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Using combinatorial sputtering to uncover new magnetic high entropy alloys

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Nicola A. Morley ; Richard M. Rowan-Robinson; Tom Wilkinson; Jing Bai ; Jianglong Gu



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ABSTRACT

The search for new magnetic thin films has been accelerated by the development of high-throughput fabrication using combinatorial sputtering followed by high-throughput characterization methods including x-ray diffraction, scanning electron microscopy, and magneto-optic Kerr effect magnetometry. From the compositional-property plot obtained from these measurements, down-selection of the most promising compositions for further in-depth studies can be carried out. This paper studies two different thin magnetic film systems with the compositions CoFeNiMn-*X*, where *X* is Al or Si. For each film system, the composition of each element ranges between 10 and 40 at. %, allowing for a wide selection of different compositions to be studied. Focusing on soft magnetic properties, the compositions down-selected are further characterized using SQUID magnetometry to determine the saturation magnetization and coercive field. This paper shows that, for combinatorially grown CoFeNiMn-Al films over a wide composition range, the main phase was BCC. When comparing the saturation magnetization measured for different CoFeNiMn-Al compositions from the combinatorial wafer, with those in the literature for single composition films, there was good agreement between them. For the CoFeNiMn-Si films, when the ratio of the elements was roughly equimolar, the composition was magnetic, with the XRD showing an amorphous phase and possible intermetallic phases. While reducing the Co, Fe, and Ni concentrations with respect to Mn and Si, the films were nonmagnetic and amorphous.

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I. INTRODUCTION

Since their conception over 20 years ago,¹ research into high entropy alloys (HEAs) has rapidly increased, due to some of their unique properties.^{1–4} These include high strength, good wear resistance, and high temperature stability.^{4,5} HEAs have been defined by different criteria⁴ including those based on composition (i.e., five or more elements in equimolar ratio or principal elements with the concentration between 35 and 5 at. %), those based on entropy (i.e., configurational entropy, $\Delta S_{conf} = R \ln x_i$, where *R* is the gas constant and *x_i* is the number of elements, and HEAs are those with $\Delta S_{conf} \geq 1.5R$), those that combine both composition

and entropy, and one that states the HEA must be single-phase solid-solution. For this work, we took the definition to be a composition of five or more elements, with concentrations between 35 and 5 at. %.

One research area of focus for HEAs has been their use as functional magnetic materials, either as soft magnetic materials or magnetocaloric materials.^{3,6,7} This is due to their base elements often being Co, Fe, and Ni, which all have magnetic moments, further the element Mn can have a ferro-magnetic moment when paired with certain elements and within certain structures. For example, Zuo *et al.*⁸ used DFT calculations to determine the

electronic and magnetic structure of CoFeNiMn, CoFeNiMn–Cr, and CoFeNiMn–Al alloys. They determined the predicted magnetic moment of each element in the HEA. For CoFeNiMn, they showed that Co, Fe, and Ni were all ferromagnetic, while Mn behaved antiferromagnetically, with the addition of Cr, a reduction in the magnetic moment of the other elements was predicted, while the addition of Al reduced the number of down-spins for Mn, which meant that Mn behaved ferromagnetically. This behavior between Al and Mn is also observed in the metastable Mn–Al τ -phase, where around the equimolar composition (40–60% Mn) the alloy is magnetic and a promising permanent magnet candidate.⁹

Due to the vast compositional space of HEAs, finding a promising composition for a new magnetic material often relies on researchers' knowledge along with trial and error, so takes a lot of time and effort. One way to explore these large composition spaces is to use combinatorial sputtering, this involves the sputtering of two or more different elements onto a wafer, to produce a composition gradient across the wafer. The number of different compositions produced across the wafer depends on the number of elements being sputtered, the angle of the sputter guns and spatial resolution of the measurements. Previous work has used this technique to create large material libraries on one 3-in. substrate.^{3,10,11} This provides a high-throughput fabrication method to create 100 s of compositions within a short period of time (i.e., half a day per wafer). Once fabricated, a broad overview of the properties can be determined using high-throughput characterisation, which can include x-ray diffraction (XRD), scanning electron microscopy (SEM), and magneto-optic Kerr effect (MOKE) magnetometry. These results provide composition-property maps, from which the most interesting compositions, such as those with the smallest or largest coercive field or highest Kerr voltage can be down-selected for further study using higher resolution techniques, such as SQUID magnetometry, four-probe resistivity measurements, and transmission electron microscopy (TEM).

There are two common sets of base elements used to investigate HEAs as magnetic materials, these are CoFeNi–Cr and CoFeNi–Mn. In Fig. 1, a set of violin plots are given for the CoFeNi–Cr–X and CoFeNi–Mn–X alloys measured magnetization presented in the literature.^{8,12–18} As Al is the most common fifth element; 13 different alloy compositions for both CoFeNi–Cr–Al and CoFeNi–Mn–Al, along with fifth different compositions for CoFeNi–CrMn–Al and four different compositions for CoFeNi–Al, they are plotted separately, with the size and the shape representing the variation in magnetization with the addition of Al. The maximum saturation magnetisation plotted was 160 Am²/kg, which is the cut-off for the violin plots. Other fifth elements, such as Ga, Sn, and Zr, are less common additions within the literature, so the saturation magnetisations are plotted together within the violin plot, with nine different compositions for the CoFeNi–Cr–X plot and only three for the CoFeNi–Mn–X plot. From Fig. 1 and the literature, for equimolar HEAs, it is observed that the addition of Cr or Mn to CoFeNi reduces the saturation magnetisation (M_s), from $M_s = 156$ Am²/kg for CoFeNi,¹⁶ to <1 Am²/kg for CoFeNiCr¹² and 18 Am²/kg for CoFeNiMn.⁸ The addition of Al can help maintain a higher M_s within the HEAs, i.e., CoFeNiMn–Al⁸ $M_s = 148$ Am²/kg and CoFeNiCr–Al¹³ $M_s = 54$ Am²/kg. The

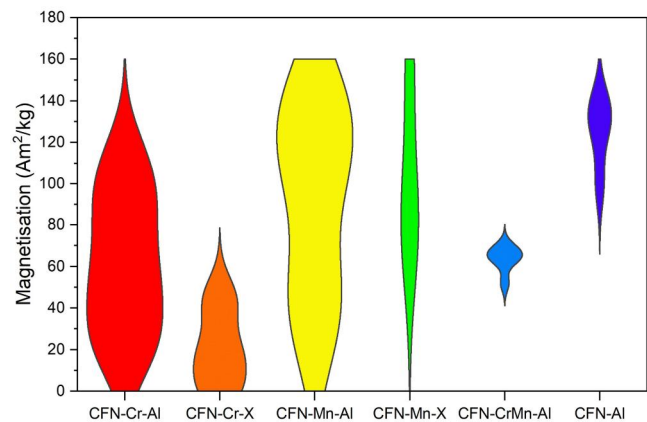


FIG. 1. Saturation magnetization of different HEAs with either CoFeNi–Mn (CFN–Mn) or CoFeNi–Cr (CFN–Cr) bases measured at 300 K. Data taken from Refs. 8 and 12–18, where X is either Ga, Sn, or Zr.

addition of other elements, such as Ga, Sn to CoFeNi–Mn⁸ can result in a higher magnetization than when added to CoFeNi–Cr.¹² For example, CoFeNiMn–Ga⁸ $M_s = 80$ Am²/kg, while CoFeNiCr–Ga¹² $M_s = 38$ Am²/kg. The alloy CoFeNiCrMn was determined by Zuo *et al.*⁸ to be nonmagnetic at room temperature. Figure 1 also shows that the majority of the research has focused on CoFeNi–Cr base alloys (27 different compositions),^{12–15} while fewer papers have focused on CoFeNi–Mn alloys (21 different compositions)^{8,16–18} with most of these using Al as the fifth element (18 different compositions). Thus, CoFeNi–Mn HEAs are an under explored research field and perfect to demonstrate the capability of high-throughput research. This paper uses combinatorial sputtering to fabricate a series of CoFeNiMn–X (X = Al and Si) films, followed by high-throughput experiments (XRD, SEM, MOKE), to try and find new compositions of soft magnetic materials that have promising properties.

II. EXPERIMENT

A. Alloy design

Figure 2 gives the enthalpy of mixing for the different element pairs within the film sets studied, taken from the Bakker & Miedema paper.¹⁹ For the base elements, Co, Fe, Ni, and Mn, the enthalpy of mixing between different pairs such as Mn–Co and Mn–Fe, is close to zero or zero, tending toward solid-solutions forming in the films. The addition of the fifth element makes a substantial difference, as both Si and Al have large negative enthalpy of mixing with all four base elements. This can affect the distribution of the atoms within the alloy and may result in more ordered or intermetallic phases forming within the films.

Over the past 20 years, HEA research has attempted to understand their main properties, by linking them to fundamental variables, such as the atomic radius of each of the separate elements, the valence electron concentration (VEC), and the enthalpy of mixing between the different element pairs.^{4,20} From these

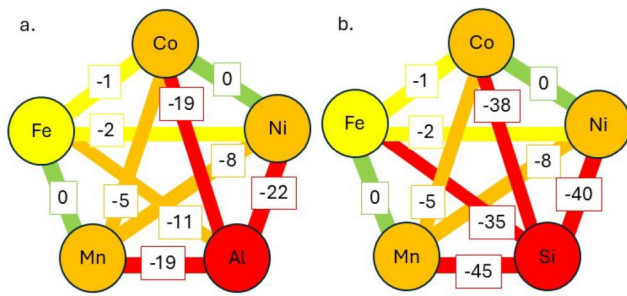


FIG. 2. Enthalpy of mixing in J/mol for (a) CoFeNiMn–Al and (b) CoFeNiMn–Si films studied.

fundamental variables, average values for each HEA are predicted, including the average VEC, average enthalpy of mixing, and the atomic radius difference. These empirical rules that have been proposed can be used to predict the phases present, or whether a solid-solution will form within the proposed HEAs.

Guo *et al.*²⁰ presented the concept that the average VEC of a HEA could predict the stability of solid-solution phase present in the HEA. The empirical rules for the VEC are that, for VEC > 8, the HEA is predicted to have a single FCC phase, while between 8 and 6.87, the HEA is predicted to have mixed BCC and FCC phases, and then for VEC < 6.87 the phase is predicted to be BCC. For the alloys studied in this paper, the VEC for the base composition (CoFeNiMn) assuming an equal ratio is 8.5, which means that the alloy is predicted to be FCC. The additional fifth elements added to the CoFeNiMn base, both have a VEC less than those of the base elements (Al VEC = 3, and Si VEC = 4), resulting in a lower average VEC and increasing probability that a BCC phase will form along with the FCC phase. Figures 3(a) and 3(c) show the predicted VEC for each of the different film compositions (at. %), along with the VEC boundaries (solid black lines) and the regions for the film compositions studied in this paper (dashed black lines). It is observed that the addition of Al should have a more dramatic effect on the phases present in the alloy, compared to Si. These changes in the phase could lead to changes in the magnetic properties. For the films studied in this paper, the majority of the compositions are predicted to be within the BCC + FCC mixed region (Tables II and III), with the edges close to the single-phase regions. As this is an empirical rule, not all HEAs fabricated will conform to the predicted VEC, as it does not take account of the synthesis process. This is because the synthesis process can change the phases present in the HEA. For example, Uporov *et al.*¹³ showed that the synthesis route of CoFeNiCrAl changed the phases present in the bulk alloy. The CoFeNiCrAl sample that was quenched had a majority BCC phase with the FCC phase being only just visible within the XRD, while the high-temperature annealed sample had both BCC and FCC phases present in roughly equal amounts. Similarly, the morphology of sputtered thin films depends strongly on the fabrication parameters such as target power and pressure,²¹ meaning that nonequilibrium phases can be produced. Thus, the average VEC can be taken as a guide to what could be expected for each composition.

Another empirical parameter used within the study of HEAs is the atomic radius difference (δr), which is the difference between the sum of the different element radii to the average atomic radius for the HEA. The following equation is used to determine it:⁴

$$\delta r = \sqrt{\sum_{i=0}^N c_i \left(1 - \frac{r_i}{\bar{r}}\right)^2},$$

where c_i is the concentration of element i , r_i is the atomic radius of element i , and $\bar{r}$ is the average atomic radius of the HEA. The atomic radius difference is normally given as a percentage, with the larger the δr the more likely the HEA will contain mixed phases. In the literature, there are a number of different values of δr below which a solid-solution phase has the probability of forming. Zhang *et al.*²² showed that the atomic radius difference could be used to suggest whether a HEA would have a solid-solution or contain intermetallics, although no numbers were given, they showed a larger δr gave rise to intermetallics. While work by Yang *et al.*²³ suggested that the boundary is $\delta r < 6.6\%$ for bulk alloys and Wiczerak *et al.*²⁴ suggested the boundary is $\delta r < 7.7\%$ for thin films grown via PVD. Furthermore, they note that the processing route can change the phases and morphology of HEAs (as noted above), and, therefore, δr is useful to use as a suggestion of whether a solid-solution, amorphous or intermetallics are formed. For this work, the boundary taken is $\delta r < 7.7\%$, where below it a solid-solution is expected, while for $\delta r > 7.7\%$ the HEA is more likely to have more than one phase present or be amorphous. The boundary was chosen as the work is mostly closely related to the work presented by Wiczerak.²⁴ For the two film compositions studied, the ternary plots for δr are given in Figs. 3(b) and 3(d), with the dashed line highlighting the compositional area studied in this paper and the solid line $\delta r = 7.7\%$. The van der Waals atomic radii were used within the calculations. It is observed for the CoFeNiMn–Al HEA, the larger δr occurs for high CoNiFe concentration, and low Al and Mn concentration. This is because the Ni atomic radius (163 pm) is much smaller than the other elements radii (Co—192 pm, Fe—194 pm, Mn—97 pm, and Al—184 pm). For all the CoFeNiMn–Al films down-selected in this study, δr was less than 7%, suggesting they should be solid-solutions, and not contain intermetallics. While for the CoFeNiMn–Si HEAs, the largest difference is observed for CoFeNi concentrations between 50 and 80 at. % with less than 10 at. % Mn and 10–30 at. % Si. This is because the atomic radius of Si is larger than the other elements (210 pm). The large difference in atomic radii between Ni and Si increases the radius difference within the compositions studied, such that δr is around the 7.7% boundary, suggesting that intermetallic phases may occur.

The choice of Al as the 5th element was made, based on the information in Fig. 1 where it is clear that Al helps maintain a high magnetisation, likely due to it driving the BCC phase in the alloys due to its low average VEC [Fig. 3(a)]. Furthermore, the availability of existing studies on both bulk and thin film CoFeNiMn–Al alloys^{8,25} enables a comparison of these results achieved from combinatorial sputtering with those in the

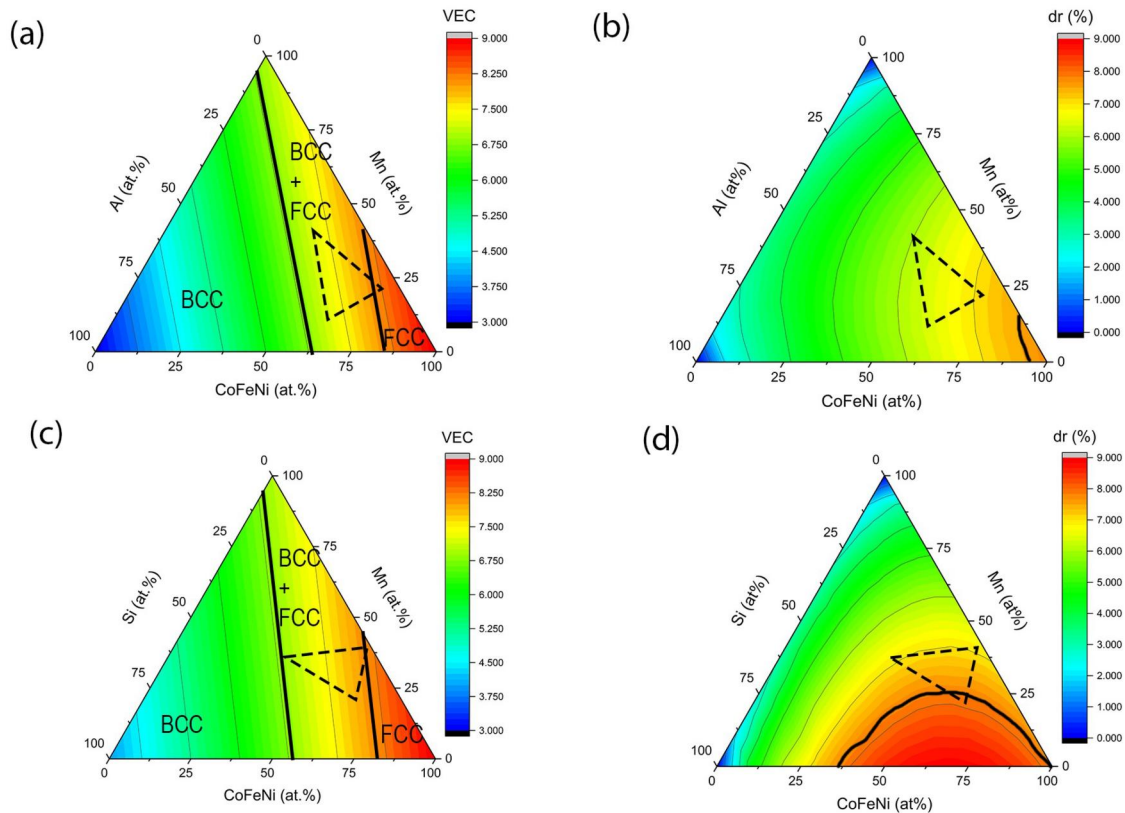


FIG. 3. (a) and (c) Ternary plots of the VEC for the CoFeNiMn-X films and (b) and (d) the atomic radius difference for the two different film compositions studied. The dashed lines show the compositional area studied in this paper. The solid lines in (a) and (c) are the VEC boundaries predicted for the different phase regions and in (b) and (d) they represent $\delta r = 7.7\%$.

literature, to validate this methodology. The choice of Si was made as there is very limited research on the structural and magnetic properties of the CoFeNiMn-Si alloy providing an exciting opportunity to demonstrate the high-throughput methodology once validated with the CoFeNiMn-Al alloy. Furthermore, HEA magnetic alloys maybe used in soft magnetic applications, where three of the requirements are a high saturation magnetisation (>800 kA/m), a near zero coercive field (<20 A/m), and a high resistivity ($>40 \mu\Omega$ cm).²⁶ The addition of Si is likely to increase the resistivity, thus if promising magnetic properties are measured, further resistivity measurements on the compositions of interest could be made.

B. Experimental methodology

All the films studied were grown using a 3-gun Lesker deposition system. Before growth, the 3-in. silicon wafers were cleaned using acetone and IPA. The targets used were an equimolar Co:Fe:Ni, a Mn and then either an Al or Si target. The films were grown at a pressure of 0.7 Pa, with the power on the target guns being 50 W for CoFeNi, 30 W for Mn, then 30 W for both Al and

Si. The films were grown for 20 min. The thickness of the films was calculated across the substrate using a combinatorial sputtering deposition model implemented in Python code, which was adapted from the Frisk²⁷ model of sputter deposition. The combinatorial sputtering deposition model takes account of the geometry of the sputter system including the power and the position of the three sputter guns in relation to the target wafer and required the experimental determination of calibration factors based on the target growth rates at the magnetron focal point. For this system the film thickness across the substrate for each combinatorial film was determined (Table I).

The combinatorial sputtering deposition model also predicts the compositions gradients across the substrate, which can then be used to tune the compositional region being looked at across the substrate. The composition regions are highlighted in Figs. 3(a) and 3(c), with the dashed lines on the ternary plots.

Compositional regions of interest can subsequently be cut out from the original 3-in. combinatorial sputtered sample. This was achieved by using the grid pattern associated with the MOKE measurements laid on top of the 3-in. wafer, so that the samples of interest were within 2 mm from MOKE position.

TABLE I. Predicted and measured element composition for each of the film set studied.

Film	CoFeNi (at. %)		Mn (at. %)		Al (at. %)		Thickness (nm)
	Predicted	Measured	Predicted	Measured	Predicted	Measured	
A1	41	39	40	39	19	22	218
A2	43	42	30	25	27	33	231
A3	53	54	31	27	17	19	182
A4	55	66	29	18	15	16	186
A5	61	64	24	17	16	19	212

Film	CoFeNi (at. %)		Mn (at. %)		Si (at. %)		Thickness (nm)
	Predicted	Measured	Predicted	Measured	Predicted	Measured	
S1	44	37	30	42	26	21	224
S2	46	47	32	35	22	18	233
S3	50	53	30	30	20	17	246
S4	53	55	29	29	18	15	256
S5	56	58	26	27	18	15	256
S6	61	63	22	23	17	14	227

The samples were $5 \times 5 \text{ mm}^2$. Table I gives the summary of the different CoFeNiMn–X films studied that were cut out from the original 3-in. wafer, including the thickness, the predicted compositions and the measured compositions using energy dispersive x-ray (EDX). It is observed that the predicted and actual compositions in most cases are within 5 at. % of each other, more commonly 1–2 at. % different, validating the combinatorial sputter deposition model implemented in this work. The film composition for areas around the edge of the film and near the clamps that held the wafer in the chamber, tended to have a larger difference between the model and the measured values, due to shadowing.

For both the combinatorial samples studied, the normalized hysteresis loops were measured on a high-throughput automated magneto-optic Kerr effect (MOKE) magnetometer, which measures a magnetisation versus magnetic field (M - H) loop at every 4 mm across the substrate. The max Kerr voltage, which is taken to be proportional to the magnetisation (M) as well as the coercive field (H_c), were determined from each hysteresis loop. These values were then outputted into two ternary diagrams (examples Figs. 4 and 8), which are then used to down-select compositions for further analysis. The shadowing effect that gives rise to differences between the composition predicted at the edges and the actual composition is the reason why there seems to be an abrupt

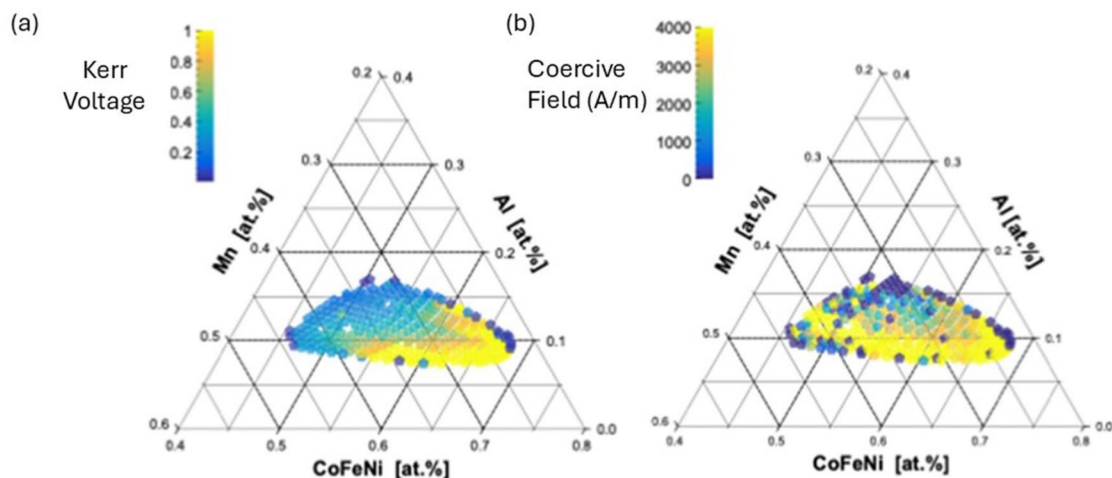


FIG. 4. Ternary plots of the (a) Kerr voltage and (b) coercive field determined from the MOKE magnetometer for the CoFeNiMn–Al combinatorial sputtered wafer.

change in magnetic properties at the boundaries within the ternary plots. For example, in Fig. 8(a), at around 60 at. % CoFeNi, the Kerr voltage seems to be 1 then 0 then 1 again.

X-ray diffraction (XRD) was used on these down-selected samples to determine the phases present within the different regions of the compositions produced. 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 was used. For any peaks measured, Bragg's law was applied to determine the lattice constant, and the Scherrer equation used to estimate the crystallite size.²⁸ The actual composition of the films was determined using EDX, along with backscattered electrons (BSE) and secondary electrons (SE) to determine whether any grains or topology existed in the films. For a selection of chosen compositions, a Quantum Design MPMS-3 SQUID magnetometer was used to measure the magnetization hysteresis loops at 300 K and magnetic fields up to 2 T, to determine the saturation magnetization. The magnetization as a function of temperature between 5 and 350 K at an applied field (either 0.025 or 0.05 T) was measured to study the magnetic behavior at low temperatures. To study the film surface, a Bruker Multimode 8 AFM in tapping mode was used to determine the surface roughness of the films, as this can be linked to the morphology and the grain size.

III. RESULTS AND DISCUSSION

A. CoFeNiMn-Al films

From Fig. 4, the CoFeNiMn-Al compositions that have a large Kerr voltage and, therefore, are likely to be strongly magnetic at room temperature, are those with less than 10 at. % Al or those with less than 35 at. % Mn. Further from the Kerr voltage ternary plot, it is observed that all the compositions studied are magnetic at room temperature. From the coercive field ternary plot, the coercive fields for the compositions with Al less than 10 at. % are the highest, with $H_c \sim 4 \text{ kA/m}$, increasing the Al concentration, results in a decrease in the coercive field. There is a region at around 13 at. % Al, 27 at. % Mn and 65 at. % CoFeNi, which has a high Kerr voltage and a coercive field 2–3 kA/m, which may be an interesting composition region for further investigation with regard to soft magnetic materials.

From Fig. 5(a) and Table II, it is observed that the CoFeNiMn-Al films all have a BCC phase, with film A1 also likely containing a FCC phase as well, due to the asymmetric peak at $2\theta \sim 44^\circ$. The black vertical lines in Fig. 5(a) are the expected peak positions for CoFeNi FCC phase with a lattice constant of 0.3582 nm ²⁹ and the HEA $\text{CoFeMn}_{0.5}\text{Al}_{0.5}$ BCC phase with a lattice constant of 0.289 nm .³⁰ The BCC (211) is at $2\theta \sim 81.5^\circ$, which is seen in all the films, with films A3 and A4 having clearer peaks. Also, for these films, there is a peak at $2\theta \sim 55.1^\circ$ just above the noise, which is likely to be the BCC (111) peak, suggesting that these compositions have ordered BCC structures (B2 phase). Due to the low signal to noise ratio, it is not possible to tell whether the films only have an ordered BCC phase or both an ordered and disordered BCC phases, which have been observed previously in bulk HEAs. Typically, the disordered BCC phase is observed within thin magnetic films containing Co and Fe,³¹ while the B2 phase has been previously observed within bulk HEAs including

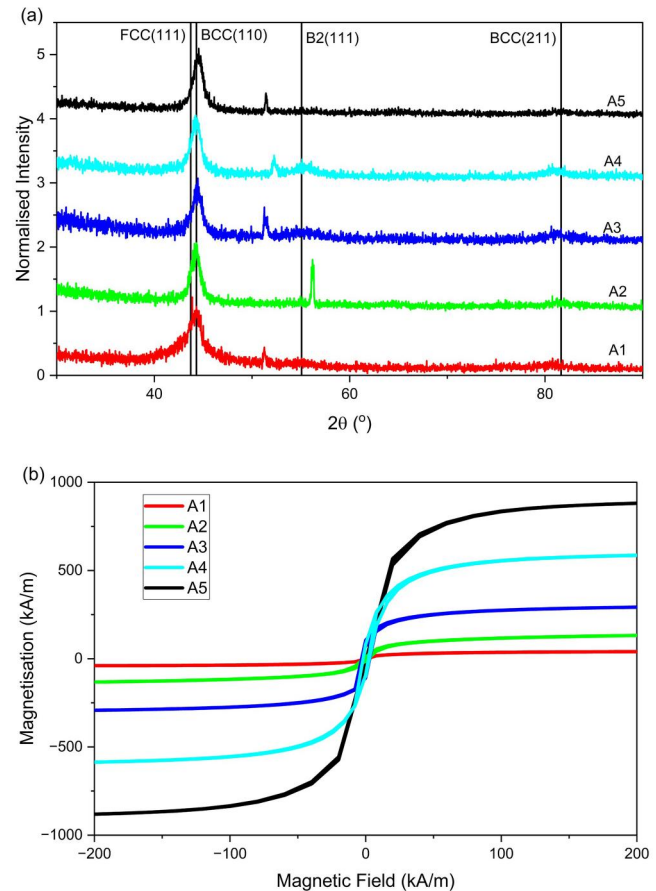


FIG. 5. (a) XRD of the five CoFeNiMn-Al films taken from the larger combinatorial sputtered wafer. (b) Magnetization as a function of magnetic field for the five CoFeNiMn-Al films studied.

CoFeNiCrAl_x ³² and CoFeNiMnCrAl ,³³ and binary alloys such as Fe-Al.³⁴ The addition of Al drives the ordered BCC phase formation within HEAs. To fully confirm this ordered phase and whether there is also a disordered BCC phase, higher resolution experiments, such as TEM would be required.

The films A1, A3, and A5, all have a sharp peak at $2\theta \sim 51.5^\circ$, fitting this peak in Highscore software, the peak appears to correspond to cubic Fm-3m compounds, but it is difficult to determine more accurately, which one due to the higher reflections at $\sim 75^\circ$ and $\sim 90^\circ$ being obscured in the noise. The “best” fit using the CIF files within Highscore for A1 was $\text{Al}_{0.06}\text{Co}_{0.39}\text{Fe}_{0.4}\text{Ni}_{0.15}$ (record: 04-030-1280), and for A3 and A5 it was $\text{Co}_{0.1}\text{Mn}_{0.4}\text{Ni}_{0.6}$ (record: 04-017-6736). Film A4 has a peak at a slightly higher angle (52.3°), again fitting to this peak without additional higher intensity peaks was difficult, with the closest match being P4/nm tetragonal FeNi (record: 01-083-3765). For the A2 film, no peak is observed at $2\theta \sim 51.5^\circ$, suggesting that for this composition no cubic Fm-3m occurred, which agrees with the prediction from the average VEC,

TABLE II. Radius difference, average enthalpy of mixing, VEC, predicted phases, actual phase, lattice constant, and crystallite size for the CoFeNiMn–Al films.

Film	Radius difference (%)	Average enthalpy of mixing (J/mol)	VEC	Predicted phase	Actual phase	Lattice constant ± 0.001 nm	Crystallite size (± 1 nm)
A1	5.7	−15.2	6.89	BCC + FCC	BCC FCC	0.29 0.358	7
A2	5.8	−18.	6.53	BCC	BCC	0.289	6
A3	6.4	−12.1	7.34	BCC + FCC	BCC	0.289	6
A4	6.5	−12	7.65	BCC + FCC	BCC	0.29	7
A5	6.7	−13.5	7.54	BCC + FCC	BCC	0.288	6

that only a BCC phase should exist. The A2 film does have an additional peak at $\sim 55.8^\circ$, which was very difficult to match to, again due to the absence of higher intensity peaks, with the closest match being hexagonal $\text{Al}_{0.75}\text{Fe}_{0.25}$ (record: 03-065-3654). These peaks suggest that there are second phases present within the films, but due to the low signal to noise ratio, it is very difficult to determine what these phases are and how much of each is present.

From HEA VEC theory, it was predicted that all the films apart from A2 should contain both BCC and FCC phases, the XRD data suggest that the predominant phase is a nanocrystalline BCC phase in four of the films, with the possibility of a minority cubic Fm-3m phase in three of them, while film A1 contained both phases. This agrees with the atomic radius difference prediction, that HEAs with δr less than 7.7% will be a solid-solution. The A2 film had the most negative average enthalpy of mixing, and the smallest VEC, and was the only film not to have a peak that could be associated with a cubic Fm-3m phase, rather its additional peak was possibly an intermetallic phase. The crystallite sizes are all similar, which is to be expected as they are all grown on the same wafer, with the same growth conditions. The EDX maps of the different elements for the CoFeNiMn–Al films ([supplementary material](#)) suggest that all the elements are homogeneously mixed, within the spatial resolution of the EDX instrument with no clear spatial differences between the elements. Noting that the grains within the films are less than 50 nm (Fig. 6), which is smaller than the interaction volume of the EDX. Thus, unlike in bulk alloys where mixed phases can occur, and are observed within EDX images due to the microstructure being larger than $1\text{ }\mu\text{m}$ (i.e., larger than the EDX resolution), EDX and XRD for these combinatorial films do not have high enough resolution to resolve nanostructures and multiple phases.

From the BSE images it is observed that the A5 film surface is cracked [Fig. 6(a)], with spherical grains at the surface. Typically for magnetic films such as FeCo and FeCoCr,³⁵ the surface roughness is in the range of 0.5–2 nm, with no cracking observed, thus these features were studied in more detail using an AFM [Figs. 6(b) and 6(c)] for the A5 film. From the BSE and AFM images the average width of the cracks were determined to be between 50 and 60 nm. While the average grain size width was 33 ± 7 nm, with an average height of 5 nm. The difference between the grain size measured from the SEM and AFM images (Fig. 6) and the crystallite size calculated from the XRD peaks (Table II), is often observed in thin films, due to the XRD peak being

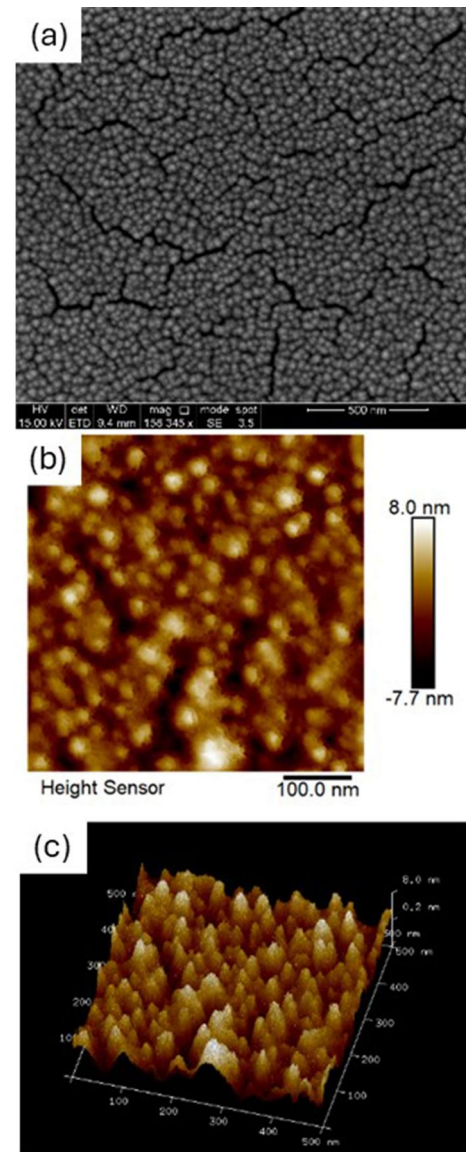


FIG. 6. For the film A5. (a) Backscattered electron image (BSE), (b) 2D and (c) 3D AFM images of the surface.

broaden due to inhomogeneous strain with the films.²¹ The increase in the width of the XRD peak, due to the strain means that the calculated crystallite size from the Scherrer equation within the film is often smaller than the actual grain size.

The magnetization hysteresis loops were measured on a SQUID, to determine the saturation magnetisation [Fig. 5(b)]. These saturation magnetizations were then plotted against the CoFeNi–Mn–Al (at. %) concentrations [Fig. 9(a)] along with the CoFeNiMn–Si films from the next section]. To verify that the combinatorial films give the same functional properties as single composition films, the magnetizations are compared with the literature, for films with similar compositions. Alsebaie *et al.*²⁵ single composition CoFeNiMn–Al films were grown systematically changing either the Al concentration or the Mn concentration. For their film with 46 at. % CoFeNi, 34 at. % Mn, and 20 at. % Al, they measured $M_s = 20$ kA/m, which has a similar composition to the A1 film and is in good agreement with the saturation magnetization. While their 59 at. % CoFeNi, 18 at. % Mn, 23 at. % Al film had $M_s = 600$ kA/m, which is in good agreement with the combinatorial A4 film. This verifies that the combinatorial films method can be used to study HEA films, with the functional properties measured in good agreement with single composition grown films. It is also observed that for films with less than 50 at. % CoFeNi, the saturation magnetization is less than 100 kA/m, but from 50 at. % to 70 at. % CoFeNi concentration the saturation magnetization increases up to 900 kA/m. As expected the highest magnetisations is for Mn < 20 at. % and Al < 20 at. %. The coercive fields measured on the SQUID magnetometer ranged from 0.74 kA/m (A2 and A5) to 3 kA/m (A3). Comparing them with the values determined from the high-throughput MOKE measurement [Fig. 4(b)], it was found that there is good agreement between the two measurements, demonstrating that the high-throughput MOKE is exact enough to down-select compositions of interest.

B. CoFeNiMn–Si films

For the CoFeNiMn–Si films, the range of compositions is within 40–65 at. % magnetic elements, from the ternary plots (Fig. 7) it is observed that the majority of the compositions are weakly magnetic at room temperature, with the highest Kerr voltage being measured for the highest concentration of magnetic elements. Most of the coercive fields measured are less than 3 kA/m so in the soft to semi-soft magnetic regions. For this combinatorial film composition, it was decided to study six films in the compositional range from CoFeNi:Mn:Si = 1:1:1 to 3:1:1, i.e., film S6 is roughly equimolar for all the elements.

From the XRD data [Fig. 8(a)], it is observed for films S1, S2, S3, there is a shallow amorphous hump at around 48° . This is likely to be due to both the Mn and Si being added. Previous work on CoFeNiMn films, found that the addition of Mn to CoFeNi caused the morphology to change from a FCC phase for CoFeNi to an amorphous phase for CoFeNiMn.²⁵ While Si is often used to promote amorphous and nanocrystalline morphologies in soft magnetic alloys, such as FeSiB (Metglas) and Finemet. Although there is a higher concentration of Si within these films, no additional phases were observed.

For films S4, S5, and S6, the compositions are close to equimolar, this means that the amount of Mn in the samples has been reduced, and the large negative enthalpy of mixing between Si and the other elements could dominate the phases (Fig. 2). Two further peaks are observed above the noise the first at $\sim 35.4^\circ$ and the second at $\sim 54^\circ$. These peaks are likely to be due to two different intermetallic phases that Si forms with Fe, Co, and Mn separately or in mixed combinations. The two of the possible intermetallic cubic phases that could form are ϵ -MSi and M_3 Si, where M is either the single element or a combination of the elements such as FeCo_2Si .³⁶ For ϵ -MSi, the main peaks occur at $20 \sim 35.4^\circ$ (111), 46.1° (210), and $\sim 51^\circ$ (211). As these are thin

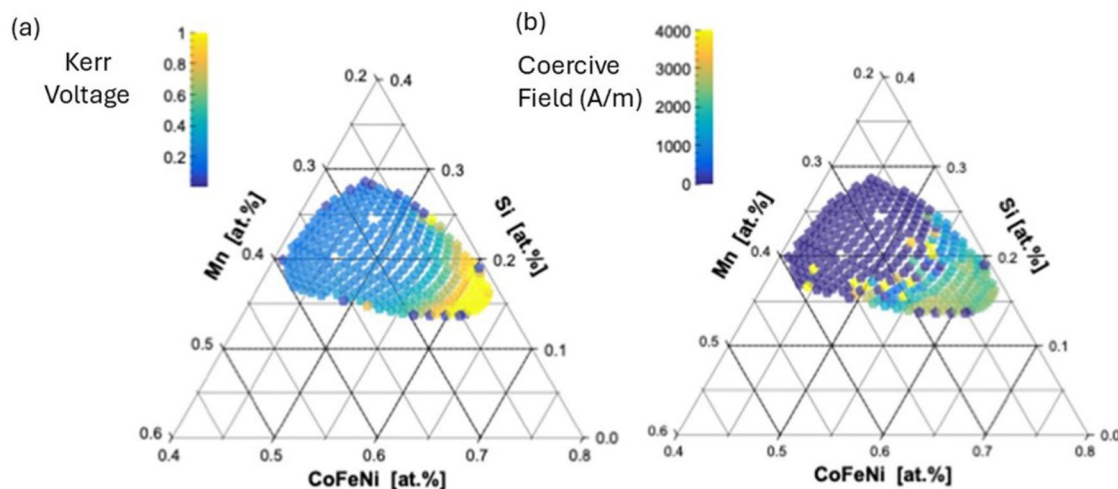


FIG. 7. Ternary plots of the magnetic properties determined from the automated MOKE magnetometer for the CoFeNiMn–Si combinatorial sputtered wafer, (a) Kerr voltage and (b) coercive field.

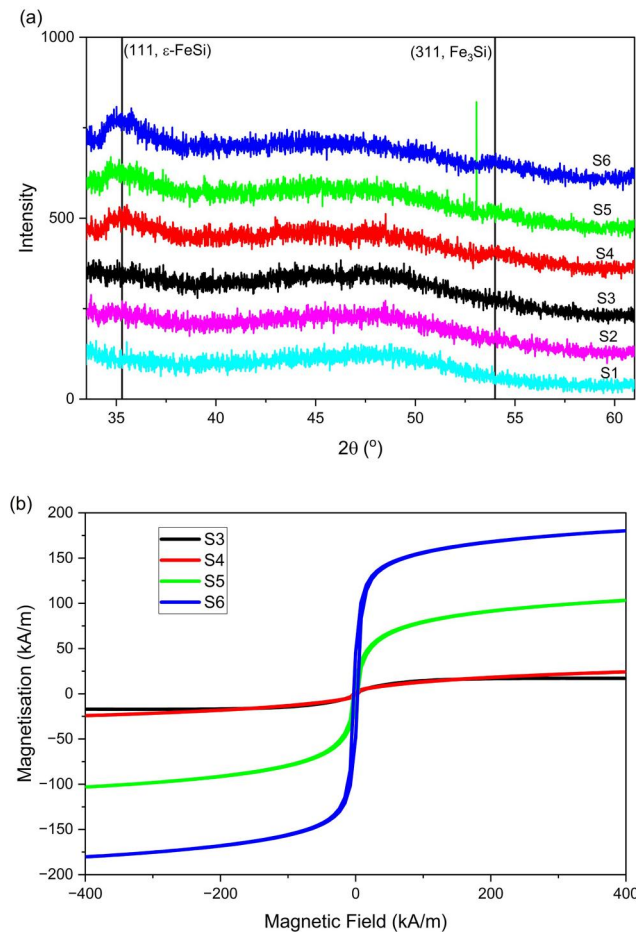


FIG. 8. (a) XRD of the six selected CoFeNiMnSi films and (b) magnetization as a function of the magnetic field for the CoFeNiMn–Si films measured on a SQUID magnetometer.

films, there could be some preferred orientation growth within the films, which might be why only the (111) peak is observed, with the (210) within the broad amorphous hump. For M_3Si , the main peaks occur at $2\theta \sim 31.8^\circ$ (200), 45.6° (220), and $\sim 54^\circ$ (311), again preferred orientation growth could be the reason why only the (311) peak is observed, while the (220) peak is likely to be within the amorphous hump. To fully confirm the presence of these intermetallic phases, a higher resolution technique such as TEM would have to be used. Previous work on CoFeNi films²⁵ found that the preferred orientation growth for this sputter system was along the (111) direction, with no other peaks observed. An estimate of the lattice constants of the ϵ -MSi and M_3Si phases were calculated within the FITYK³⁷ software. Due to the signal to noise ratio being low, the error on the lattice constants is larger than those calculated for the CoFeNiMn–Al films. This means that the lattice constant determined for each phase for each film is the same. The lattice constants³⁶ are in agreement with the bulk lattice constants of ϵ -MSi and M_3Si , where for ϵ -MSi the lattice constant ranges from 0.438 nm for CoSi to 0.45 nm for NiSi, and for M_3Si the lattice constant for Fe_3Si is 0.563 nm and for Mn_3Si is 0.564 nm.

As the as-grown CoFeNiMn–Si films all have an amorphous structure, this means that during the growth, the equilibrium crystal structure of the compositions has not been reached. To achieve this, the films either could be grown at an elevated temperature between 100 and 200 °C or annealed after growth, although this could also promote the intermetallic phases observed in films S4, S5, and S6.

From Table III, it is observed that the average VEC is not as good predictors for thin films, as they are for bulk alloys, to predict which the phases are present in the films, as the films' morphology was either amorphous or mixed phase with no solid-solution phases observed. This is likely due to the films not being in an equilibrium state. However, the compositions with an average enthalpy of mixing above -25 J/mol were amorphous, while those with a less negative enthalpy of mixing were amorphous with intermetallic phases present. For the two films (S5 and S6), where the atomic radius difference was close to or above 7.7%,

TABLE III. Atomic radius difference, average enthalpy of mixing, VEC, predicted phases, actual phase, and lattice constant for the CoFeNiMn–Si films.

Film	Radius difference (%)	Average enthalpy of mixing (J/mol)	VEC	Predicted phase	Actual phase	Lattice constant (nm) (± 0.01 nm)
S1	6.55	−30.4	7.1	BCC + FCC	Amorphous	—
S2	7.37	−27.4	7.42	BCC + FCC	Amorphous	—
S3	7.48	−26	7.56	BCC + FCC	Amorphous	—
S4	7.37	−23.7	7.12	BCC + FCC	Amorphous	—
					ϵ -FeSi	0.44
					Fe_3Si	0.56
S5	7.67	−23.7	7.23	BCC + FCC	Amorphous	—
					ϵ -FeSi	0.44
					Fe_3Si	0.56
S6	7.88	−22.3	7.4	BCC + FCC	Amorphous	—
					ϵ -FeSi	0.44
					Fe_3Si	0.56

the films had additional intermetallic phases present, along with an amorphous morphology. This suggests that in HEA thin films the atomic radius difference and the average enthalpy of mixing may be good predictors of whether a film will be solid-solution or contain intermetallics, plus if Mn and Si are included in the composition, then the films have a higher chance of being amorphous. For the CoFeNiMn–Si films studied, films S1 and S2 were not magnetic at room temperature, this will be due to the nonmagnetic content of the films' composition being greater than 50 at. % nonmagnetic. The S3 film was ~50 at. % magnetic to ~50 at. % nonmagnetic and had a very small magnetisation (~17 kA/m) at room temperature, with a very small coercive field [Fig. 8(b)]. For the S4, S5, and S6 films, they were magnetic, with an increase in saturation magnetization as the magnetic element content increased. The coercive fields measured from Fig. 8(b), were ~3 kA/m for S4, S5, and S6 films, which again are in good agreement with the coercive fields measured for those compositions using the high-throughput MOKE. As the coercive fields are greater than 1 kA/m and the magnetisations are less than 800 kA/m, it is unlikely that these alloys would be considered as possible candidates for soft magnetic materials.

C. Comparison of CoFeNiMn–X films

For both film sets studied, it is possible to suggest some general trends observed for combinatorial CoFeNiMn–X sputtered films. Looking at the structural and HEA parameters in Tables II and III, the CoFeNiMn–Al films had a smaller radius difference, and a smaller average negative enthalpy of mixing compared to the CoFeNiMn–Si films, while the VECs were all similar in the range of 6.5–8. The CoFeNiMn–Al films were polycrystalline, with a dominant BCC phase, while the CoFeNiMn–Si films were predominately amorphous, with Si based intermetallic phases appearing for the larger radius difference compositions. This suggests that in the combinatorial HEA thin films, the morphology present in the grown films can be predicted better using the atomic radius difference rather than the average VEC.

Comparing the saturation magnetization as a function of composition for both film sets and considering CoFeNiMn bulk value is 150 kA/m (Ref. 8) [Fig. 9(a)], it is observed that the magnetization of HEA films does not solely depend upon the concentration of CoFeNi within the alloy. For both CoFeNiMn–Al and CoFeNiMn–Si films, the magnetization is greater than the CoFeNiMn bulk alloy value for concentrations of CoFeNi greater than 60 at. %, clearly demonstrate that the combination of the elements is important, as well as the amount of the magnetic elements. Further, similar magnetizations were measured in both film sets as the CoFeNiMn alloy, with 50 at. % of magnetic elements, so 50 at. % of nonmagnetic elements. Previous work has shown that the phase (BCC or FCC), makes a large difference to the magnetization measured,¹⁶ with HEAs with a BCC phase tending to have higher magnetisations than those with a FCC phase, even if the FCC phase has a higher concentration of magnetic elements. For the CoFeNiMn–Al films, the increase in magnetization is likely to be due to the BCC structure within the films, as CoFeNiMn has a FCC structure. While for the CoFeNiMn–Si films, the morphology is amorphous and, therefore, suggesting

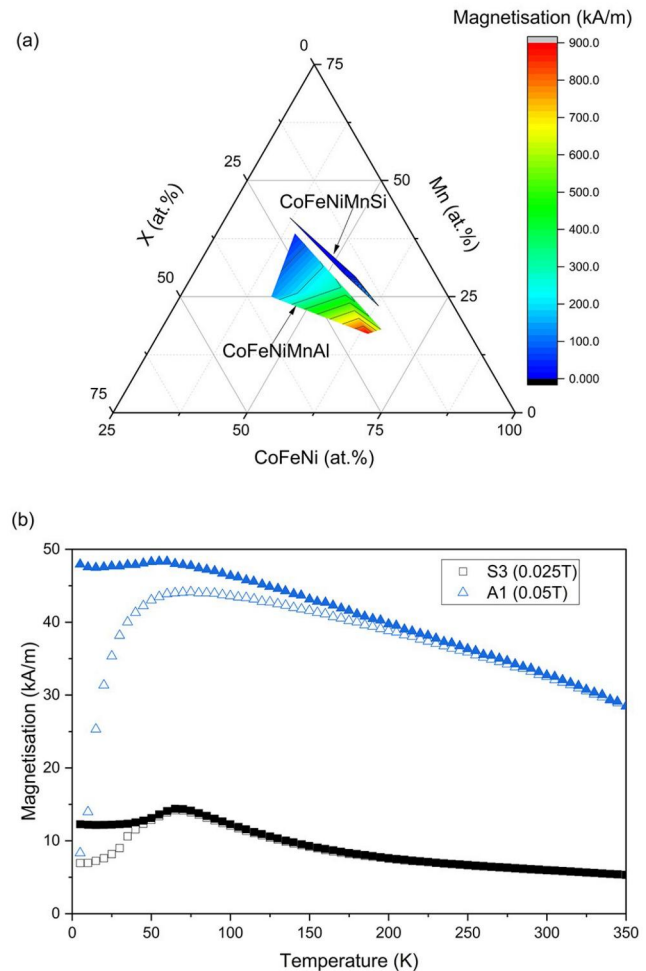


FIG. 9. (a) Saturation magnetization at 300 K as a function of composition, where X either is Al or Si. (b) Magnetization as a function of temperature for film A1 at the 0.05 T applied field and film S3 at the 0.025 T applied field. The open shapes are for the ZFC and the closed shapes for the FC.

either the Mn or the FCC phase causes the low magnetization of the base alloy.

Magnetic high entropy alloys could be used in a few different applications, such as soft magnetic materials, where high saturation magnetization and small coercive field along with high resistivity are important for applications,³⁸ or they could be used within magnetic refrigeration.³⁹ The majority of the work studying HEAs for magnetic refrigeration has focused on room temperature changes, where there has been success in achieving relatively competitive changes in entropy.⁴⁰ Another area of interest for magnetic HEAs is their magnetic low temperature behavior, where due to either formations of nanoparticles or clustering of atoms unusual low temperature behavior is observed. Therefore, two films (A1 and S3) whose magnetization was smaller than 150 kA/m at 300 K, so only weakly magnetic at room temperature,

were measured on the SQUID to study the magnetization as a function of temperature. From Fig. 9(b), it is observed for the zero-field cooled (ZFC) magnetization as a function of temperature, both films show an increase in the magnetization to a peak before decreasing. While for the field cooled (FC) magnetization data, the magnetization follows the ZFC magnetization from 300 K to the peak, before slightly decreasing and plateauing. The peaks in the ZFC data occur at 70 K for film A1 and 65 K for film S3 and is known as the blocking temperature. Two different reasons can cause this behavior, the first is the compositions are magnetic spin glasses, which has been observed in amorphous rare-earth HEA alloys⁴¹ and the second is that the films contain magnetic nanoparticles within a matrix, which has been observed in bulk CoFeNiCrAl alloys.⁴² As the CoFeNiMn-Si (S3) film is amorphous, the behavior is likely to be due to this composition being spin-glasslike. While for the CoFeNiMn-Al (A1), it is less clear of the origin, as the film is polycrystalline, with the XRD showing a majority BCC phase and a minority FCC phase, plus the surface roughness (Fig. 6) shows grains of ~33 nm. These grains might behave as nanoparticles at low temperatures, leading to the observed blocking temperature or similar to film S3, the blocking temperature could be due to a spin-glasslike behavior.

IV. SUMMARY AND CONCLUSIONS

High-throughput methodology involving combinatorial sputtering and high-throughput MOKE magnetometry and XRD provides a quicker method to identify compositions of interest within a wide compositional range. The method has been demonstrated for the CoFeNiMn-Al alloy, where the saturation magnetisation measured for the films at a range of compositions matched well with those in the literature for both bulk and thin films, showing that the high-throughput method can be trusted.

Furthermore, the alloy CoFeNiMn-Si was investigated, as it had not been studied before. It was determined that at lower Si content, second phases appeared, this could be due to the atomic radius difference being larger than 7.6%, while at higher Si content, the films were amorphous, likely due to the large negative enthalpy of mixing. For these films, the saturation magnetisation was higher for the lower Si compositions. For the six compositions studied, none had promising soft magnetic properties, as the saturation magnetization was too low and the coercive fields were greater than 1 kA/m.

From both film sets, comparing the empirical prediction parameters (average enthalpy of mixing, average VEC, and atomic radius difference), the atomic radius difference seems to be able to predict better as to whether a solid-solution, multiphase intermetallic or amorphous film is grown. It has also been shown that the magnetization of CoFeNiMn-X alloys does not solely depend upon the amount of CoFeNi within the composition, rather it is more complex involving the phases present and the other non-magnetic elements added.

SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for backscattered electron (BSE) and energy-dispersive x-ray (EDX) image of the

CoFeNiMn-Al films (Fig. S1). Backscattered electron (BSE) and energy-dispersive x-ray (EDX) image of the CoFeNiMn-Si films (Fig. S2).

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Nicola A. Morley: Conceptualization (equal); Data curation (equal); Funding acquisition (equal); Methodology (equal); Supervision (equal); Writing – original draft (equal). **Richard M. Rowan-Robinson:** Conceptualization (equal); Funding acquisition (supporting); Investigation (equal); Methodology (lead); Writing – review & editing (equal). **Tom Wilkinson:** Investigation (equal). **Jing Bai:** Conceptualization (equal); Funding acquisition (supporting); Investigation (equal); Methodology (lead); Writing – review & editing (equal). **Jianglong Gu:** Data curation (supporting); Funding acquisition (supporting); Investigation (equal); Methodology (supporting); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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