnetization decreases near 380 K (BCC Curie temperature), peaks at 830 K due to enhanced exchange coupling and compositional fluctuations, and sharply drops at 930 K, marking the experimentally determined $T_C \sim 958 \text{ K}$. Effective moments, $\mu_{eff} = 1.35 \mu_B$, and its component along the applied field, $\mu_H = 0.646 \mu_B$ at 2.5 K, reflect temperature-dependent exchange weakening in the BCC phase.

At higher temperatures, beyond structural transformations, vacancy-mediated atomic diffusion and compositional fluctuations facilitate Fe, Co, and Ni redistribution, modify-

ing local exchange interactions and reinforcing moment alignment. These fluctuations, combined with the presence of nanoprecipitates, act as pinning sites, altering coercivity and hysteresis behavior. The combined effects of phase stability, dipolar interactions, dendritic connectivity, and vacancy-driven compositional changes shape the alloy's magnetic response, offering a framework for temperature-sensitive scientific and industrial applications.

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