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Supporting Information for

Accelerated design for magnetic high entropy alloys using data-driven multi-objective optimization

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Table S1. Descriptive statistics of the target properties.

Target	Count	Mean	STD	Min	25%	50%	75%	Max
M_s	151	90.7	41.2	4.2	70.8	91.0	120.0	176.9
H	279	403	197	110	198	418	556	850

Notes: Count is the total number of alloying compositions with experimental M_s and H data in the HEA dataset; Mean, STD Min and Max are the mean, standard deviation, minimum and maximum of M_s or H, respectively; 25%, 50% and 75% are the 25th, 50th, and 75th percentiles of M_s or H, respectively.

Table S2. The list of candidate features for the prediction of HEAs' magnetic or mechanical properties.

Class	No.	Description	Formula
Chemical components	1-20	Atomic percent of chemical elements (c)	c_i, i = B, C, Al, Si, P, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Ga, Zr, Nb, Mo, Pd, Sn, Gd
Related to atomic structures	21	Theoretical density (ρ)	$100/(\sum \frac{w_i}{\rho_i})$
	22	Mean metallic atomic radius (R_m)	$\sum c_i r_i$
	23	Atomic size difference (δ_R)	$\sqrt{\sum c_i \left(1-\frac{r_i}{\bar{r}}\right)^2}$
	24	Theoretical molar volume (V_m)	$V_m = \sum_{i=1}^{n} \frac{c_i m_i}{\rho_i}$
Related to thermodynamics	25	Mixing entropy (ΔS_{mix})	$\Delta S_{mix} = -R \sum c_i \ln c_i$

	26	Mixing enthalpy (ΔH_{mix})	$\Delta H_{\mathrm{mix}} = \sum 4c_i c_j \Delta H_{AB}^{\mathrm{mix}}\ , i \neq j$
	27	Ω	$\frac{T_{\mathrm{m}}\Delta S_{\mathrm{mix}}}{\lvert\Delta H_{\mathrm{mix}}\rvert}$
	28	Mixing Gibbs free energy (ΔG_{mix})	$\Delta G_{\mathrm{mix}} = \Delta H_{\mathrm{mix}} - T_{\mathrm{m}}\Delta S_{\mathrm{mix}}$
	29	Melting temperature calculated by the rule of mixtures (T_{m})	$T_{\mathrm{m}} = \sum c_i (T_{\mathrm{m}})_i$
Related to electronic properties	30	Pauling electronegativity (χ)	$\sum c_i \chi_i$
	31	Pauling electronegativity difference (δ_χ)	$\sqrt{\sum c_i \left(1 - \frac{\chi_i}{\bar{\chi}}\right)^2}$
	32	Relative Pauling electronegativity ($R\chi$)	$\sum c_i\chi_i - \sum c_k\chi_k$, k = Fe, Co, Ni, Gd
	33	Valence electron concentration (VEC)	$\sum c_i VEC_i$
	34	Valence electron concentration difference (δ_{VEC})	$\sqrt{\sum c_i \left(1 - \frac{VEC_i}{\overline{VEC}}\right)^2}$
	35	Relative valence electron concentration ($RVEC$)	$\sum c_i VEC_i - \sum c_k VEC_k$, k = Fe, Co, Ni, Gd
	36	Work function (W)	$\sum c_i W_i$
	37	Work function difference (δ_{W})	$\sqrt{\sum c_i \left(1 - \frac{W_i}{\overline{W}}\right)^2}$
Related to magnetism	38	Average atomic moment of ferromagnetic components ($\bar{\mu}$)	$c_{\mathrm{Fe}}\mu_{\mathrm{Fe}} + c_{\mathrm{Co}}\mu_{\mathrm{Co}} + c_{\mathrm{Ni}}\mu_{\mathrm{Ni}} + c_{\mathrm{Gd}}\mu_{\mathrm{Gd}}$
	39	Theoretical saturated magnetization of ferromagnetic components (B_{s})	$\frac{\mathrm{N_A}\bar{\mu}\mu_B}{V_m}$

Notes: w_i, ρ_i, r_i, χ_i, VEC_i, m_i and $T_{\mathrm{m}i}$ are atomic weight percentage, density, atomic radius, electronegativity, valence electron concentration, atomic weight and melting point of the i^{th} constituent element in each composition, respectively. R is the gas constant, and $\Delta H_{AB}^{\mathrm{mix}}$ is the mixing enthalpy of the liquid binary alloy containing i^{th} and j^{th} elements[1]. The average magnetic moment of Fe, Co and Ni atoms are $\mu_{\mathrm{Fe}} = 2.2\mu_B$, $\mu_{\mathrm{Co}} = 1.7\mu_B$ and $\mu_{\mathrm{Ni}} = 0.6\mu_B$, respectively, where μ_B is the Bohr magneton.

Table S3. Elemental properties used for feature calculation.

Element	Atomic weight (Da)	ρ (g/cm^3)	R_{m} (Å)	T_{m} (K)	χ	*VEC*	*W*
Fe	55.8	7.874	1.241	1811	1.83	8	4.7
Ni	58.7	8.912	1.246	1728	1.91	10	5.15
Co	58.9	8.860	1.251	1768	1.88	9	5.00
Al	27.0	2.698	1.432	933	1.61	3	4.28
Cr	52.0	7.150	1.249	2180	1.66	6	4.50
Cu	63.5	8.960	1.278	1358	1.9	11	4.65
Mn	54.9	7.440	1.350	1519	1.55	7	4.10
Si	28.1	2.330	1.153	1687	1.9	4	4.85
V	50.9	6.110	1.316	2183	1.63	5	4.30
B	10.8	2.340	0.820	2349	2.04	3	4.45
Ti	47.9	4.540	1.462	1941	1.54	4	4.33
Zr	91.2	6.506	1.603	2128	1.33	4	4.05
C	12.0	2.267	0.773	4000	2.55	4	5.00
P	31.0	1.820	1.060	317	2.19	5	0.00
Ga	69.7	5.907	1.392	303	1.81	3	4.20
Nb	92.9	8.570	1.429	2750	1.6	5	4.30
Mo	96.0	10.220	1.363	2896	2.16	6	4.60
Sn	118.7	7.287	1.620	505	1.96	4	4.42
Pd	106.4	12.020	1.375	1828	2.2	10	5.12
Gd	157.3	7.895	1.801	1585	1.2	3	3.10

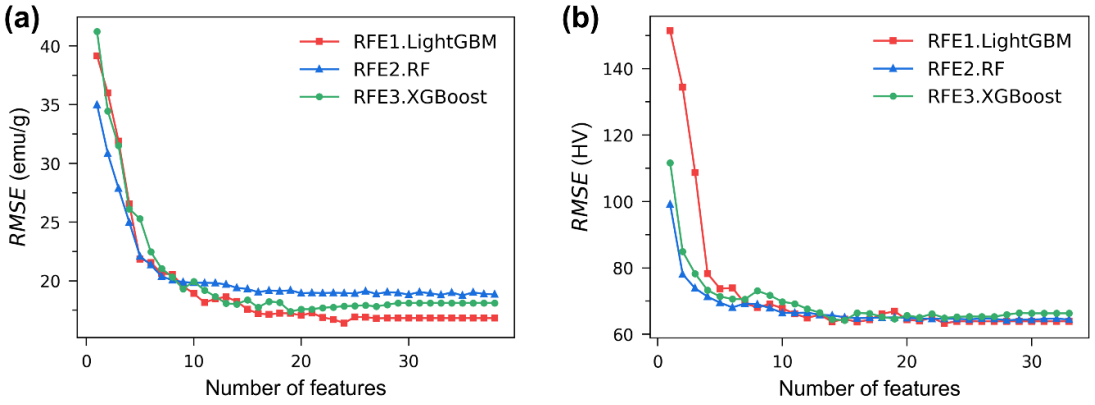

Fig. S1. Feature selection based on RFE with different cores for (a) M_s and (b) H prediction.

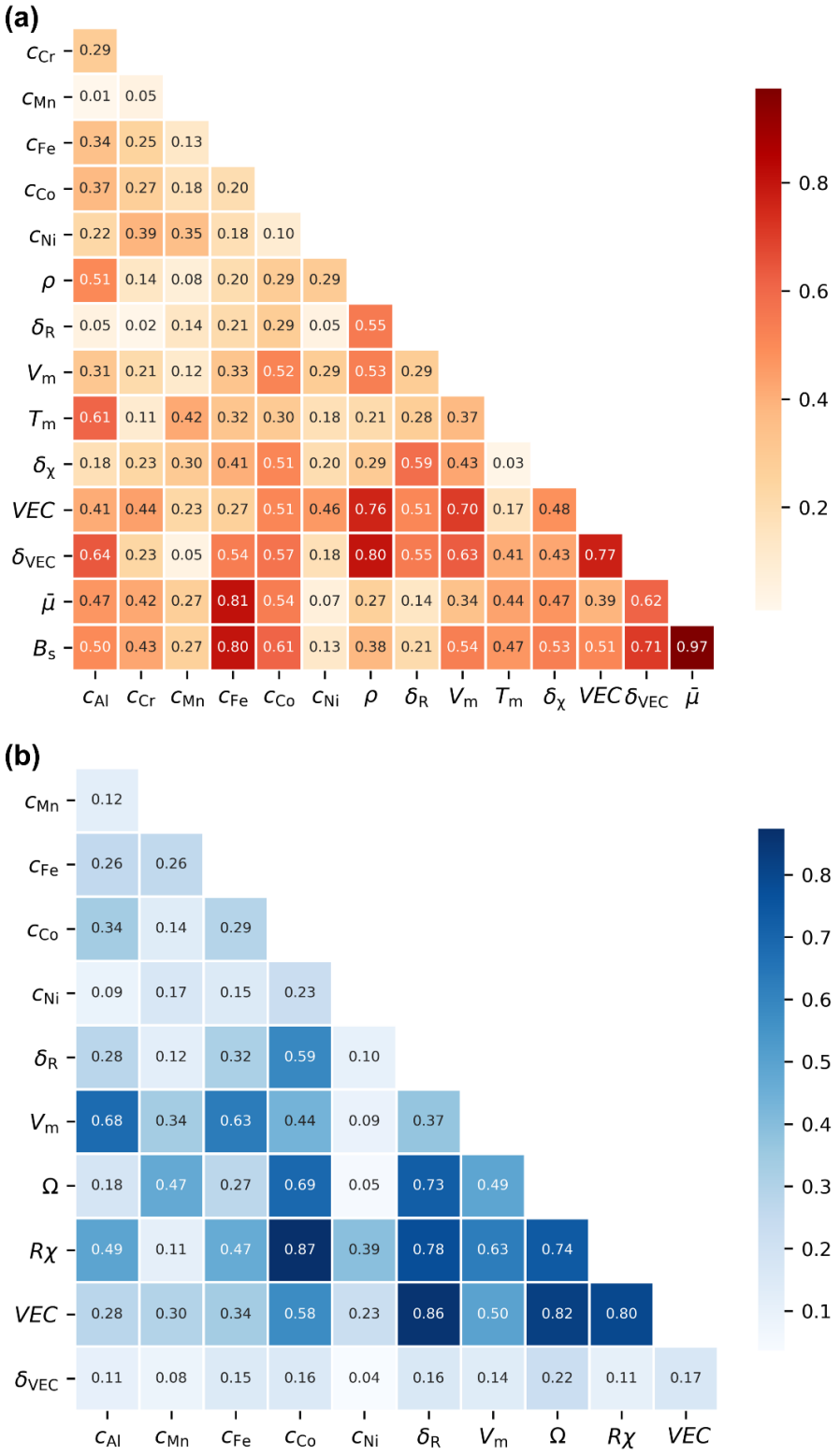

Fig. S2. The absolute PCC maps of the features selected by REF for (a) M_s and (b) H prediction.

Table S4. Alloying compositions suggested by the weighted-sum method for different α/β.

α/β	Composition (at.%)					$\widehat{M}_s$ (emu/g)	$\widehat{H}$ (HV)
	Fe	Co	Ni	Al	Si		
1.0	44	25	7	16	8	144.2 ± 8.4	696 ± 30
1.1	44	25	7	16	8	144.2 ± 8.4	696 ± 30
1.2	44	25	7	16	8	144.2 ± 8.4	696 ± 30
1.3	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.4	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.5	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.6	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.7	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.8	43	25	10	13	9	149.2 ± 7.6	667 ± 32
1.9	43	25	10	13	9	149.2 ± 7.6	667 ± 32
2.0	43	25	10	13	9	149.2 ± 7.6	667 ± 32

Table S5. Pareto optimal set calculated by MOEA.

No.	Composition (at.%)					$\widehat{M}_s$ (emu/g)	$\widehat{H}$ (HV)
	Fe	Co	Ni	Al	Si		
1	35	29	8	17	11	128.8 ± 8.1	707 ± 39
2	35	29	7	18	11	124.2 ± 8.1	724 ± 39
3	37	29	10	16	8	142.0 ± 7.6	664 ± 34
4*	37	26	13	16	8	143.6 ± 7.1	658 ± 33
5	37	29	9	17	8	139.4 ± 7.9	676 ± 32
6	37	30	7	18	8	134.7 ± 8.1	695 ± 34
7	37	29	8	16	10	137.1 ± 8.2	689 ± 33
8	38	29	8	18	7	138.9 ± 8.0	688 ± 33
9	38	29	7	16	10	137.1 ± 8.3	692 ± 32
10	49	29	10	3	9	153.4 ± 16.3	431 ± 39
11*	49	22	11	9	9	148.8 ± 10.1	561 ± 43
12	49	24	7	9	11	146.6 ± 8.8	652 ± 35
13*	49	23	8	9	11	147.3 ± 8.8	633 ± 38
14	50	22	9	9	10	147.4 ± 9.5	602 ± 42

Note: "*" represents the alloying compositions chosen for experimental validation.

Table S6. Compositions and properties of some reported Fe-Co-Ni-Al-Si magnetic HEAs.

Composition	Fe (at.%)	Co (at.%)	Ni (at.%)	Al (a.t%)	Si (at.%)	M_s (emu/g)	H (HV)	ΔS_{mix} ($J \cdot K^{-1} \cdot mol^{-1}$)	Phase(s)
$(Fe_{0.3}Co_{0.5}Ni_{0.2})_{1-x}(Al_{1/3}Si_{2/3})_x$ [2]									
x = 0.05	28.5	47.5	19.0	1.7	3.3	153.5	-	10.05	FCC
x = 0.10	27.0	45.0	18.0	3.3	6.7	131.0	-	10.94	FCC
x = 0.15	25.5	42.5	17.0	5.0	10.0	117.1	-	11.59	BCC + FCC
x = 0.25	22.5	37.5	15.0	8.3	16.7	116.5	-	12.42	BCC
$FeCoNi(AlSi)_x$ [3]									
x = 0.10	31.3	31.3	31.3	3.1	3.1	143.4	147	10.87	FCC
x = 0.30	27.8	27.8	27.8	8.3	8.3	113.8	344	12.32	FCC + BCC
x = 0.50	25.0	25.0	25.0	12.5	12.5	107.1	644	12.97	BCC
x = 0.70	22.7	22.7	22.7	15.9	15.9	78.3	771	13.26	BCC
$(Fe_{0.45}Co_{0.3}Ni_{0.25})_{1-x}(Al_{0.4}Si_{0.6})_x$ [4]									
x = 0.125	39.4	26.3	21.9	5.0	7.5	123.2	260	11.60	BCC + FCC
x = 0.15	38.3	25.5	21.3	6.0	9.0	125.8	345	11.90	BCC + FCC
x = 0.16	37.8	25.2	21.0	6.4	9.6	126.3	421	12.00	BCC + FCC
x = 0.17	37.4	24.9	20.8	6.8	10.2	132.8	494	12.11	BCC + FCC
x = 0.18	36.9	24.6	20.5	7.2	10.8	148.5	599	12.20	BCC
x = 0.19	36.5	24.3	20.3	7.6	11.4	141.9	612	12.29	BCC
x = 0.2	36.0	24.0	20.0	8.0	12.0	133.3	621	12.38	BCC

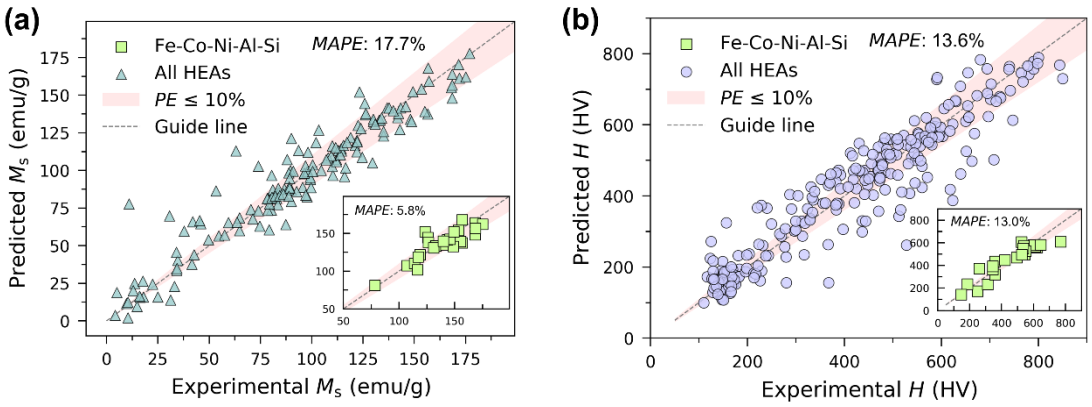

Fig. S3. Cross-validated predictions of (a) M_s *via* SVR and (b) *H via* LightGBM as compared against corresponding experimental values in the dataset after adding the experimental data of the newly synthesized HEAs.

References

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