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Research Article

Structural and magnetic evolution of FeCoNiAlMn thin films under varying Mn content and annealing temperatures

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ABSTRACT

This study focuses on FeCoNiAl films with varying concentrations of Mn added, deposited using the DC sputtering technique. All films were fabricated with a uniform thickness of approximately 100 nm. Four series of FeCoNiAl-Mn films were prepared at room temperature and subsequently annealed at different temperatures: 400°C, 600°C, and 800°C. The annealing was carried out in a furnace under an argon atmosphere, with a heating rate of 2°C/min for a duration of 1 h. The X-ray diffraction (XRD) analysis reveals that the crystal structure changes as a function of both Mn concentration and annealing temperature. The structure transitions from face-centred cubic (FCC) in FeCoNi to body-centred cubic (BCC) upon the addition of Al and Mn, and eventually to an amorphous phase at higher AlMn concentrations. The magnetic properties also exhibit significant changes upon annealing at 400°C. For the Fe_{1.2}CoNiAl_{1.4}Mn_{2.5} film, the saturation magnetization increases by approximately 35-fold. Similarly, the Fe_{1.1}CoNiAl_{1.5}Mn_{3.3} film demonstrates a tenfold increase in saturation magnetization, accompanied by marked reductions in coercivity and remanence, by factors of roughly 29 and 43, respectively.

1. Introduction

FeCoNi alloys have garnered significant research interest due to their remarkable soft magnetic performance and favorable thermal stability (Osaka, Sawaguchi *et al.* 1999; Saito, Yamada *et al.* 1999; Osaka, Asahi *et al.* 2005; Wu, Xu *et al.* 2008). Owing to their exceptional magnetic characteristics, these alloys hold strong potential for widespread application in sensors, actuators, and magnetic data storage technologies (Wu, Xu *et al.* 2008; Alsebaie and Morley 2025). High-entropy alloys (HEAs) are distinguished by four fundamental features (Yeh, Chen *et al.* 2004; Lu, Wang *et al.* 2015; Varvenne, Leyson *et al.* 2017): high configurational entropy, significant lattice distortion, slow atomic diffusion, and the so-called cocktail effect. These characteristics, collectively, tend to promote the formation of simplified phase structures, in contrast to the multiple brittle and complex intermetallic compounds typically anticipated by conventional metallurgical principles (Salishchev, Tikhonovsky *et al.* 2014). The addition of selected elements to the FeCoNi ternary alloy appears to offer a promising pathway for developing materials with enhanced comprehensive properties, attributed to the intrinsic 'cocktail effect' characteristic of the HEAs (Xu, Zhang *et al.* 2020; Akbari, Vafaei *et al.* 2024). Moreover, the incorporation of additional alloying elements into FeCoNi-based systems induces distinct physical and magnetic behaviors, enabling their adaptation for targeted applications such as transformers, electric motors, and electromagnetic devices (Zuo, Li *et al.* 2014).

In FeCoNi(MnAl)_x HEAs, strong atomic interactions are observed among Al, Ni, Co, and Mn atoms, primarily due to the significant negative mixing enthalpies of the Al-Ni, Al-Co, and Al-Mn pairs,

reported as -22, -19, and -19 kJ/mol, respectively (Alsebaie and Morley 2025). These strong affinities among constituent elements promote the formation of short-range order (SRO) structures (Akbari, Vafaei *et al.* 2024; Alsebaie and Morley 2025). However, numerous studies have demonstrated that even minor variations in heat treatment conditions can lead to significant changes in both the structural and magnetic properties of a material. Consequently, precise control of the heat treatment process is often essential for optimizing the magnetic performance of magnetic materials (Belmeguenai, Gabor *et al.* 2016; Bormio-Nunes, Cardoso *et al.* 2022). Akbari *et al.* investigated the effect of annealing on FeCoNi(MnAl)_x alloys and reported an enhancement in their soft magnetic properties (Akbari, Vafaei *et al.* 2024). A previous investigation examined the FeCoNiCrCu_{0.5} alloy in both its as-cast state and after annealing at various temperatures, revealing a clear influence of annealing temperature on phase transformation and material properties. Nevertheless, limited research has comprehensively explored these alloy systems, particularly regarding the impact of annealing conditions on their microstructural evolution, thermal behavior, and electrochemical corrosion resistance (Lin and Tsai 2011). Lu *et al.* (2021) investigated the effect of annealing at various temperatures on the magnetic and mechanical properties of FeCoNiMn_{0.25}Al_{0.25} HEA synthesized via casting. The as-cast alloy exhibited a single-phase face-centered cubic (FCC) structure. However, annealing at elevated temperatures significantly altered the phase composition. Specifically, a heterogeneous microstructure consisting of an FCC matrix with body-centered cubic (BCC) precipitates was observed in the alloys annealed at 750°C, 850°C, and 950°C (Lu, He *et al.* 2021).

The majority of the research into FeCoNiMnAl alloys has studied the heat treatment of bulk alloys, while this paper presents an investigation

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into the magnetic and structural changes induced by varying Mn concentrations in FeCoNiAl alloys in thin films, along with the effects of annealing at different temperatures on these films.

2. Experimental Work

In this study, a series of samples was prepared using direct current (DC) magnetron sputtering using a three-gun sputtering system. The films were deposited onto (100)-oriented silicon substrates, each measuring 10 mm × 10 mm. Prior to deposition, the substrates were cleaned using acetone and isopropanol in an ultrasonic bath for 10 min, followed by drying with nitrogen gas. The base pressure of the chamber before sputtering was approximately 3×10^{-7} Torr. Three targets were employed in the sputtering process: a FeCoNi alloy (with an atomic composition of 33.3% for each element), aluminum (99.999% purity), and manganese (99.9% purity). The powers applied to the FeCoNi and Al targets were fixed at 70 W and 35 W, respectively, across all samples. In contrast, the power applied to the Mn target was either 35 W, 70 W, or 105 W for the different samples. Argon gas was used as the sputtering plasma at a constant pressure of 5×10^{-3} Torr. All films were deposited at room temperature with a fixed thickness of 100 nm. Prior to each deposition, a 2-min pre-sputtering step was conducted to clean the target surfaces. Subsequently, the shutters were opened simultaneously from all targets to initiate the film growth.

After the deposition, the samples were annealed at different temperatures: 400°C, 600°C, and 800°C. Each annealing process was conducted for 1 h with a controlled heating rate of 2°C/min. Both heating and cooling were performed in a furnace under an argon (Ar) atmosphere. The crystal structure of the films was studied by X-ray diffraction (XRD). The XRD measurements were performed using a Bruker D2 Phaser diffractometer equipped with a Cu $K\alpha_1$ radiation source ($\lambda = 1.54184 \text{ \AA}$), operating at 40 kV and 40 mA. $\theta/2\theta$ scans were carried out over a limited angular range of 25° to 65° with a step size of 0.020° to selectively investigate the structural characteristics of the deposited films while avoiding the intense diffraction peaks associated

with the silicon substrate, which typically appear around 65°. X-ray photoelectron spectroscopy (XPS) analyses were performed using a Kratos Supra instrument equipped with a monochromated Al source (photon energy = 1486.6 eV), which was used to identify the film's composition. Two analysis points were measured for each sample, with an analysis area of $700 \mu\text{m} \times 300 \mu\text{m}$. Survey spectra were acquired over a binding energy range of 1200 to 0 eV, using a pass energy of 160 eV, with a step size of 1 eV. Each scan was collected in a single sweep with a total acquisition time of 300 s. The magnetic properties, including saturation magnetization (M_s), coercivity (H_c), and remanence (M_r), were measured using a Superconducting Quantum Interference Device (SQUID) magnetometer. Hysteresis loops (M–H curves) were recorded at a temperature of 300 K under an applied magnetic field ranging from +800 kA/m to −800 kA/m.

3. Results and Discussion

The results for the different techniques are split into two sections: one studying the phase and composition of the films, and the other the magnetic properties.

3.1 XPS and XRD results

XPS was employed to determine the elemental composition of the films. As shown in Fig. 1, the intensity of the Mn 2p peaks at binding energies of approximately 831 eV and 845 eV increases with the applied sputtering power, which was varied from 35 W to 105 W. The elemental compositions for each film, calculated using CasaXPS software, have been presented in Table 1.

Fig. 2 provides a schematic overview of the phase transformations induced by annealing, serving as a visual guide for the structural analysis presented in Fig. 3. Fig. 3(a) presents the XRD patterns of FeCoNi in its as-deposited state and after annealing at 400°C, 600°C, and 800°C. In its as-deposited form, the FeCoNi alloy exhibits a face-centered cubic (FCC) metallic crystal structure (Yang, Tsau *et al.* 2018,

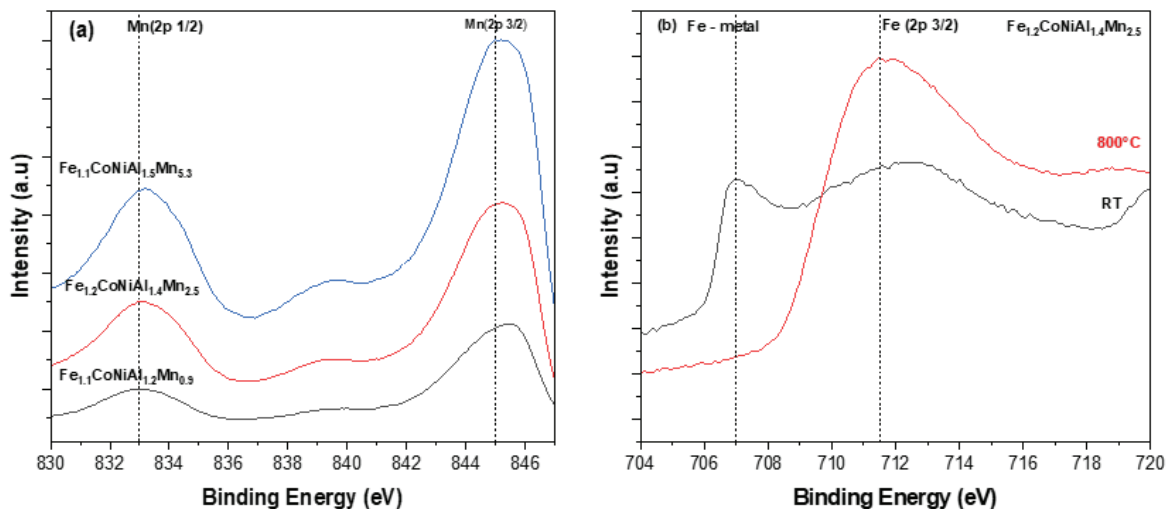


Fig. 1. XPS spectra of FeCoNiAlMn thin films with different Mn concentrations. (a) Mn 2p_{1/2} and Mn 2p_{3/2} spectra for Fe_{1.1}CoNiAl_{1.5}Mn_{0.9}, Fe_{1.2}CoNiAl_{1.4}Mn_{2.5}, and Fe_{1.1}CoNiAl_{1.5}Mn_{5.3} films deposited at room temperature. The increase in Mn peak intensity corresponds to higher sputtering power applied to the Mn target. (b) Fe 2p spectra of Fe_{1.2}CoNiAl_{1.4}Mn_{2.5} showing the metallic Fe peak at room temperature and its disappearance after annealing at 800°C, confirming oxide formation.

Table 1.

Elemental composition of FeCoNiAlMn thin films measured by XPS. Values are given in atomic percent (at%). Films were deposited at room temperature using DC magnetron sputtering, while Mn concentration was varied by adjusting the sputtering power applied to the Mn target.

Film	Annealing	Fe(at%)(±4)	Co(at%)(±4)	Ni(at%)(±4)	Al(at%)(±4)	Mn(at%)(±4)
Fe _{1.2} Co _{1.2} Ni	RT	36	35	29	0	0
Fe _{1.1} CoNiAl _{1.2} Mn _{0.9}	RT	21	19	19	23	18
Fe _{1.2} CoNiAl _{1.4} Mn _{2.5}	RT	18	14	14	20	34
Fe _{1.1} CoNiAl _{1.5} Mn _{5.3}	RT	11	10	10	15	54

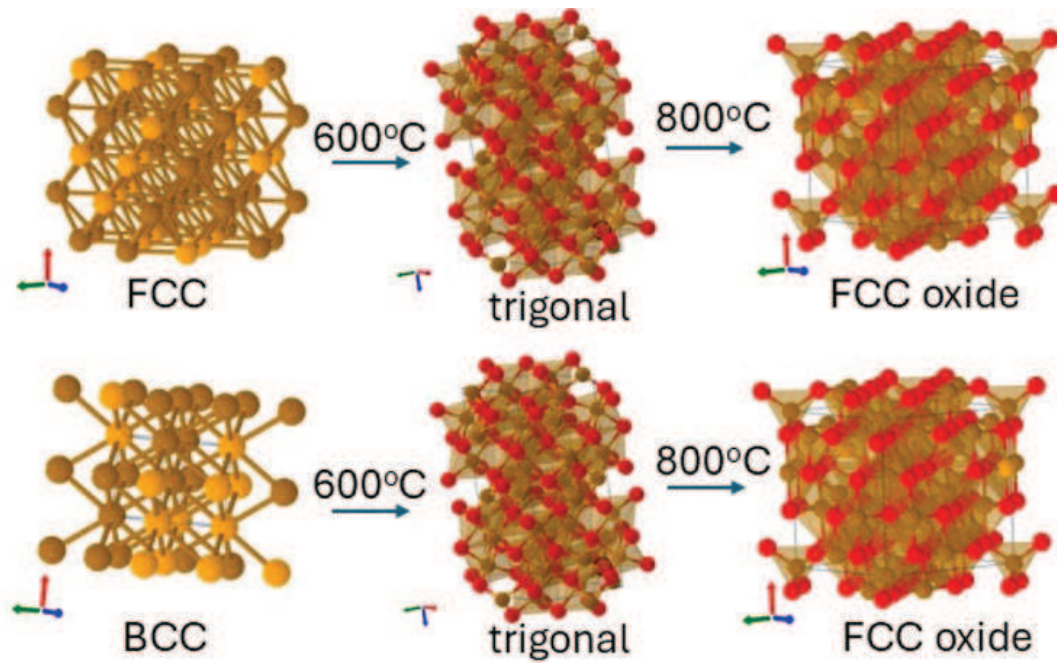


Fig. 2. shows the change in crystal structure within the films upon annealing. The brown/orange atoms are the metallic elements (Co, Fe, Ni, Mn, and Al), and the red atoms are oxygen ions (Project 2025).

Alsebaie and Morley 2025). Upon annealing at 400°C, the diffraction peak at 44.5° becomes sharper, indicating increased crystallinity and grain growth as confirmed by the grain size data in Table 2. When the annealing temperature reaches 600°C, the FCC peak disappears, and a new peak emerges at 33°, corresponding to the trigonal phase of Fe_2O_3 . Further annealing at 800°C results in a transformation to an FCC oxide structure, consistent with the findings reported by Yang *et al.* (Yang, Tsau *et al.* 2018) and the standard diffraction pattern (PDF No. 04-026-0037). Fig. 2 shows the crystal structure changes with annealing.

Fig. 3(b) displays the XRD pattern of $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$ film. In the as-deposited state, the sample exhibits a body-centered cubic (BCC) crystal structure, which has previously been observed in other CoFeNiAlMn films (Bazioti, Løvvik *et al.* 2022, Alsebaie and Morley 2025). It is the addition of the Al that drives the transition to the BCC phase (Alsebaie and Morley 2025). Upon annealing at 400°C, the diffraction peaks become narrower, indicating enhanced crystallinity. With further annealing at 600°C, the emergence of peaks corresponding to the trigonal phase of Fe_2O_3 is observed, similar to the transformation noted in the FeCoNi film under the same conditions (Fig. 2). At 800°C, the annealed sample reveals an FCC-type oxide structure, confirmed by the presence of three distinct characteristic peaks 30.4°, 35.9° and 63.1° (Yang, Tsau *et al.* 2018; Data 2021).

Fig. 3(c) presents the XRD patterns of the $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ film in the as-deposited state and after annealing at 400°C, 600°C, and 800°C. The as-deposited sample exhibits an amorphous phase, likely due to the high Mn content, which has been noticed to promote amorphization in the absence of post-deposition thermal treatment (Alsebaie and Morley 2025). Upon annealing at 400°C, the sample transitions to a BCC crystal structure. Further annealing at 600°C and 800°C results in the formation of an FCC oxide phase (Yang, Tsau *et al.* 2018, Data 2021). At 800°C, the XRD spectrum shows a notable increase in peak intensity, indicating enhanced crystallinity and the development of a preferred orientation among grains. Additionally, a consistent leftward shift in the FCC oxide peaks is observed between the 600°C and 800°C annealing treatments. This suggests that at 600°C, the peak may be at the beginning of the formation of the FCC oxide phase. Also, this shift suggests an increase in the interplanar spacing with higher annealing temperature, in accordance with Bragg's Law ($2d \sin \theta = n\lambda$). The FCC oxide peaks are confirmed from the XPS data, Fig. 1(b), where the metallic Fe peak is observed for the as-grown film, but disappears for the 800°C annealed film, showing that an oxide phase is present.

Table 2.

The Lattice constants and grain sizes of FeCoNiAlMn films extracted from XRD patterns at different annealing temperatures (RT, 400°C, 600°C, 800°C). Lattice constants were calculated using the cubic lattice spacing formula, and grain sizes were estimated using the Scherrer equation. The table summarizes the structural evolution of the films, including transitions between FCC, BCC, amorphous, and FCC oxide phases.

Film	Annealing (°C)	2 θ	Miller Indices	Lattice constant (Å)	Grain size (Å)
$\text{Fe}_{1.2}\text{Co}_{1.2}\text{Ni}$	RT	44.2	(111)	3.55	108
	400	44.5	(111)	3.51	136
	600	32.9	(104)	11.2	225
	800	30.2	(220)	8.35	522
		35.6	(311)	8.34	198
$\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$	RT	44.7	(110)	2.86	105
	400	45	(110)	2.84	53
	600	32.9	(104)	11.19	210
	800	30.4	(220)	8.31	522
		35.9	(311)	8.28	318
		63.1	(440)	8.32	296
$\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$	400	45.1	(110)	2.84	105
	600	30.1	(220)	8.39	174
		62.6	(440)	8.38	80
	800	30.5	(220)	8.27	348
		36	(311)	8.25	122
		63.5	(440)	8.28	237
$\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$	400	45.6	(110)	2.81	71
	800	30.5	(220)	8.27	313
		35.9	(311)	8.28	318
		43.2	(400)	8.36	361
		63.3	(440)	8.31	187

Fig. 3(d) presents the X-ray diffraction pattern of the $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ samples under different thermal treatments. At

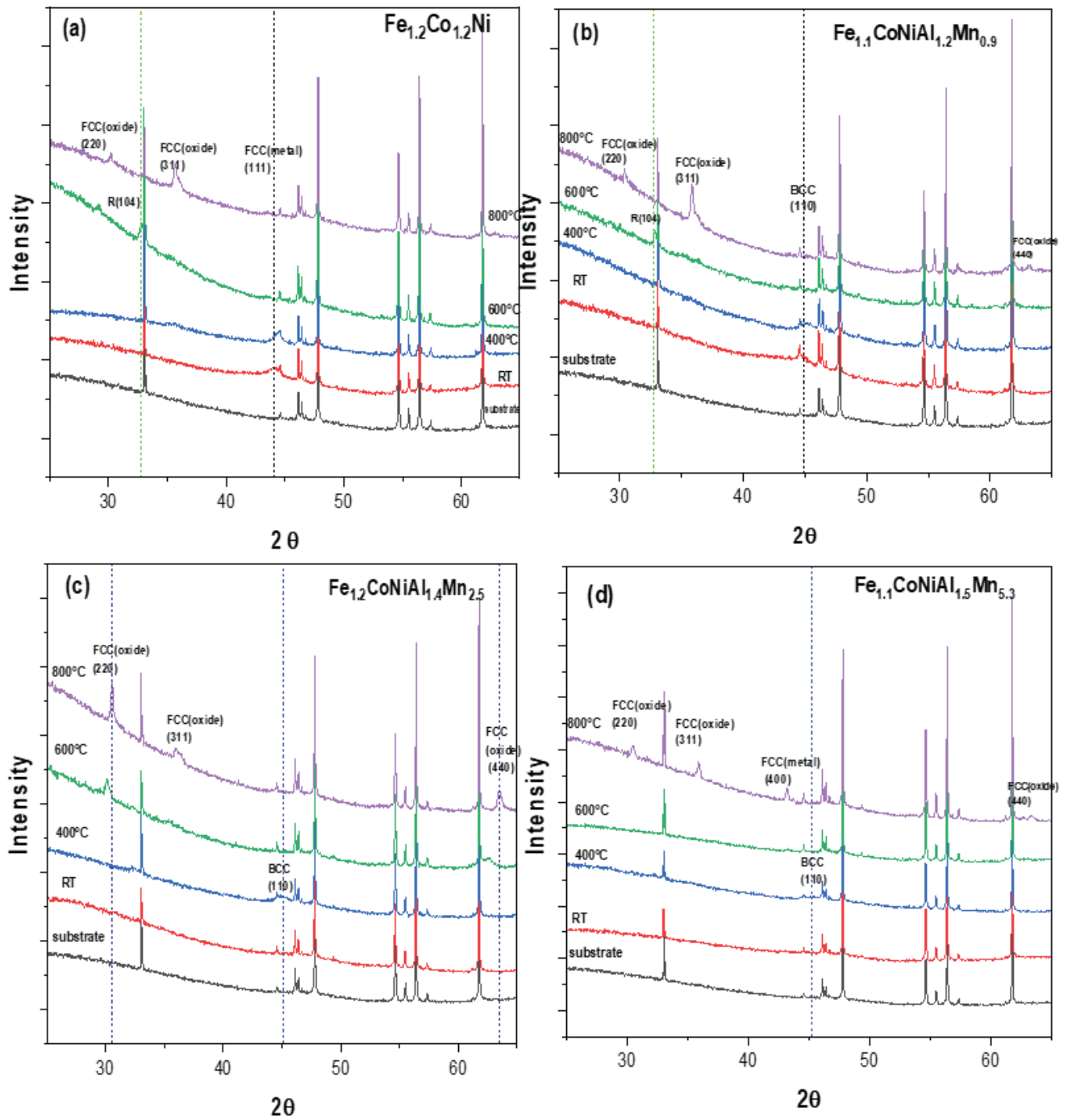


Fig. 3. XRD patterns of FeCoNiAlMn films at RT, 400°C, 600°C, and 800°C. (a) FeCoNi showing FCC structure and oxide formation at higher temperatures. (b) $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$ showing BCC at RT and FCC oxide at 800°C. (c) $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ transitioning from amorphous to BCC and then FCC oxide. (d) $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ transitioning from amorphous to mixed FCC metal/oxide at 800°C.

room temperature, the sample exhibits a fully amorphous structure, as indicated by the absence of distinct diffraction peaks. Upon annealing the sample at 400°C, a broad peak begins to appear, corresponding to a BCC structure. However, the peak remains relatively wide, indicating that the crystallinity is still limited and that the structure is not fully ordered. When the sample is annealed at 600°C, it returns to an amorphous state, with the diffraction pattern showing no sharp peaks once again. This may suggest a thermal destabilization of the previously formed BCC phase or the formation of a disordered phase at this temperature. However, the broad diffraction features may not necessarily correspond to a fully amorphous phase; instead, they could indicate the presence of nanocrystalline domains or short-range ordered structures. Finally, at 800°C, several distinct peaks emerge in

the XRD spectrum. These peaks correspond to both FCC metal and FCC oxide phases, indicating the formation of multiple crystalline structures (Yang, Tsau *et al.* 2018; Alsebaie and Morley 2025). The appearance of FCC oxide peaks at 800°C is most likely related to trace oxygen impurities in the annealing atmosphere, possibly from residual oxygen in high-purity Ar gas or disruption from furnace walls or substrate surfaces during heating. Such effects become more pronounced at elevated temperatures due to enhanced diffusion kinetics. A minor furnace leak could also contribute to oxide formation, although the primary source is expected to be residual oxygen. Although the lattice constant in Table 2 was calculated from the diffraction peaks using the cubic lattice spacing formula and fitted with Fityk software, the obtained value closely matches those reported by Li *et al.* (Li, Bai *et*

al. 2020) and the standard diffraction pattern (PDF No. 04-026-0037) (Data 2021). Table 2 shows that the grain size, calculated from the Scherrer equation (Muniz, Miranda *et al.* 2016), increases notably with annealing temperature, particularly beyond 600°C, due to enhanced atomic diffusion and recrystallisation. This trend reflects the typical grain coarsening behavior associated with thermal treatment and is further influenced by structural phase transformations, especially the formation of oxide phases at higher temperatures.

3.2 Magnetic properties

Fig. 4 presents the variation in coercive field and remanence for the investigated samples as a function of annealing temperature. As shown in Fig. 4(a), the coercivity values for FeCoNi and Fe_{1.1}CoNiAl_{1.2}Mn_{0.9} films are approximately the same at room temperature, 400°C, and 800°C, indicating relatively stable magnetic softness under those conditions, even though there is a change in phase within the films due to the annealing. However, at 600°C, there is a noticeable and significant increase in coercivity, nearly doubling compared to other temperatures. This sharp increase suggests a possible change in the microstructure at this specific annealing condition. It is worth noting

that the XRD patterns in Figs. 3(a and b) show a peak at 32.7°, from the oxide phase, which may be correlated with the observed rise in coercivity, as the oxide phase will be an ordered structure (Fig. 2) with larger grain sizes (Table 1). From Herzer's paper (Herzer 1990), the coercive field increases with grain size as D^6 . This peak could be attributed to a phase transformation or the appearance of a secondary phase that enhances magnetic anisotropy. Furthermore, this shift in coercivity at 600°C implies that both FeCoNi and Fe_{1.1}CoNiAl_{1.2}Mn_{0.9} alloys transition from soft magnetic materials to semi-hard magnetic materials (Gutfleisch, Willard *et al.* 2011).

Fig. 4(b) illustrates the coercivity behavior of Fe_{1.2}CoNiAl_{1.4}Mn_{2.5} and Fe_{1.1}CoNiAl_{1.5}Mn_{5.3} alloys at different annealing temperatures. Since these films exhibit an amorphous structure at room temperature with no long-range atomic order, the magnetocrystalline anisotropy is essentially negligible. The magnetic behavior is therefore dominated by random anisotropy and magnetoelastic strain, consistent with observations in amorphous Fe-based alloys, where the anisotropy originates from internal strain rather than from the crystal lattice (Abbas and Morley 2017). At room temperature, this means that both samples exhibit relatively low coercivity values, with only slight differences between them, due to having no grains as they are amorphous. However, upon

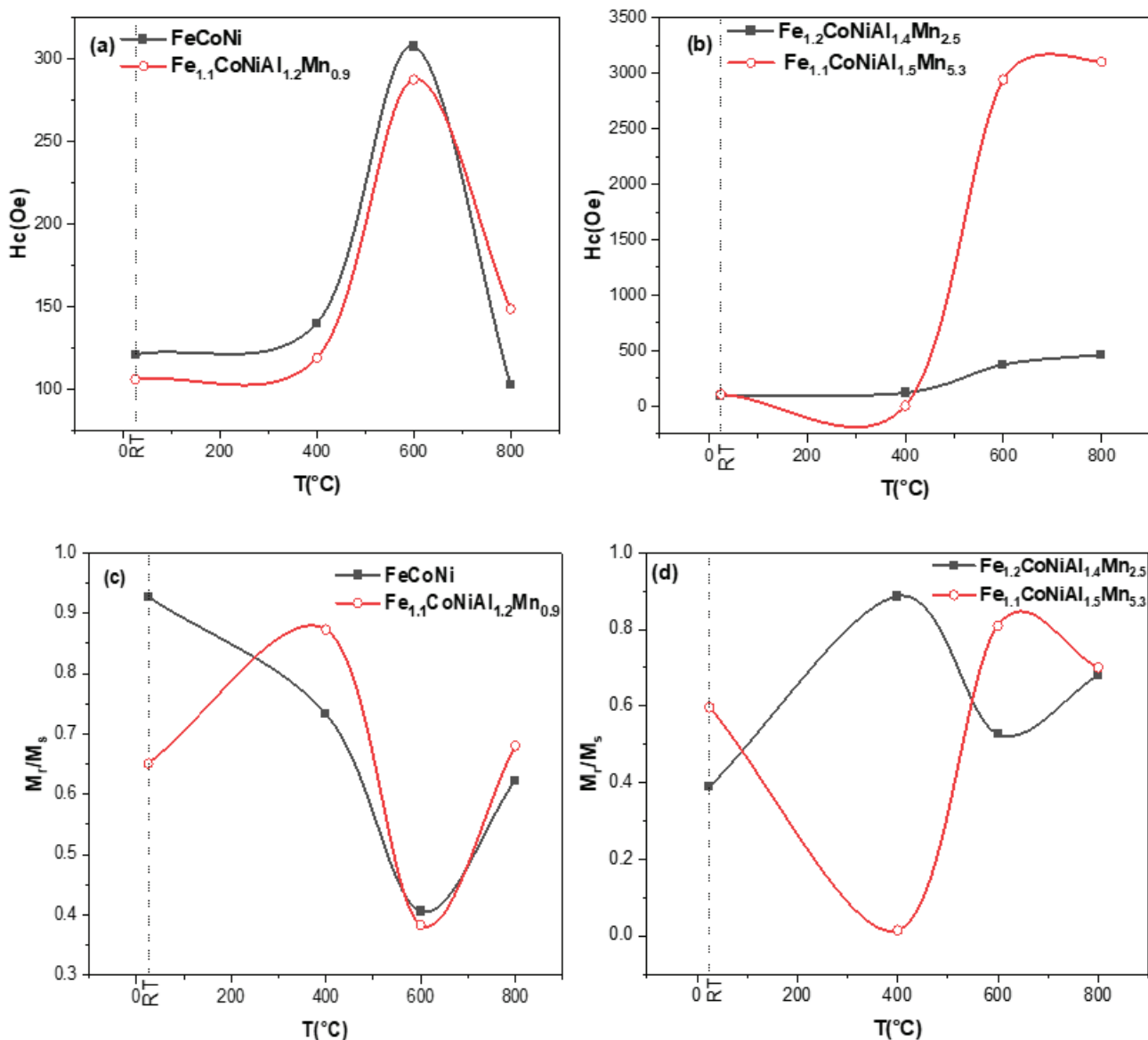


Fig. 4. (a-d) Show the Coercivity (Hc) and normalized remanence (Mr/Ms) of FeCoNiAlMn thin films measured at different annealing temperatures (RT, 400°C, 600°C, 800°C). (a) and (c) FeCoNi and Fe_{1.1}CoNiAl_{1.2}Mn_{0.9}. (b) and (d) Fe_{1.2}CoNiAl_{1.4}Mn_{2.5} and Fe_{1.1}CoNiAl_{1.5}Mn_{5.3}. The plots show the evolution of magnetic behavior as a function of thermal treatment for each composition.

annealing at 400°C, the $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ sample shows a noticeable drop in coercivity to approximately 4 Oe, which may be attributed to the presence of a very broad BCC peak, suggesting nano-size grains within the film. As the annealing temperature increases to 600°C and 800°C, both samples undergo significant changes in their magnetic behavior. $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ transitions from a soft magnetic to a hard magnetic material, most likely due to a phase transformation from BCC to FCC oxide, as confirmed by XRD results. In addition to this phase evolution, Herzer's random-anisotropy model (Herzer 1990) supports this trend by predicting that coercivity increases sharply as the grain size approaches the exchange length and decreases again for excessively large grains. Since Table 2 shows a clear grain-size growth with increasing annealing temperature, the combined effects of oxide formation and grain coarsening explain the observed hardening at 600°C and the subsequent reduction in coercivity at higher temperatures. In contrast, $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ also shows a structural change from BCC at 400°C to FCC at higher annealing temperatures; however, its magnetic behavior only shifts from soft to semi-hard, which can likewise be attributed to its more moderate grain-size evolution, consistent with Herzer's model. Further, one possible explanation for the difference in magnetic hardness between the two alloys is the variation in manganese content. The higher Mn concentration in $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ may have played a key role in promoting structural ordering and stabilizing the FCC oxide phase, which ultimately led to its transition into a hard magnetic. While the correlation between structural phase transitions and magnetic properties is evident, other factors such as domain wall pinning, exchange coupling between different phases, and magnetoelastic effects could also contribute to the observed behavior.

Figs. 4(c and d) present the remanence values (M_r/M_s) for FeCoNi, $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$, $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$, and $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ alloys at various annealing temperatures. Fig. 4(c) highlights the behavior of FeCoNi and $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$ films, both of which exhibit relatively high remanence values at room temperature and 400°C. Such elevated values are typically undesirable for soft magnetic materials, as they indicate stronger magnetic retention and lower reversibility. Upon increasing the annealing temperature to 600°C, the remanence of both alloys decreases significantly, falling within the ideal range for semi-hard magnetic materials. This suggests a possible phase transformation from FCC for FeCoNi and BCC for $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$ to the trigonal phase of Fe_2O_3 as observed in Figs. 3(a and b) and shown in Fig. 2. However, at 800°C, the remanence increases again, reaching approximately 0.8 M_r/M_s . This level is generally considered high and, therefore, unsuitable for applications requiring soft magnetic behavior. The increase may be related to structural evolution from the trigonal phase to FCC oxide,

as shown in Figs. 3(a and b), such as grain growth, or stabilization of a high-anisotropy phase at elevated temperatures. Although linked to structural transitions, domain wall pinning, phase interactions, and magnetoelastic coupling may also affect the remanence behavior.

Fig. 4(d) displays the normalized remanence (M_r/M_s) for $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ and $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ alloys at various annealing temperatures. For $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ films, the M_r/M_s value at room temperature is relatively high for a soft or semihard magnetic material. However, after annealing at 400°C, the remanence drops significantly, entering a range that is suitable for soft magnetic applications. When this behavior is correlated with the low coercivity observed at the same temperature, the alloy demonstrates potential as a soft magnetic material under these conditions. As the annealing temperature increases to 600°C and 800°C, the normalized remanence rises again, reaching approximately 0.8. When combined with the corresponding increase in coercivity, this indicates a shift toward hard magnetic behavior. Although the overall magnetization remains relatively low, $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ may still be considered a promising candidate for certain hard magnetic applications at these annealing temperatures.

In the case of $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ films, the M_r/M_s value starts at around 0.4 at room temperature. After annealing at 400°C, the remanence exhibits a sharp increase to approximately 0.9, which is considered too high for both soft and semi-hard magnetic classifications. This behavior may be attributed to structural change from amorphous to BCC, as shown in Fig. 3(c), or magnetic ordering induced by annealing. Upon further annealing at 600°C, the M_r/M_s decreases back into the semihard magnetic range, indicating a potential improvement in magnetic reversibility. Finally, at 800°C, the remanence slightly increases to around 0.7, which is still relatively high but suggests the material maintains semihard magnetic characteristics, and that might be due to the change to FCC oxide structure as seen in Fig. 3(c).

Fig. 5 presents the variation in saturation magnetization (M_s) for the different alloy compositions as a function of annealing temperature. In Fig. 5(a), FeCoNi exhibits a noticeable decline in M_s with increasing annealing temperature. Between 600°C and 800°C, the M_s stabilizes at a relatively low value. This reduction and subsequent stabilization could be attributed to the absence of a crystal structure transition, as the alloy maintains an FCC phase throughout the annealing process. Similarly, the $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$ alloy follows a comparable trend, with M_s decreasing as the annealing temperature increases. In this case, the alloy initially possesses a BCC structure, but transitions to an FCC phase upon annealing. The drop in M_s can therefore be linked to this structural transformation, specifically the shift from BCC to FCC,

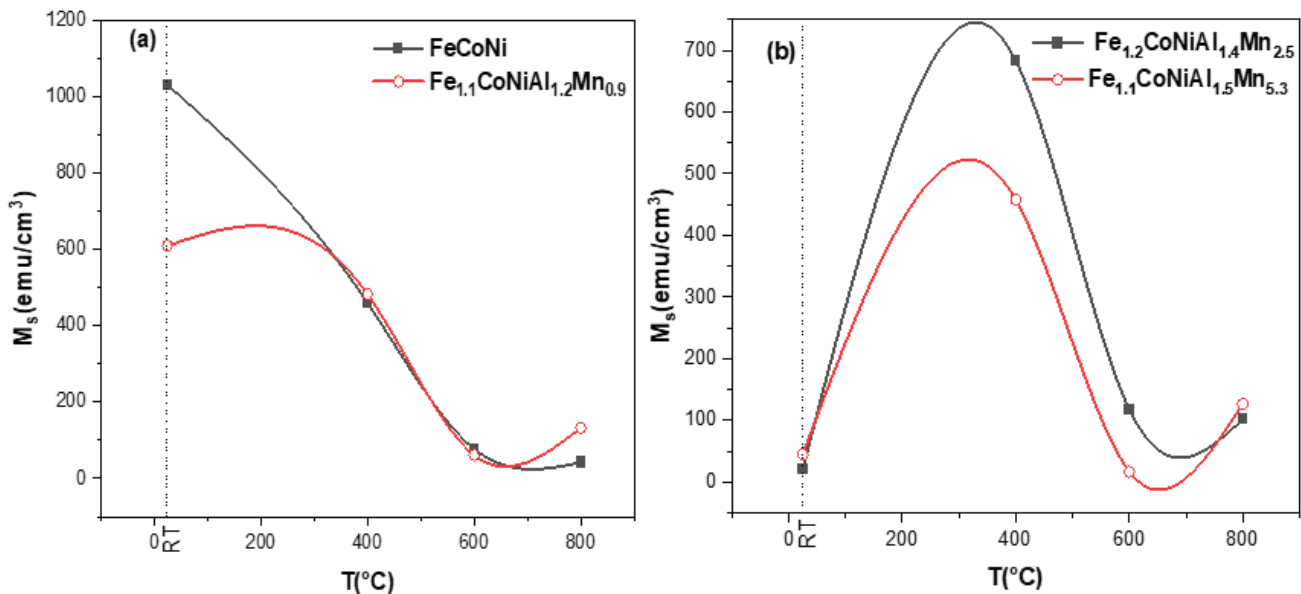


Fig. 5. Saturation magnetization (M_s) of FeCoNiAlMn thin films at different annealing temperatures (RT, 400°C, 600°C, 800°C). (a) FeCoNi and $\text{Fe}_{1.1}\text{CoNiAl}_{1.2}\text{Mn}_{0.9}$. (b) $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ and $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$. The figure highlights the changes in M_s with increasing temperature and the influence of Mn content on magnetic behavior.

which, unlike the typical FCC-to-BCC transition, does not lead to an improvement in M_s .

Fig. 5(b) reveals a significant enhancement in the saturation magnetization (M_s) for the $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ alloy. At room temperature, the M_s is approximately 20 emu/cm³. However, after annealing at 400°C, it dramatically increases to nearly 700 emu/cm³, representing an increase of about 3500%. This sharp improvement is most likely attributed to the transformation of the crystal structure from an amorphous phase to a BCC structure, which typically supports better magnetic ordering. A similar trend is observed for the $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ alloy. Initially exhibiting an amorphous structure with low magnetic saturation at room temperature, the M_s rises sharply, by more than 1000% times, after annealing at 400°C, which correlates with the transition to a BCC phase. This is due to the amorphous phase only having very short-range magnetic order, leading to random anisotropy. When transitioned to a BCC structure (Fig. 2), the magnetic order is now over a longer range, even though the atoms are randomly distributed on each of the lattice sites, the regular crystal structure allows for long-range magnetic order, which increases the magnetization. It also means that the film has magnetocrystalline anisotropy. For both alloys, further annealing at 600°C and 800°C leads to a structural shift from BCC to FCC oxide. This transformation is the main reason behind the subsequent drop in M_s at higher annealing temperatures, as the FCC oxide phase generally offers lower saturation magnetization, due to the addition of non-magnetic oxygen to the structure (Fig. 2). Also the FCC oxide structure is likely to have 2+ and 3+ ions aligned in an anti-parallel arrangement, making the films a ferrimagnet rather than a ferromagnet (Teja and Koh 2009, Gareev 2023).

For all the films studied, the films that were annealed at 800 °C all had an FCC oxide structure, and the saturation magnetization was all ~100 emu/cm³. This suggests that the oxide lattice arrangement gives a similar magnetization, independent of the concentration of the elements, due to the formation of magnetic ions.

Considering the significant enhancement in magnetic properties upon annealing, it is evident that thermal treatment plays a key role in tailoring the magnetic behavior of FeCoNi-based alloys. For instance, the $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ alloy shows a more than thirtyfold increase in saturation magnetization when annealed from room temperature to 400°C. Similarly, the $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ alloy exhibits a substantial improvement in soft magnetic performance after annealing at 400°C, with saturation magnetization increasing by more than an order of magnitude compared to the as-deposited state. This enhancement is accompanied by dramatic reductions in coercivity and remanence by approximately 2900% and 4300%, respectively. As the annealing temperature increases further, the films transition from soft to hard magnetic behavior, as reflected by the rising coercivity and remanence. These results highlight the crucial influence of thermal processing on the phase evolution and magnetic response of high-entropy thin films.

4. Conclusions

In this study, a series of FeCoNiAlMn films with varying Mn content was fabricated using DC sputtering. Four films were prepared, and the effect of heat treatment on both the crystal structure and magnetic properties was systematically investigated for each. The addition of small amounts of Mn and Al to FeCoNi alters the crystal structure from FCC to BCC, while further increasing the Mn content leads to the formation of an amorphous structure. However, upon annealing, the initially amorphous films recrystallize. The magnetic properties exhibit significant enhancement in films with high Mn content, specifically $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ and $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$, after annealing at 400°C. The saturation magnetization of $\text{Fe}_{1.2}\text{CoNiAl}_{1.4}\text{Mn}_{2.5}$ increases substantially upon annealing to 400°C, while $\text{Fe}_{1.1}\text{CoNiAl}_{1.5}\text{Mn}_{5.3}$ also demonstrates an increase in saturation magnetization accompanied by a pronounced decrease in both coercivity and remanence, indicating its potential as a soft magnetic alloy.

These results demonstrate that Mn concentration and annealing temperature together play a decisive role in tailoring the structural and magnetic evolution of FeCoNiAlMn thin films. Moderate annealing (~400°C) enhances soft magnetic behavior through structural ordering, whereas higher annealing temperatures (600–800°C) promote oxide

formation and harder magnetic states. Such findings provide valuable insights for designing and optimizing HEA thin films for targeted applications ranging from soft magnetic devices (e.g., sensors, transformers) to hard magnetic applications (e.g., data storage and permanent magnets).

CRediT authorship contribution statement

Dkhilallah Meslet Alsebaie: Data curation, formal analysis, investigation, writing – original draft. **Nicola Morley Morley:** Conceptualization, project administration, resources, supervision, writing – original draft, writing – review and editing.

Declaration of competing interest

The authors declare that they have no competing financial interests or personal relationships that could have influenced the work presented in this paper.

Declaration of generative AI and AI-assisted technologies in the writing process

The authors confirm that there was no use of artificial intelligence (AI)-assisted technology for assisting in the writing or editing of the manuscript and no images were manipulated using AI.

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