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Highly Tunable Magnetic and Mechanical Properties in an $\text{Al}_{0.3}\text{CoFeNi}$ Complex Concentrated Alloy

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Abstract

Electrical rotating machines, including motors, account for a significant portion of total energy consumption in the world. Improving the magnetic materials used in motors is a key challenge to increase their performance. Specifically, higher rotation frequency requires appropriate site specific magnetic properties as well as good mechanical properties. Hence, we studied both the magnetic and mechanical properties of an $\text{Al}_{0.3}\text{CoFeNi}$ complex concentrated alloy (CCA). Heat treatment, guided by phase diagram modeling, was employed to develop a novel eutectoid-like nano-lamellar (FCC+ L_{12}) / (BCC+B2) microstructure as well as a coarser FCC+B2 microstructure. The coarser microstructure exhibits soft magnetic properties with saturation magnetization (M_s) of ~ 127 emu/g, coercivity (H_c) of ~ 151 A/m and microhardness of ~ 195 VHN. On the other hand, the semi-hard nano-lamellar microstructure exhibits $M_s \sim 138$ emu/g, a high $H_c \sim 12732$ A/m and a very high microhardness ~ 513 VHN. This corresponds to more than eighty times increase in H_c and double the hardness in the same alloy. These results demonstrate the feasibility of producing a range of mechanical and magnetic properties by thermo-mechanical

treatment of a single CCA composition, making them potential candidates for metamorphic manufacturing.

Keywords: Complex Concentrated Alloys; High Entropy Alloys; Nano-lamellar microstructure; Eutectoid decomposition; Magnetic properties

1. Introduction

Metamorphic manufacturing is a novel manufacturing approach to produce engineering components [1], which relies on incremental forming to achieve site-specific properties dictated by local microstructures. This new manufacturing approach requires materials whose properties can be varied over a wide range via thermomechanical treatment. Complex concentrated alloys (CCA) are potentially suitable materials for such manufacturing methods since the structural and functional properties of a single alloy composition can be varied over a wide range by suitable processing strategies [2, 3]. In 3d transition (Co, Cr, Fe, Ni) based CCAs property manipulation is often achieved by systematically changing the phase stability and microstructure. Alloying elements such as Al and Ti can also be added both these elements have large atoms and can induce chemical ordering. With increasing Al content in such CCAs, the phases present typically change from single phase FCC (or FCC + L1₂), to a two phase FCC + BCC mixture (possibly also with L1₂ and/or B2 precipitates), and eventually to BCC + B2 [4-6]. This effect of changing composition on the phase stability and consequently on the mechanical and magnetic properties has been previously demonstrated in Al_xCoCrFeNi [7], Al_xCuCrFeNi₂ [8], and AlCo_xCr_{1-x}FeNi [9] CCAs. Chang and Yeh [10] reported microstructural changes with varying Al content and its influence on high temperature mechanical properties in Al_xCo_{1.5}CrFeNi_{1.5}. Yang et al. [11]

recently developed an $\text{Al}_{0.25}\text{Ti}_{0.25}\text{CoFeNi}$ CCA, based on the medium entropy CoFeNi system, with a high $L1_2$ volume fraction, this composition exhibited an excellent combination of tensile yield strength and ductility. These results suggest that we can systematically tune the mechanical as well as magnetic properties in Al containing CoFeNi CCAs. Hence, we selected an $\text{Al}_{0.3}\text{CoFeNi}$ alloy composition and employed systematic heat treatment to vary the microstructure. This microstructural variation was shown to dramatically influence magnetic coercivity and hardness, while retaining the magnetization values.

Soft magnetic materials are vital for many applications such as electrical power generation, rotating electrical machines, magnetic shielding, electromagnets etc. The global soft magnetic materials (SMM) market was ~ \$47 billion in 2018, and is expected to reach ~\$87 billion by 2026. SMM are vital in power electronics systems such as inductors, transformers, generators, electric motors. Electric Power Research Institute (EPRI) estimated that 60% to 65% of electrical energy is consumed by motor drive applications. Emerging electrical transportation systems also require better electrical conversion systems. Developing high performance, high frequency, high temperature electric motors require magnetic materials possessing high mechanical strength and good magnetic properties [12-21].

The power of an electrical rotating machine is the product of the speed, thermal utilization, magnetic utilization and volume [22]. This implies that we need a material with high magnetic saturation, low core loss, high thermal conductivity and high strength. Conventional metallic SMM possess good magnetic properties but suffer from poor strength. Further, in rotors, the mechanical strength needs to be higher on the periphery compared to the central regions due to the larger

centrifugal stresses. Site specific properties will facilitate spatial optimization of the properties. Metamorphic manufacturing [1] can achieve site-specific properties by changing the local microstructures in a single material. Here, we demonstrate a large tunability in the magnetic and mechanical properties of $\text{Al}_{0.3}\text{CoFeNi}$ CCA via thermo-mechanical processing. This study could support in designing a next generation electric motor where site specific properties within a same material is highly desired.

Semi hard magnetic materials with high mechanical strength can also be useful in other applications, e.g., electronic article surveillance materials [23]. CCAs can open up a materials space exhibiting promising combinations of mechanical strength and magnetic properties. There have been a few investigations relating the Al content to the magnetic properties of CCAs. Examples include $\text{Al}_x\text{CoCrFeNi}$ alloys exhibiting ferromagnetic behavior for $x = 0.5, 1.25$ and 2 , and paramagnetic behavior for $x = 0, 0.25$ and 0.75 [24]. Zuo et al.[25], tuned the magnetic properties by changing the Al content in Al_xCoFeNi alloys, the H_c was found to be ~ 215 A/m and 344 A/m for $x = 0.25$ and 0.50 , respectively. Despite these previous reports it is important to note that metamorphic manufacturing will require alloys whose properties can be substantially changed via thermo-mechanical processing without changing the alloy composition. This study demonstrates that for a single $\text{Al}_{0.3}\text{CoFeNi}$ CCA, guided by thermodynamic modeling, the type and morphology of the precipitates in the complex multi-phase microstructure can be tuned via thermo-mechanical processing. This leads to large range of microhardness values (195 VHN to 513 VHN), and a very wide range of coercivity values (151 A/m to 12732 A/m). The extremely high microhardness and coercivity can be attributed to a novel eutectoid-like nano-lamellar microstructure.

2. Experimental Procedure

$\text{Al}_{0.3}\text{CoFeNi}$ alloy was produced using conventional arc melting. The composition of the cast ingot was found to be 9.4Al-30.1Co-31.9Fe-28.6Ni (at. %) by Energy dispersive spectroscopy in a Scanning electron microscope (SEM-EDS). The cast alloy was homogenized at 1250°C for 30 min, followed by 85% reduction in thickness by cold rolling. The samples were then encapsulated in quartz tubes backfilled with argon for subsequent heat treatments. The rolled samples were solution annealed at 1250°C for 5min (CRSA condition), followed by heat treatment at either 600°C for 50 h (CRSA600) or 700°C for 50 h (CRSA700). Some samples were only heat treated at 700°C for 50 h after cold rolling (CR700). Here, the acronym CR stands for cold rolled to 85% reduction in thickness and SA corresponds to solution annealing treatment at 1250°C for 5 min. The temperature for solutionizing, to obtain a single phase microstructure was chosen based on the isopleth in Fig. 1(a). Microstructural characterization was performed using a FEI Nova-NanoSEM 230™ Scanning electron microscope coupled with an energy dispersive spectrometer (EDS) and Hikari Super Electron Backscattered Diffraction (EBSD) detectors, FEI Tecnai G2 TF20™ transmission electron microscope operated at 200 kV and a Cameca LEAP 5000XS 3D Atom Probe Microscope operated at 30K with a pulse fraction of 20% and detection rate of 0.5 in Laser mode. Microhardness measurements were carried out in a Vickers microhardness tester using a load of 500 gf. The magnetic properties were measured using a Lakeshore 7404 Vibrating Scanning Magnetometer (VSM).

3. Results and Discussion

3.1 Phase stability modeling

An isopleth for the Al_xCoFeNi system was calculated using Thermocalc (TCHEA3 database) and is shown in Fig. 1(a). This isopleth can be treated as a pseudo-binary section of the complex phase diagram. The composition of $\text{Al}_{0.3}\text{CoFeNi}$ was chosen since this composition traverses several phase fields at different temperatures. Two different heat treatment temperatures were identified, 600°C and 700°C . The isopleth predicts a substantial change in phase stability from a three-phase FCC + L1_2 + B2 region at 600°C to a two-phase FCC + B2 region at 700°C . The calculated phase fractions as a function of temperature is shown in Fig. 1(b).

3.2 Scanning Electron Microscopy

In the single phase CRSA condition, large recrystallized grains ($\sim 150\ \mu\text{m}$) of the FCC phase were observed with no intragranular or grain boundary precipitation, Fig. 2(a). A backscatter SEM image from the CRSA600 condition is shown in Fig. 2(b). The predicted phase stability at this temperature of 600°C , is a three-phase FCC+ L1_2 +B2 field (refer to Fig. 1). The image in Fig. 2(b) reveals a highly refined lamellar microstructure in all the grains, resembling the pearlite-like microstructure observed in steels. The inset in Fig. 2(b), recorded at higher magnification, indicates further decomposition within these lamellae.

The CRSA700 and CR700 samples correspond to heat treatment at 700°C in a different region of phase stability (FCC + B2). A backscattered SEM image from the CRSA700 condition is shown in Fig. 2(c), revealing recrystallized FCC grains, similar in size to those observed in the CRSA sample. The higher magnification inset in the same figure shows a small fraction of grain boundary precipitates, which are likely to be B2 precipitates [26]. Previous reports on $\text{Al}_{0.3}\text{CoCrFeNi}$ rationalized that the high nucleation barrier of the B2 phase within the FCC matrix makes it rather

difficult to homogeneously nucleate intra-granular B2 precipitates, favoring heterogeneous nucleation at grain boundaries [2, 26]. Therefore, the number density of heterogeneous B2 nucleation sites has been increased by cold-rolling the sample to 85% reduction in thickness, followed by heat treatment only at 700°C for 50 h (CR700 condition). Fig. 2(d) shows a backscatter SEM image of a CR700 condition sample. The microstructure consists of substantially refined FCC grains compared to the CRSA700 condition with coarse grain boundary precipitates in darker contrast (inset shows a high magnification image). Coupled EBSD-EDS analysis of this microstructure (Fig. 3) identifies the darker contrast regions as BCC based precipitates, based on the phase map (they were found to be B2 precipitates based on subsequent TEM analysis) with a volume fraction of ~21%. The EDS mapping in Fig. 3(b) shows the enrichment of Al in grain boundary precipitates. The refined FCC matrix grains in the CR700 condition samples compared to the CRSA700 condition samples can be attributed to the concurrent recrystallization of the matrix grains and B2 precipitation occurring in CR700 samples, pinning the growing FCC grains.

3.3. Transmission Electron Microscopy

Further characterization of the CR700 and CRSA600 conditions has been carried out via transmission electron microscopy (TEM). Fig. 4(a) shows a bright-field TEM image of a sample in the CR700 condition revealing multiple FCC grains and a precipitate in the center. The diffraction pattern obtained from the [001] zone axis of the BCC based precipitate exhibits {100} superlattice reflections (inset in Fig. 4(a)), confirming that the crystal structure is B2. In the CRSA600 condition, this alloy exhibits a periodic lamellar structure (lamellae thickness of ~100-150 nm) as revealed by the bright field TEM micrograph shown in Fig. 4(b). Electron diffraction analysis revealed that {011} BCC // {111} FCC and <001> BCC // <011> FCC, indicating the

Nishiyama-Wasserman orientation relationship between the lamellae [27]. The dark-field image in Fig. 4(c), recorded from a $\{100\}$ FCC superlattice reflection in the $[011]$ FCC zone axis micro-diffraction pattern (inset) highlights the $L1_2$ regions within the FCC lamella. Similarly, the dark-field image in Fig. 4(d), obtained from the $\{100\}$ superlattice reflection in the $[011]$ BCC zone selected area diffraction pattern (inset) highlights the B2 regions within the BCC lamella. Therefore, the CRSA600 condition exhibits a complex nanolamellar pearlite-like microstructure, consisting of alternating FCC+ $L1_2$ and BCC+B2 lamellae. This indicates that concurrent growth of FCC/ $L1_2$ and BCC/B2 lamellae in the form of eutectoid-like colonies can overcome the nucleation barrier for the formation of B2 precipitates within the parent FCC matrix. Dark-field images (Figs. 4(c) and (d)) indicate that within each lamella, the FCC and $L1_2$ phases and the BCC and B2 phases can be co-continuous, suggesting a synergistic spinodal decomposition coupled with chemical ordering within both FCC and BCC lamellae [28, 29].

3.4 Atom probe tomography

Atom probe tomography (APT) was performed to determine the constitution of phases in the four-phase microstructure of the CRSA600 condition. Figs. 5(a) and (b) show images of the two different APT reconstructions exhibiting several interphase interfaces. The composition of each phase was determined by averaging the volume of the phase within a sphere of 10 nm diameter, placed such that it is within that phase. The BCC phase, as expected, was lean in Al and Ni and rich in Fe and Co with a composition of 4Al-45Co-47Fe-4Ni. The B2 precipitates were rich in Ni and Al with a composition of 30Al-11Co-10Fe-49Ni. Within the FCC based lamella, the continuous matrix appears to be rich in Ni and Al and has thus been assigned as the $L1_2$ phase, with a composition of 23Al-5Co-5.5Fe-66.5Ni. The APT reconstructions and analysis clearly

reveal Ni and Al lean regions within the L1₂ lamellae, and these regions are expected to be FCC with the composition 6Al-30Co-30Fe-34Ni. The compositions of each phase are tabulated in Supplementary Table I. Based on these compositions obtained from APT, the site occupancies in L1₂ regions could be represented as (Ni,Fe,Co)₃(Al,Fe) [30] and the B2 regions as (Ni,Co)(Al,Fe,Co) [30, 31].

3.5 Mechanical properties

Fig. 6 shows the engineering stress-strain curves for the four conditions in tension and Table 1 shows the results from microhardness measurements. The single-phase FCC condition (CRSA) of Al_{0.3}CoFeNi exhibited a yield stress (YS) ~457 MPa, an ultimate tensile stress (UTS) ~852 MPa and a net tensile strain ~76%. The sample did not fail during the test as it reached the maximum elongation. The microhardness of this condition was measured to be ~196 VHN. When compared to conventional solid solutions, the relatively high yield stress and hardens in CRSA can be rationalized based on the lattice distortion theory in HEA/CCAs and the solid solution strengthening caused by the size and modulus misfit of Al atoms in CoFeNi matrix. The nano-lamellar microstructure, with substantial phase fractions of L1₂ and B2 intermetallic phases within the lamellae in CRSA600 condition, exhibited a high yield stress of ~1074 MPa, UTS of ~1302 MPa and a net plastic strain of ~8%. This condition also showed the highest hardness (~513 VHN) among the four conditions studied.

Interestingly, the CRSA700 condition exhibits a yield stress marginally lower than CRSA condition. This could potentially be due to reduced solid solution strengthening in the FCC matrix because of the depletion of Al and Ni caused by B2 precipitation. Additionally, the volume fraction

of B2 phase is too low to have a substantial strengthening contribution. The hardness of CRSA600 was found to be similar to that of CRSA condition. This similarity in hardness can be attributed to two factors. First, the diameter of hardness indent was around 65 μm while the grain size was around 171 μm . The plastic deformation, mediated by dislocation glide, may not have spread to the grain boundaries to encounter the effect of the B2 precipitates. However, in the case of the CR700 (direct annealed at 700°C after cold-rolling) sample, a large number of heterogenous defects, such as deformation bands (containing a high density of dislocations) were produced during rolling, resulting in a higher, near equilibrium volume fraction of the B2 phase. The fine recrystallized FCC grain structure provided Hall-Petch strengthening and B2 phase provided precipitation strengthening. This resulted in a good balance of tensile properties with yield stress ~ 893 MPa, ultimate tensile strength ~ 1187 MPa and a tensile ductility $\sim 26\%$. The hardness of CR700 was found to be ~ 232 VHN, which is also substantially higher than CRSA and CRSA700 conditions.

3.6 Magnetic properties

The magnetization curves were obtained by plotting magnetization (M) as a function of applied magnetic field (H), for each of the four conditions, CRSA, CRSA600, CRSA700, and CR700 (Fig. 6). The inset shows a magnified view of the region near zero applied field, revealing that the area under the magnetization curve is maximum for the CRSA600 condition. Table I summarizes the magnetization properties (saturation magnetization, M_s , and coercivity, H_c) and the microhardness values.

The value of M_s varies from 126.8 emu/g in the case of the CRSA700 condition to 138.7 emu/g in the case of the nano-lamellar CRSA600 condition. The value of M_s is not a strong function of the microstructure. The coercivity (H_c), on the other hand, varies dramatically and is dependent on the microstructure. The coercivity of the single-phase FCC solid solution condition in the case of CRSA is 199 A/m, increases to 604 A/m in the case of the CR700 sample, and finally becomes 12732 A/m in the case of the nano-lamellar CRSA600 condition. This corresponds to more than eighty times increase in the value of H_c in the same alloy. The nano-lamellar structure with sharp chemical/phase change in CRSA600 results in high resistance to magnetic domain wall movement and therefore higher value of coercivity. Lowe et al. also reported high value of coercivity in Alnico samples which exhibited a spinodal microstructure consisting of FeCo rich and AlNi rich phases with sharp chemical contrast [32].

For convenience, magnetic materials can be divided in three categories based on the value of coercivity; (i) soft ($H_c < 25$ Oe (2000 A/m)), (ii) semi hard (25 Oe $< H_c < 700$ Oe (55720 A/m)) and (iii) hard ($H_c > 700$ Oe (55720 A/m))[33]. CRSA, CRSA700 and CR700 belong to the soft magnetic category while CRSA600 belongs to the semi hard magnetic category. Fig 6 (b) shows the M_s and H_c for our $Al_{0.3}FeCoNi$ along with other reported FeCoNi based soft magnetic $FeCoNi(MnAl)_x$ ($x = 0.25, 0.50, 0.75$ and 1) alloys [34], $FeCoNi(AlSi)_x$ ($x = 0.1, 0.2$ and 0.5)[35] and $FeCoNi(MnAl)_{0.25}$ (as cast, cold rolled and annealed)[36] CCAs. CRSA and CRSA700 exhibit an excellent combination of low H_c and high M_s (highlighted in green color in Fig. 6(b)). The low coercivity for CRSA700 can be attributed by the fact that it has an average grain size higher than that of CRSA and CR700. Fig 6(c) shows the M_s and H_c for CRSA600 along with other FeCoNi based semi hard magnetic $FeCoNi(MnSi)_x$ ($x = 0.25, 0.50$ and 0.75) [37] and $FeCoNi(AlSi)_x$ ($x =$

0.3, 0.4 and 0.8) CCAs. Recently, Zhang et al., explored $\text{Al}_{0.5}\text{CoFeNi}$ CCAs, however, the H_c values were changed only in the range of 60 Oe to 90 Oe [38]. Some of the earlier reported CCAs exhibit larger values of H_c than CRSA600. However, the M_s for those CCAs is inferior (<100 emu/g). High M_s is a key requirement for both soft and semi (hard) magnetic applications. Our CRSA600 exhibits high saturation magnetization (138.7 emu/g) along with reasonable high H_c (12720 A/m).

Fig. 6(d) shows the Vickers hardness of some of the commercialized soft and hard magnetic materials along with our $\text{Al}_{0.3}\text{FeCoNi}$ studied in four different conditions. The hardness of CRSA, CRSA700 and CR700 are higher or close to the hardness of famous soft magnetic materials, e.g., $\text{Fe}_{50}\text{Ni}_{50}$, permalloy and permendur. On the other hand, hardness of CRSA600 is very close to the values for Alnico, Sm-Co and Nd-Fe-B types of permanent magnets. The high hardness value coupled with high saturation magnetization and coercivity in CRSA600 condition makes this alloy a potential non-rare earth containing candidate for semi hard magnet applications which also require good mechanical strength. While the tensile ductility decreased with increase in yield stress, all four conditions exhibited a ductility reasonable for magnetic applications. Our results clearly demonstrate the feasibility of tailoring the site specific mechanical and magnetic properties within a component via site-specific thermo-mechanical treatment of a single alloy composition for room temperature applications. Such an approach has been recently referred to as metamorphic manufacturing [1, 39]. In addition to room temperature applications, this alloy can also be potentially used for intermediate temperature ($<500^\circ\text{C}$) applications. In particular, the two-phase microstructure in CR700 (FCC+B2) and the four-phase microstructure in CRSA600 (FCC+L1₂+BCC+B2) are in agreement with the equilibrium thermodynamic modeling shown in

Fig. 1(a). The obtained microstructures in these two conditions are expected to have reached equilibrium because of the long-term annealing (50 hrs at 700°C and 600°C) carried out in this study. However, the high temperature properties of these microstructures is a subject of future study.

4. Summary and Conclusions

The mechanical and magnetic properties of an $\text{Al}_{0.3}\text{CoFeNi}$ alloy can be tailored over a wide range, paving the way for metamorphic manufacturing of mechanically strong magnetic components.

1. A novel eutectoid-like nano-lamellar microstructure, consisting of complex hierarchically decomposed alternating lamellae of FCC+ L_{12} /BCC+B2, has been found in an $\text{Al}_{0.3}\text{CoFeNi}$ CCA. This structure arises from Al alloying addition, which results in a competition between the tendency for L_{12} ordering within the FCC lattice and BCC/B2 phase formation. The experimentally observed phase stability as a function of temperature could be largely rationalized based on CALPHAD modeling (ThermoCalc with TCHEA3 database).
2. The nano-lamellar microstructure exhibits a hardness of ~ 513 VHN, saturation magnetization M_s ~138 emu/g and coercivity H_c ~12732 A/m, making this a mechanically and magnetically semi hard alloy.
3. On the other hand, the microstructure with fine recrystallized FCC grains and near equilibrium B2 phase fraction exhibited saturation magnetization of ~132 emu/gm, reasonably low coercivity 604 A/m and high hardness ~232 VHN.

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Competing interests

None.

Table 1: Summary of the magnetic and microhardness properties for the four conditions.

Condition	Ms (emu/g)	Hc (Oe) Hc (A/m)	Average Microhardness (VHN)
Solutionized (CR85+1250°C/5min) CRSA	131.4	2.5 199	196.6 ± 12.61
Indirect 700 (CR85+1250°C/5min +700°C/50h) CRSA700	126.8	1.9 151	195.52 ± 3.73
Direct 700 (CR85+700°C/50h) CR700	132.6	7.6 604	232.34 ± 3.84
Indirect 600 (CR85+1250°C/5min +600°C/50h) CRSA600	138.7	160.0 12732	513 ± 7.25

Figure Captions

Fig. 1: (a) Pseudo-binary isopleth for varying Al in ternary CoFeNi system (b) Phase fraction vs temperature diagram showing the equilibrium volume fractions of phases at various temperatures.

Fig. 2 Backscattered SEM micrographs of (a) CRSA (b) CRSA600 (c) CRSA700 and (d) CR700. The insets show high magnification pictures from the respective conditions.

Fig. 3: Coupled EBSD-EDS mapping of CR700 (a) Phase map from EBSD (b) Al map showing the enrichment in the grain boundary precipitates.

Fig. 4 TEM of CR700 in (a) and CRSA600 in (b-d), (a) Bright field image showing ultra-fine FCC grains and a B2 precipitate (dark in color). Inset shows the diffraction pattern recorded from $[001]_{B2}$ (b) Bright-field image showing the nano-lamellar structure (c) Dark-field image taken from L12 superlattice reflection. Inset shows the diffraction pattern from $[011]_{FCC}$ zone axis taken from region A in (b); (d) Dark-field image taken from B2 superlattice reflection. Inset shows the diffraction pattern from $[011]_{BCC}$ taken from region B in (b)

Fig. 5: (a-b) Two different atom probe reconstructions of CRSA600 showing the FCC and BCC lamellae and the intermetallic precipitates within each lamella.

Fig. 6 (a) Magnetization versus applied magnetic field curves of $Al_{0.3}FeCoNi$ studied at four condition. Comparison of saturation magnetization (left axis, square) and coercivity values (right axis, circle) of $Al_{0.3}FeCoNi$ CCA with associated FeCoNi based (b) soft magnetic HEA (c) semi

hard magnetic HEA, Black and red lines are guide to the eye. (d) Vickers hardness of Al_{0.3}FeCoNi studied at four conditions and some of the commercial soft and hard magnets.

References

- [1] G. Spanos, G. Daehn, J. Allison, E. Bilitz, D. Bourne, J. Cao, K. Clarke, J. DeLoach Jr, E. Herderick, J. Lewandowski, *Metamorphic Manufacturing: Shaping the Future of On-Demand Components*, The Minerals Metals and Materials Society, Inc Pittsburgh United States, 2019.
- [2] B. Gwalani, S. Gorsse, D. Choudhuri, Y.F. Zheng, R.S. Mishra, R. Banerjee, Tensile yield strength of a single bulk Al_{0.3}CoCrFeNi high entropy alloy can be tuned from 160 MPa to 1800 MPa, *Scripta Mater* 162 (2019) 18-23.
- [3] J. Su, D. Raabe, Z.M. Li, Hierarchical microstructure design to tune the mechanical behavior of an interstitial TRIP-TWIP high-entropy alloy, *Acta Mater* 163 (2019) 40-54.
- [4] W.-R. Wang, W.-L. Wang, J.-W. Yeh, Phases, microstructure and mechanical properties of Al_xCoCrFeNi high-entropy alloys at elevated temperatures, *J Alloy Compd* 589 (2014) 143-152.
- [5] J.W. Yeh, S.K. Chen, S.J. Lin, J.Y. Gan, T.S. Chin, T.T. Shun, C.H. Tsau, S.Y. Chang, Nanostructured high-entropy alloys with multiple principal elements: novel alloy design concepts and outcomes, *Adv Eng Mater* 6(5) (2004) 299-303.
- [6] A. Manzoni, H. Daoud, R. Völkl, U. Glatzel, N. Wanderka, Phase separation in equiatomic AlCoCrFeNi high-entropy alloy, *Ultramicroscopy* 132 (2013) 212-215.
- [7] W.R. Wang, W.L. Wang, S.C. Wang, Y.C. Tsai, C.H. Lai, J.W. Yeh, Effects of Al addition on the microstructure and mechanical property of Al_xCoCrFeNi high-entropy alloys, *Intermetallics* 26 (2012) 44-51.
- [8] T. Borkar, B. Gwalani, D. Choudhuri, C. Mikler, C. Yannetta, X. Chen, R.V. Ramanujan, M. Styles, M. Gibson, R. Banerjee, A combinatorial assessment of Al_xCrCuFeNi₂ (0 < x < 1.5) complex concentrated alloys: Microstructure, microhardness, and magnetic properties, *Acta Mater* 116 (2016) 63-76.
- [9] T. Borkar, V. Chaudhary, B. Gwalani, D. Choudhuri, C.V. Mikler, V. Soni, T. Alam, R. V. Ramanujan, R. Banerjee, A combinatorial approach for assessing the magnetic properties of high entropy alloys: role of Cr in AlCo_xCr_{1-x}FeNi, *Adv Eng Mater* 19(8) (2017) 1700048.

- [10] Y.J. Chang, A.C. Yeh, The evolution of microstructures and high temperature properties of $\text{Al}_x\text{Co}_{1.5}\text{CrFeNi}_{1.5}\text{Ti}_y$ high entropy alloys, *J Alloy Compd* 653 (2015) 379-385.
- [11] T. Yang, Y.L. Zhao, Y. Tong, Z.B. Jiao, J. Wei, J.X. Cai, X.D. Han, D. Chen, A. Hu, J.J. Kai, K. Lu, Y. Liu, C.T. Liu, Multicomponent intermetallic nanoparticles and superb mechanical behaviors of complex alloys, *Science* 362(6417) (2018) 933-937.
- [12] J.M. Silveyra, A.M. Leary, V. DeGeorge, S. Simizu, M.E. McHenry, High speed electric motors based on high performance novel soft magnets, *J Appl Phys* 115(17) (2014).
- [13] S. Collocott, *Magnetic Materials: Domestic Applications*, (2016).
- [14] G. Bertotti, General-Properties of Power Losses in Soft Ferromagnetic Materials, *Ieee T Magn* 24(1) (1988) 621-630.
- [15] J.M. Silveyra, P. Xu, V. Keylin, V. DeGeorge, A. Leary, M.E. McHenry, Amorphous and Nanocomposite Materials for Energy-Efficient Electric Motors, *J Electron Mater* 45(1) (2016) 219-225.
- [16] M. Filippini, P. Alotto, V. Cirimele, M. Repetto, C. Ragusa, L. Dimauro, E. Bonisoli, Magnetic Loss Analysis in Coaxial Magnetic Gears, *Electronics-Switz* 8(11) (2019).
- [17] M.K. Mu, Q. Li, D.J. Gilham, F.C. Lee, K.D.T. Ngo, New Core Loss Measurement Method for High-Frequency Magnetic Materials, *Ieee T Power Electr* 29(8) (2014) 4374-4381.
- [18] R.M. Bozorth, *Ferromagnetism*, *Ferromagnetism*, by Richard M. Bozorth, pp. 992. ISBN 0-7803-1032-2. Wiley-VCH, August 1993. (1993) 992.
- [19] M. Lefik, K. Komez, E.N. Juszczak, D. Roger, P. Napieralski, N. Takorabet, H. Elmadah, High temperature machines: topologies and preliminary design, *Open Phys* 17(1) (2019) 657-669.
- [20] J. Karthaus, K. Hameyer, Static and cyclic mechanical loads inside the rotor lamination of high-speed PMSM, 2017 7th International Electric Drives Production Conference (EDPC), IEEE, 2017, pp. 1-6.
- [21] R. Saidur, A review on electrical motors energy use and energy savings, *Renewable and Sustainable Energy Reviews* 14(3) (2010) 877-898.
- [22] P. Ohodnicki, *Soft Magnetic Alloys for Electrical Machine Applications: Basics, State-of-the-Art, and R&D Opportunities*, NETL Office of Research and Development, 2015.
- [23] N.-C. Liu, R.C. O'handley, W. Ho, R. Copeland, Semi-hard magnetic elements and method of making same, Google Patents, 1994.

- [24] Y.F. Kao, S.K. Chen, T.J. Chen, P.C. Chu, J.W. Yeh, S.J. Lin, Electrical, magnetic, and Hall properties of $\text{Al}_x\text{CoCrFeNi}$ high-entropy alloys, *J Alloy Compd* 509(5) (2011) 1607-1614.
- [25] T.T. Zuo, R.B. Li, X.J. Ren, Y. Zhang, Effects of Al and Si addition on the structure and properties of CoFeNi equal atomic ratio alloy, *J Magn Magn Mater* 371 (2014) 60-68.
- [26] B. Gwalani, S. Gorsse, D. Choudhuri, M. Styles, Y.F. Zheng, R.S. Mishra, R. Banerjee, Modifying transformation pathways in high entropy alloys or complex concentrated alloys via thermo-mechanical processing, *Acta Mater* 153 (2018) 169-185.
- [27] K. Koumatos, A. Muehleemann, A theoretical investigation of orientation relationships and transformation strains in steels, *Acta Crystallogr A* 73 (2017) 115-123.
- [28] S. Banerjee, P. Mukhopadhyay, Phase transformations: examples from titanium and zirconium alloys, Elsevier 2010.
- [29] W.A. Soffa, D.E. Laughlin, Decomposition and ordering processes involving thermodynamically first-order order \rightarrow disorder transformations, *J Appl Phys* 37(11) (1989) 3019-3028.
- [30] N.V. Allaverdova, V.K. Portnoy, L.A. Kucherenko, A.V. Ruban, V.I. Bogdanov, Atomic distribution of alloying additions between sublattices in the intermetallic compounds Ni_3Al and NiAl II: Microscopic calculations and X-ray diffraction analysis, *J Appl Phys* 141(2) (1988) 191-200.
- [31] Y.L. Hao, R. Yang, Y. Song, Y.Y. Cui, D. Li, M. Niinomi, Concentration of point defects and site occupancy behavior in ternary NiAl alloys, *Mat Sci Eng a-Struct* 365(1-2) (2004) 85-89.
- [32] K. Lowe, M. Durrschnabel, L. Molina-Luna, R. Madugundo, B. Frincu, H.J. Kleebe, O. Gutfleisch, G.C. Hadjipanayis, Microstructure and magnetic properties of melt-spun Alnico-5 alloys, *J Magn Magn Mater* 407 (2016) 230-234.
- [33] S. Constantinides, *Semi-Hard Magnets: The important role of materials with intermediate coercivity*, Magnetics, San Antonio, TX, 2011.
- [34] P. Li, A. Wang, C.T. Liu, Composition dependence of structure, physical and mechanical properties of $\text{FeCoNi}(\text{MnAl})_x$ high entropy alloys, *Intermetallics* 87 (2017) 21-26.
- [35] Y. Zhang, T. Zuo, Y. Cheng, P.K. Liaw, High-entropy Alloys with High Saturation Magnetization, Electrical Resistivity and Malleability, *Scientific Reports* 3(1) (2013) 1455.
- [36] P. Li, A. Wang, C.T. Liu, A ductile high entropy alloy with attractive magnetic properties, *Journal of Alloys and Compounds* 694 (2017) 55-60.

[37] P. Sahu, S. Solanki, S. Dewangan, V. Kumar, Microstructure and magnetic behavior of FeCoNi(Mn-Si)_x (x = 0.5, 0.75, 1.0) high-entropy alloys, *Journal of Materials Research* 34(5) (2019) 829-840.

[38] J. Xu, J.Y. Zhang, Y.Q. Wang, P. Zhang, J. Kuang, G. Liu, G.J. Zhang, J. Sun, Annealing-dependent microstructure, magnetic and mechanical properties of high-entropy FeCoNiAl_{0.5} alloy, *Materials Science and Engineering: A* (2020) 139003.

[39] G.S. Daehn, A. Taub, *Metamorphic manufacturing: The third wave in digital manufacturing*, *Manufacturing Letters* 15 (2018) 86-88.

Supplementary Information

Table S1: The composition of various phases determined by APT sphere analysis

Elements		FCC	L12	BCC	B2
Al	at. %	5.83	22.90	4.03	30.36
	sigma%	0.14	0.28	0.10	0.25
Co	at %	29.81	5.19	44.72	11.13
	sigma%	0.34	0.12	0.38	0.14
Fe	at. %	29.94	5.41	47.37	9.81
	sigma%	0.34	0.13	0.39	0.13
Ni	at. %	34.42	66.50	3.87	48.70
	sigma%	0.37	0.56	0.09	0.34