**SUPPLEMENTARY INFORMATION**

**ENHANCED MAGNETIZATION WITH INCREASED CHROMIUM CONCENTRATION IN FeCoCrxNi2Al HIGH-ENTROPY ALLOY**

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**SECTION I: Calculation Details**

1. **Enthalpy of Mixing (ΔHmix):**

The enthalpy of mixing (ΔHmix) for HEA with N elements can be determined using the following equation [1],

ΔHmix = $\sum\_{i=1\dot{i}\ne j}^{N}4ΔH\_{AB}^{mix} c\_{i}c\_{j}$

N is Total number of components in the system,

*ci* & *cj* are the composition of the ith & jth component (in a molar ratio).

Total molar concentration for Sample A (FeCoCr1.5Ni2Al) = 6.5

Total molar concentration for Sample B (FeCoCr0.5Ni2Al) = 5.5

From Miedema's model [2] the enthalpy of mixing$(ΔH\_{AB}^{mix})$ for binary alloys can be written from the below mentioned table

|  |  |
| --- | --- |
| **Binary Alloys** | **Enthalpy of mixing (**$ΔH\_{AB}^{mix}$**)** |
| Fe-Co | -1 |
| Fe-Cr | -1 |
| Fe-Ni | -2 |
| Fe-Al | -1 |
| Co-Cr | -4 |
| Co-Ni | 0 |
| Co-Al | -19 |
| Cr-Ni | -7 |
| Cr-Al | -10 |
| Ni-Al | -22 |

Enthalpy of mixing (ΔHmix) = 4 {[(-1) x $\frac{1}{6.5}$ x $\frac{1}{6.5}$] + [(-1) x $\frac{1}{6.5}$ x $\frac{1.5}{6.5}$] + [(-2) x $\frac{1}{6.5}$ x $\frac{2}{6.5}$] +

 of Sample A [(-1) x $\frac{1}{6.5}$ x $\frac{1}{6.5}$] + [(-4) x $\frac{1}{6.5}$ x $\frac{1.5}{6.5}$] + [(0) x $\frac{1}{6.5}$ x $\frac{2}{6.5}$] +

 [(-19) x $\frac{1}{6.5}$ x $\frac{1}{6.5}$] + [(-7) x $\frac{1.5}{6.5}$ x $\frac{2}{6.5}$] + [(-10) x $\frac{1.5}{6.5}$ x $\frac{1}{6.5}$] +

 [(-22) x $\frac{1}{6.5}$ x $\frac{1}{6.5}$]}

**ΔHmix = -14.87 kJ mol-1**

Enthalpy of mixing (ΔHmix) = 4 {[(-1) x $\frac{1}{5.5}$ x $\frac{1}{5.5}$] + [(-1) x $\frac{1}{5.5}$ x $\frac{0.5}{5.5}$] + [(-2) x $\frac{1}{5.5}$ x $\frac{2}{5.5}$] +

 of Sample B [(-1) x $\frac{1}{5.5}$ x $\frac{1}{5.5}$] + [(-4) x $\frac{1}{5.5}$ x $\frac{0.5}{5.5}$] + [(0) x $\frac{1}{5.5}$ x $\frac{2}{5.5}$] +

 [(-19) x $\frac{1}{5.5}$ x $\frac{1}{5.5}$] + [(-7) x $\frac{0.5}{5.5}$ x $\frac{2}{5.5}$] + [(-10) x $\frac{0.5}{5.5}$ x $\frac{1}{5.5}$] +

 [(-22) x $\frac{1}{5.5}$ x $\frac{1}{5.5}$]}

**ΔHmix = -11.04 kJ mol-1**

1. **Entropy of Mixing (ΔSmix):**

The entropy of mixing (ΔSmix) is calculated using the formula below [1],

$ΔS\_{mix}$ = - R$\sum\_{i=1}^{N}c\_{i}ln c\_{i}$

R = 8.314 J. (mol.K)-1

N is Total number of components in the system,

*ci* is the composition of the ith component (in a molar ratio).

Entropy of mixing (ΔSmix) of Sample A = - R [$\frac{1}{6.5} ln \frac{1}{6.5}$ + $\frac{1}{6.5} ln \frac{1}{6.5}$ + $\frac{1.5}{6.5} ln \frac{1.5}{6.5}$ +

 $\frac{2}{6.5} ln \frac{2}{6.5}$ + $\frac{1}{6.5} ln \frac{1}{6.5}$]

**ΔSmix = 13.01 J. (mol. K)-1**

Entropy of mixing (ΔSmix) of Sample B = - R [$\frac{1}{5.5} ln \frac{1}{5.5}$ + $\frac{1}{5.5} ln \frac{1}{5.5}$ + $\frac{1.5}{5.5} ln \frac{1.5}{5.5}$ +

 $\frac{2}{5.5} ln \frac{2}{5.5}$ + $\frac{1}{5.5} ln \frac{1}{5.5}$]

**ΔSmix = 12.59 J. (mol. K)-1**

**SECTION II: High Temperature X-Ray Diffraction (HT-XRD):**

In-situ High Temperature XRD experiments were carried out using BRUKER make D8 discover X-ray diffractometer equipped with a Mo (λKα = 0.7093 Å) radiation source. The heating of samples was carried out using a heating stage manufactured by Anton Paar (Model DHS 1100) maintained at a vacuum better than 10-4 mbar. Figure S1 shows the in-situ HT- XRD pattern obtained for Sample A from Room temperature (RT) to 900oC. The temperature stability of the phases can be understood from these diffraction results. It is found that similar to our XRD results in Fig 1(a), we observe a dual phase FCC + BCC peaks up to 900oC with no formation of any additional phases. The FCC and BCC lattice parameters at Room Temperature are found to be 3.58 Å and 2.87 Å respectively.



Figure S1: In-situ HT-XRD results of Sample A = FeCoCr1.5Ni2Al from RT to 900oC in vacuum. The observed phases are stable from room temperature to 900oC with a slight increase in the ‘BCC’ peak intensity of 2θ angle at 20­o.

**SECTION III: Magnetization vs Temperature measurement.**

Figure S2 & S3 (a-b) represents the Magnetization vs Temperature plot for two different temperature ranges such as (a) 15 K -300K (b) 300 K – 900 K for Sample A and Sample B respectively. In order to calculate the Curie Temperature (Tc) from the plot, the corresponding first order derivative plot of Magnetization are obtained. (c-d) The dM/dT vs T represents the Tc for low and high temperature regime.

Due to the coexistence of multiple phases in the samples, we notice more than one ordering temperature for each sample. We understand that in general for the high entropy alloy the paramagnetic to ferromagnetic phase transition temperature (Curie temperature) can be well estimated from the minima of the plot of dM/dT vs T plot [3]. However, due to multiphase character of these samples it is quite non-trivial to estimate the ordering temperature. The values of ordering temperature were estimated by either noticing peak in the dM/dT vs T plot or by extrapolating the linear part of the M vs T where it intersects M = 0 [4]. Note that for sample A it is quite challenging to assign minima peak in dM/dT vs T plot in figure R3 (c) and (d). Hence by extrapolating the linear part of the M vs T crossing M = 0, we estimate the ordering temperature for sample A ~ 750 K. For sample B, it is hard to explain the origin of the peak in figure R4(c) at 100K, however, we observe two minima in figure R4 (d) and the corresponding ordering temperatures are defined as 550 K and 810 K. In both these samples we find that the magnetization slowly decreases to zero beyond 800K.



Figure S2 (a-d) Magnetization versus temperature response for sample A in range (a) 15 K – 300 K and (b) 300 K - 900 K. Measurement was performed at H = 100 Oe magnetic field. Corresponding dM/dT vs T plot is shown for (c) 15 K - 300 K and (d) 300 K – 900 K.



Figure S3: Magnetization versus temperature response for sample A in range (a) 15 K - 300 K and (b) 300 K - 900 K. Measurement was performed at H = 100 Oe magnetic field. Corresponding dM/dT vs T plot is shown for (c) 15 K - 300 K and (d) 300 K - 900 K.

**References**

[1] Y. Zhang *et.al.,* Adv Eng Mater., 10 (6), pp 534-538, 2008.

[2] A. Takeuchi and A. Inoue: Mater. Trans., 41 (11), pp 1372–1378, 2000.

[3] T. Borkar *et al.,* Adv. Eng. Mater.,19(8), pp 1700048, 2017.

[4] J. Cieslak *et al.,* Intermetallics., 118, pp 106672, 2020.