## Supplementary Materials

# Machine learning assisted design of high entropy alloys with desired property

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The experimental HEAs and associated hardness of alloys obtained from Iterative Loop I are shown in Table S1.

**Table.S1. Composition and hardness values for newly-synthesized alloys from Iterative Loop I.**

| Input | Iteration No. | Alloy No. | Elemental content, at% | | | | | | Hardness, HV |
|---|---|---|---|---|---|---|---|---|---|
| | | | Al | Co | Cr | Cu | Fe | Ni | |
| composition | 1 | C1-1 | 34 | 17 | 34 | 0 | 5 | 10 | 699±37 |
| | | C1-2 | 47 | 14 | 15 | 5 | 13 | 6 | 728±42 |
| | | C1-3 | 38 | 11 | 6 | 9 | 31 | 5 | 604±31 |
| | 2 | C2-1 | 36 | 10 | 31 | 6 | 9 | 8 | 790±41 |
| | | C2-2 | 43 | 17 | 15 | 0 | 14 | 11 | 806±31 |
| | | C2-3 | 47 | 12 | 12 | 5 | 13 | 11 | 811±31 |
| | 3 | C3-1 | 43 | 19 | 19 | 0 | 11 | 8 | 855±26 |
| | | C3-2 | 39 | 10 | 31 | 5 | 8 | 7 | 802±18 |
| | | C3-3 | 47 | 13 | 13 | 5 | 12 | 10 | 799±42 |
| | 4 | C4-1 | 43 | 20 | 22 | 0 | 8 | 7 | 857±43 |
| | | C4-2 | 47 | 14 | 19 | 5 | 10 | 5 | 855±15 |
| | | C4-3 | 43 | 20 | 19 | 0 | 9 | 9 | 816±37 |
| | 5 | C5-1 | 46 | 14 | 23 | 5 | 7 | 5 | 811±34 |
| | | C5-2 | 44 | 14 | 22 | 5 | 9 | 6 | 790±48 |
| | | C5-3 | 43 | 19 | 24 | 0 | 8 | 6 | 855±30 |
| | 6 | C6-1 | 43 | 18 | 24 | 0 | 10 | 5 | 856±53 |
| | | C6-2 | 47 | 14 | 20 | 5 | 9 | 5 | 859±26 |
| | | C6-3 | 43 | 24 | 22 | 0 | 5 | 6 | 875±58 |
| | 7 | C7-1* | 43 | 25 | 22 | 0 | 5 | 5 | 865±39 |
| | | C7-2◊ | 43 | 24 | 23 | 0 | 5 | 5 | 868±45 |
| | | C7-3 | 43 | 23 | 21 | 0 | 8 | 5 | 863±34 |

**Notes:** "*" and "◊" represent the same alloys found by Iteration Loop I and Iteration Loop II, respectively.

The feature pool used in Iteration Loop II is illustrated by Table S2, which contains 20 features, 11 of which describe the formation of different phases (*i.e.*, phase parameters) and the other 9 are related to the mechanical properties (*i.e.*, mechanical parameters) obtained from literature reports and our understanding of the physics.

**Table.S2. The feature pool consisting of twenty features from insights of the materials science.**

| Phase parameter | Formula | Description | Mechanical parameter | Formula | Description |
|---|---|---|---|---|---|
| **δr**[1] | $\sqrt{\sum_{i=1}^{n} C_i * (1-\frac{r_i}{r})^2}$ | difference of atomic radii | **η**[2] | $\sum_{i=1}^{n} \frac{C_i * \frac{2(G_i-G)}{G_i+G}}{1+0.5*\lvert C_i * \frac{2(G_i-G)}{G_i+G}\rvert}$ | modulus mismatch in strengthening model |
| **Δχ**[1] | $\sqrt{\sum_{i=1}^{n} C_i * (\chi-\chi_i)^2}$ | difference of electronegativity | **D.r**[3] | $\sum_{i=1}^{n}\sum_{j=1,i\neq j}^{n} C_i C_j * \lvert r_i - r_j\rvert$ | local size mismatch |
| **VEC**[1] | $\sum_{i=1}^{n} C_i * VEC_i$ | valence electron concentration (containing d orbital electrons) | **A**[4] | $G * \delta r * (1+\mu)/(1-\mu)$ | energy term in strengthening model |
| **ΔH**[1] | $\sum_{i=1,j>i}^{n} 4C_i C_j * \Delta H_{ij}^{mix}$ | mixing enthalpy | **F**[5] | $\frac{2G}{1-\mu}$ | Peierls-Nabarro factor |
| **ΔS**[1] | $-R\sum_{i=1}^{n} C_i * \ln(C_i)$ | configuration entropy | **w**[6] | $(\sum_{i=1}^{n} C_i * w_i)^6$ | six square of work function |
| **Ω**[1] | $T * \Delta S/\lvert\Delta H\rvert$ | Ω parameter | **G** | $\sum_{i=1}^{n} C_i * G_i$ | shear modulus |
| **Λ**[1] | $\Delta S/(\delta r * \delta r)$ | Λ parameter | **δG** | $\sqrt{\sum_{i=1}^{n} C_i * (1-\frac{G_i}{G})^2}$ | difference of shear modulus |
| **γ**[1] | $(1-\sqrt{\frac{(r+r_{min})^2-r^2}{(r+r_{min})^2}})$ $/(1-\sqrt{\frac{(r+r_{max})^2-r^2}{(r+r_{max})^2}})$ | γ parameter | **D.G** | $\sum_{i=1}^{n}\sum_{j=1,i\neq j}^{n} C_i C_j * \lvert G_i - G_j\rvert$ | local modulus mismatch |
| **D. χ** | $\sum_{i=1}^{n}\sum_{j=1,i\neq j}^{n} C_i C_j * \lvert\chi_i-\chi_j\rvert$ | mismatch of local electronegativity | **μ** | $\frac{1}{2} E * \delta r$ | lattice distortion energy |
| **e/a** | $\sum_{i=1}^{n} C_i * (e/a)_i$ | number of itinerant electrons | | | |
| **Ec** | $\sum_{i=1}^{n} C_i * (Ec)_i$ | cohesive energy | | | |

**Notes:** $C_i$, $r_i$, $\chi_i$, $VEC_i$, $e/a_i$, $Ec_i$, $G_i$ and $w_i$ represent mole ratio, atomic radius, Pauling electronegativity, valence electron concentration (containing d orbital electrons), electron concentration, cohesive energy, shear modulus and work function of i element, respectively. T, r, χ, G, E

and μ represent the mean value of melting point, atomic radius, Pauling electronegativity, shear modulus, Young's modulus and Poisson ratio of all the elements in HEAs. $\Delta H^{mix}_{ij}$ represents the mixing enthalpy of i and j elements. $r_{min}$ and $r_{max}$ are the maximum atomic radius and the minimum atomic radius of all elements, respectively.

The experimental HEAs and associated hardness of alloys obtained from Iteration Loop II are shown in Table S3.

**Table.S3. Composition and hardness values for the newly-synthesized alloys predicted from Iterative Loop II**

| Input | Iteration No. | Alloy No. | Elemental contents, at% | | | | | | Hardness, HV |
|---|---|---|---|---|---|---|---|---|---|
| | | | Al | Co | Cr | Cu | Fe | Ni | |
| composition+ knowledge | 1 | K1-1 | 47 | 12 | 20 | 5 | 11 | 5 | 843±52 |
| | | K1-2 | 43 | 15 | 17 | 5 | 14 | 6 | 794±35 |
| | | K1-3 | 42 | 10 | 30 | 5 | 7 | 6 | 783±27 |
| | 2 | K2-1 | 47 | 11 | 22 | 5 | 10 | 5 | 797±17 |
| | | K2-2 | 47 | 13 | 20 | 5 | 10 | 5 | 805±24 |
| | | K2-3 | 29 | 22 | 34 | 0 | 5 | 10 | 699±10 |
| | 3 | K3-1 | 47 | 7 | 16 | 9 | 16 | 5 | 749±26 |
| | | K3-2 | 43 | 14 | 21 | 5 | 12 | 5 | 828±42 |
| | | K3-3 | 47 | 18 | 15 | 5 | 10 | 5 | 849±37 |
| | 4 | K4-1 | 47 | 20 | 18 | 5 | 5 | 5 | 883±22 |
| | | K4-2 | 43 | 18 | 20 | 0 | 12 | 7 | 864±23 |
| | | K4-3 | 47 | 17 | 16 | 5 | 10 | 5 | 841±37 |
| | 5 | K5-1◊ | 43 | 24 | 23 | 0 | 5 | 5 | 868±45 |
| | | K5-2* | 43 | 25 | 22 | 0 | 5 | 5 | 865±39 |
| | | K5-3 | 46 | 21 | 18 | 5 | 5 | 5 | 840±67 |
| | 6 | K6-1 | 43 | 22 | 22 | 0 | 8 | 5 | 882±22 |
| | | K6-2 | 43 | 22 | 23 | 0 | 7 | 5 | 883±47 |
| | | K6-3 | 43 | 20 | 21 | 0 | 9 | 7 | 828±48 |
| | 7 | K7-1 | 43 | 22 | 25 | 0 | 5 | 5 | 848±27 |
| | | K7-2 | 47 | 19 | 19 | 5 | 5 | 5 | 878±25 |
| | | K7-3 | 43 | 29 | 18 | 0 | 5 | 5 | 782±49 |

**Notes:** "*" and "◊" represent the same alloys found by Iteration Loop I and Iteration Loop II.

Table S4 is the original dataset of Al-Co-Cr-Cu-Fe-Ni HEAs system with the measured Vickers hardness which are collected from 30 different publications from 2004 to 2016. There is a discrepancy of the hardness values in terms of the same alloy from different literatures. For simplicity, we averaged the hardness values reported in different references if the relative error is less than 10% and we abandoned those with relative error larger than 10% (Table S5).

**Table.S4. The original training dataset used for machine learning**

| Alloy No. | Elemental content, at% | | | | | | Hardness, HV | Ref. |
|---|---|---|---|---|---|---|---|---|
| | Al | Co | Cr | Cu | Fe | Ni | | |
| 1 | 18.2 | 9.1 | 18.2 | 18.2 | 18.2 | 18.2 | 473 | 7 |
| 2 | 18.2 | 18.2 | 9.1 | 18.2 | 18.2 | 18.2 | 367 | 7 |
| 3 | 18.2 | 18.2 | 18.2 | 18.2 | 9.1 | 18.2 | 418 | 7 |
| 4 | 18.2 | 18.2 | 18.2 | 18.2 | 18.2 | 9.1 | 423 | 7 |
| 5 | 5.3 | 21.1 | 21.1 | 0.0 | 26.3 | 26.3 | 168 | 8 |
| 6 | 12.5 | 12.5 | 12.5 | 0.0 | 12.5 | 50.0 | 225 | 9 |
| 7 | 0.0 | 0.0 | 25.0 | 25.0 | 25.0 | 25.0 | 143 | 9 |
| 8 | 0.0 | 5.0 | 23.8 | 23.8 | 23.8 | 23.8 | 146 | 9 |
| 9 | 0.0 | 10.0 | 22.5 | 22.5 | 22.5 | 22.5 | 150 | 9 |
| 10 | 0.0 | 15.0 | 21.3 | 21.3 | 21.3 | 21.3 | 158 | 9 |
| 11 | 0.0 | 33.3 | 16.7 | 16.7 | 16.7 | 16.7 | 175 | 9 |
| 12 | 0.0 | 25.0 | 0.0 | 25.0 | 25.0 | 25.0 | 154 | 9 |
| 13 | 0.0 | 23.8 | 5.0 | 23.8 | 23.8 | 23.8 | 153 | 9 |
| 14 | 0.0 | 22.5 | 10.0 | 22.5 | 22.5 | 22.5 | 158 | 9 |
| 15 | 0.0 | 21.3 | 15.0 | 21.3 | 21.3 | 21.3 | 161 | 9 |
| 16 | 0.0 | 16.7 | 33.3 | 16.7 | 16.7 | 16.7 | 172 | 9 |
| 17 | 0.0 | 25.0 | 25.0 | 25.0 | 0.0 | 25.0 | 183 | 9 |
| 18 | 0.0 | 23.8 | 23.8 | 23.8 | 5.0 | 23.8 | 182 | 9 |
| 19 | 0.0 | 22.5 | 22.5 | 22.5 | 10.0 | 22.5 | 172 | 9 |
| 20 | 0.0 | 21.3 | 21.3 | 21.3 | 15.0 | 21.3 | 171 | 9 |
| 21 | 0.0 | 16.7 | 16.7 | 16.7 | 33.3 | 16.7 | 157 | 9 |
| 22 | 0.0 | 22.5 | 22.5 | 22.5 | 22.5 | 10.0 | 170 | 9 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 23 | 0.0 | 21.3 | 21.3 | 21.3 | 21.3 | 15.0 | 167 | 9 |
| 24 | 0.0 | 16.7 | 16.7 | 16.7 | 16.7 | 33.3 | 158 | 9 |
| 25 | 0.0 | 26.7 | 26.7 | 0.0 | 26.7 | 20.0 | 140 | 9 |
| 26 | 0.0 | 20.0 | 20.0 | 0.0 | 20.0 | 40.0 | 125 | 9 |
| 27 | 14.3 | 14.3 | 14.3 | 0.0 | 14.3 | 42.9 | 242 | 9 |
| 28 | 15.4 | 15.4 | 15.4 | 0.0 | 15.4 | 38.5 | 265 | 9 |
| 29 | 16.7 | 16.7 | 16.7 | 0.0 | 16.7 | 33.3 | 335 | 9 |
| 30 | 0.0 | 28.3 | 28.3 | 0.0 | 28.3 | 15.0 | 170 | 9 |
| 31 | 5.0 | 31.7 | 31.7 | 0.0 | 31.7 | 0.0 | 475 | 9 |
| 32 | 10.0 | 30.0 | 30.0 | 0.0 | 30.0 | 0.0 | 620 | 9 |
| 33 | 18.2 | 18.2 | 18.2 | 0.0 | 18.2 | 27.3 | 503 | 9 |
| 34 | 21.3 | 21.3 | 21.3 | 0.0 | 21.3 | 15.0 | 555 | 9 |
| 35 | 22.5 | 22.5 | 22.5 | 0.0 | 22.5 | 10.0 | 548 | 9 |
| 36 | 23.8 | 23.8 | 23.8 | 0.0 | 23.8 | 5.0 | 615 | 9 |
| 37 | 25.0 | 25.0 | 25.0 | 0.0 | 25.0 | 0.0 | 720 | 9 |
| 38 | 23.8 | 5.0 | 23.8 | 0.0 | 23.8 | 23.8 | 550 | 9 |
| 39 | 22.5 | 10.0 | 22.5 | 0.0 | 22.5 | 22.5 | 539 | 9 |
| 40 | 16.7 | 33.3 | 16.7 | 0.0 | 16.7 | 16.7 | 532 | 9 |
| 41 | 23.8 | 23.8 | 5.0 | 0.0 | 23.8 | 23.8 | 438 | 9 |
| 42 | 22.5 | 22.5 | 10.0 | 0.0 | 22.5 | 22.5 | 476 | 9 |
| 43 | 21.3 | 21.3 | 15.0 | 0.0 | 21.3 | 21.3 | 510 | 9 |
| 44 | 16.7 | 16.7 | 33.3 | 0.0 | 16.7 | 16.7 | 617 | 9 |
| 45 | 25.0 | 25.0 | 25.0 | 0.0 | 0.0 | 25.0 | 712 | 9 |
| 46 | 23.8 | 23.8 | 23.8 | 0.0 | 5.0 | 23.8 | 665 | 9 |
| 47 | 22.5 | 22.5 | 22.5 | 0.0 | 10.0 | 22.5 | 587 | 9 |
| 48 | 21.3 | 21.3 | 21.3 | 0.0 | 15.0 | 21.3 | 558 | 9 |
| 49 | 16.7 | 16.7 | 16.7 | 0.0 | 33.3 | 16.7 | 510 | 9 |
| 50 | 15.0 | 28.3 | 28.3 | 0.0 | 28.3 | 0.0 | 655 | 9 |
| 51 | 20.0 | 26.7 | 26.7 | 0.0 | 26.7 | 0.0 | 695 | 9 |
| 52 | 40.0 | 20.0 | 20.0 | 0.0 | 20.0 | 0.0 | 775 | 9 |
| 53 | 5.7 | 18.9 | 18.9 | 18.9 | 18.9 | 18.9 | 185 | 10 |
| 54 | 13.8 | 17.2 | 17.2 | 17.2 | 17.2 | 17.2 | 273 | 10 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 55 | 20.6 | 15.9 | 15.9 | 15.9 | 15.9 | 15.9 | 475 | 10 |
| 56 | 26.5 | 14.7 | 14.7 | 14.7 | 14.7 | 14.7 | 558 | 10 |
| 57 | 23.1 | 15.4 | 15.4 | 15.4 | 15.4 | 15.4 | 510 | 10 |
| 58 | 31.5 | 13.7 | 13.7 | 13.7 | 13.7 | 13.7 | 603 | 10 |
| 59 | 33.3 | 13.3 | 13.3 | 13.3 | 13.3 | 13.3 | 625 | 10 |
| 60 | 35.9 | 12.8 | 12.8 | 12.8 | 12.8 | 12.8 | 655 | 10 |
| 61 | 37.5 | 12.5 | 12.5 | 12.5 | 12.5 | 12.5 | 735 | 10 |
| 62 | 2.4 | 24.4 | 24.4 | 0.0 | 24.4 | 24.4 | 118 | 11 |
| 63 | 9.1 | 22.7 | 22.7 | 0.0 | 22.7 | 22.7 | 127 | 11 |
| 64 | 14.9 | 21.3 | 21.3 | 0.0 | 21.3 | 21.3 | 338 | 11 |
| 65 | 16.7 | 20.8 | 20.8 | 0.0 | 20.8 | 20.8 | 382 | 11 |
| 66 | 18.4 | 20.4 | 20.4 | 0.0 | 20.4 | 20.4 | 527 | 11 |
| 67 | 23.1 | 19.2 | 19.2 | 0.0 | 19.2 | 19.2 | 479 | 11 |
| 68 | 31.0 | 17.2 | 17.2 | 0.0 | 17.2 | 17.2 | 482 | 11 |
| 69 | 14.3 | 42.9 | 0.0 | 14.3 | 14.3 | 14.3 | 166 | 12 |
| 70 | 20.0 | 20.0 | 0.0 | 20.0 | 20.0 | 20.0 | 536 | 12 |
| 71 | 23.8 | 4.8 | 0.0 | 23.8 | 23.8 | 23.8 | 531 | 12 |
| 72 | 22.2 | 11.1 | 0.0 | 22.2 | 22.2 | 22.2 | 545 | 12 |
| 73 | 18.2 | 27.3 | 0.0 | 18.2 | 18.2 | 18.2 | 366 | 12 |
| 74 | 16.7 | 33.3 | 0.0 | 16.7 | 16.7 | 16.7 | 249 | 12 |
| 75 | 5.9 | 23.5 | 23.5 | 0.0 | 23.5 | 23.5 | 110 | 13 |
| 76 | 8.6 | 22.9 | 22.9 | 0.0 | 22.9 | 22.9 | 131 | 13 |
| 77 | 15.8 | 21.1 | 21.1 | 0.0 | 21.1 | 21.1 | 388 | 13 |
| 78 | 17.9 | 20.5 | 20.5 | 0.0 | 20.5 | 20.5 | 538 | 13 |
| 79 | 23.8 | 19.0 | 19.0 | 0.0 | 19.0 | 19.0 | 487 | 13 |
| 80 | 3.8 | 0.0 | 19.2 | 19.2 | 19.2 | 38.5 | 162 | 14 |
| 81 | 5.7 | 0.0 | 18.9 | 18.9 | 18.9 | 37.7 | 170 | 14 |
| 82 | 7.4 | 0.0 | 18.5 | 18.5 | 18.5 | 37.0 | 200 | 14 |
| 83 | 9.1 | 0.0 | 18.2 | 18.2 | 18.2 | 36.4 | 238 | 14 |
| 84 | 10.7 | 0.0 | 17.9 | 17.9 | 17.9 | 35.7 | 278 | 14 |
| 85 | 12.3 | 0.0 | 17.5 | 17.5 | 17.5 | 35.1 | 290 | 14 |
| 86 | 13.8 | 0.0 | 17.2 | 17.2 | 17.2 | 34.5 | 315 | 14 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 87 | 15.3 | 0.0 | 16.9 | 16.9 | 16.9 | 33.9 | 339 | 14 |
| 88 | 16.7 | 0.0 | 16.7 | 16.7 | 16.7 | 33.3 | 392 | 14 |
| 89 | 19.4 | 0.0 | 16.1 | 16.1 | 16.1 | 32.3 | 521 | 14 |
| 90 | 23.1 | 0.0 | 15.4 | 15.4 | 15.4 | 30.8 | 546 | 14 |
| 91 | 24.2 | 0.0 | 15.2 | 15.2 | 15.2 | 30.3 | 550 | 14 |
| 92 | 26.5 | 0.0 | 14.7 | 14.7 | 14.7 | 29.4 | 544 | 14 |
| 93 | 28.6 | 0.0 | 14.3 | 14.3 | 14.3 | 28.6 | 567 | 14 |
| 94 | 30.6 | 0.0 | 13.9 | 13.9 | 13.9 | 27.8 | 576 | 14 |
| 95 | 33.3 | 0.0 | 13.3 | 13.3 | 13.3 | 26.7 | 593 | 14 |
| 96 | 20.0 | 0.0 | 20.0 | 20.0 | 20.0 | 20.0 | 479 | 15 |
| 97 | 21.7 | 0.0 | 21.7 | 21.7 | 21.7 | 13.0 | 494 | 15 |
| 98 | 20.8 | 0.0 | 20.8 | 20.8 | 20.8 | 16.7 | 486 | 15 |
| 99 | 19.2 | 0.0 | 19.2 | 19.2 | 19.2 | 23.1 | 408 | 15 |
| 100 | 18.5 | 0.0 | 18.5 | 18.5 | 18.5 | 25.9 | 370 | 15 |
| 101 | 16.7 | 16.7 | 16.7 | 8.3 | 16.7 | 25.0 | 358 | 16 |
| 102 | 20.0 | 20.0 | 20.0 | 10.0 | 10.0 | 20.0 | 586 | 16 |
| 103 | 16.7 | 16.7 | 25.0 | 8.3 | 16.7 | 16.7 | 601 | 16 |
| 104 | 15.4 | 30.8 | 15.4 | 7.7 | 15.4 | 15.4 | 295 | 16 |
| 105 | 22.2 | 22.2 | 0.0 | 11.1 | 22.2 | 22.2 | 584 | 16 |
| 106 | 15.4 | 15.4 | 15.4 | 7.7 | 15.4 | 30.8 | 310 | 16 |
| 107 | 16.7 | 25.0 | 16.7 | 8.3 | 16.7 | 16.7 | 451 | 16 |
| 108 | 20.0 | 20.0 | 10.0 | 10.0 | 20.0 | 20.0 | 546 | 16 |
| 109 | 15.4 | 15.4 | 30.8 | 7.7 | 15.4 | 15.4 | 607 | 16 |
| 110 | 16.7 | 16.7 | 16.7 | 8.3 | 25.0 | 16.7 | 537 | 16 |
| 111 | 15.4 | 15.4 | 15.4 | 7.7 | 30.8 | 15.4 | 514 | 16 |
| 112 | 20.0 | 20.0 | 20.0 | 10.0 | 20.0 | 10.0 | 604 | 16 |
| 113 | 22.2 | 22.2 | 22.2 | 11.1 | 22.2 | 0.0 | 639 | 16 |
| 114 | 22.2 | 0.0 | 22.2 | 11.1 | 22.2 | 22.2 | 534 | 16 |
| 115 | 30.8 | 15.4 | 15.4 | 7.7 | 15.4 | 15.4 | 609 | 16 |
| 116 | 20.0 | 10.0 | 20.0 | 10.0 | 20.0 | 20.0 | 551 | 16 |
| 117 | 11.1 | 0.0 | 22.2 | 22.2 | 22.2 | 22.2 | 382 | 17 |
| 118 | 27.3 | 0.0 | 18.2 | 18.2 | 18.2 | 18.2 | 573 | 17 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 119 | 33.3 | 0.0 | 16.7 | 16.7 | 16.7 | 16.7 | 651 | 17 |
| 120 | 7.7 | 30.8 | 0.0 | 0.0 | 30.8 | 30.8 | 135 | 18 |
| 121 | 14.3 | 28.6 | 0.0 | 0.0 | 28.6 | 28.6 | 205 | 18 |
| 122 | 20.0 | 26.7 | 0.0 | 0.0 | 26.7 | 26.7 | 380 | 18 |
| 123 | 8.0 | 17.0 | 17.0 | 8.0 | 17.0 | 33.0 | 280 | 19 |
| 124 | 23.0 | 15.0 | 23.0 | 8.0 | 15.0 | 16.0 | 580 | 19 |
| 125 | 16.7 | 0.0 | 55.6 | 0.0 | 0.0 | 27.8 | 371 | 20 |
| 126 | 11.8 | 0.0 | 29.4 | 0.0 | 44.1 | 14.7 | 450 | 20 |
| 127 | 6.3 | 0.0 | 31.3 | 0.0 | 46.9 | 15.6 | 304 | 20 |
| 128 | 38.5 | 15.4 | 15.4 | 0.0 | 15.4 | 15.4 | 695 | 21 |
| 129 | 42.9 | 14.3 | 14.3 | 0.0 | 14.3 | 14.3 | 740 | 21 |
| 130 | 9.1 | 18.2 | 18.2 | 18.2 | 18.2 | 18.2 | 207(206/208) | 7+10 |
| 131 | 28.6 | 14.3 | 14.3 | 14.3 | 14.3 | 14.3 | 576(582/570) | 10+22 |
| 132 | 20.0 | 20.0 | 20.0 | 20.0 | 0.0 | 20.0 | 415(419/410) | 23+24 |
| 133 | 25.0 | 16.7 | 16.7 | 8.3 | 16.7 | 16.7 | 602(586/619) | 16+25 |
| 134 | 0.0 | 20.0 | 20.0 | 20.0 | 20.0 | 20.0 | 155(157/153) | 9+26 |
| 135 | 0.0 | 25.0 | 25.0 | 0.0 | 25.0 | 25.0 | 139(140/138) | 9+27 |
| 136 | 25.0 | 25.0 | 0.0 | 0.0 | 25.0 | 25.0 | 430(410/450) | 9+18 |
| 137 | 20.0 | 20.0 | 20.0 | 0.0 | 20.0 | 20.0 | 516(518/510/520) | 9+21+28 |
| 138 | 43.0 | 6.0 | 33.0 | 6.0 | 6.0 | 6.0 | 764 | * |
| 139 | 42.9 | 14.3 | 7.1 | 7.1 | 7.1 | 21.4 | 591 | * |
| 140 | 42.9 | 21.4 | 7.1 | 7.1 | 7.1 | 14.3 | 701 | * |
| 141 | 40.0 | 13.3 | 6.7 | 13.3 | 20.0 | 6.7 | 768 | * |
| 142 | 46.2 | 15.4 | 7.7 | 7.7 | 15.4 | 7.7 | 702 | * |
| 143 | 42.9 | 14.3 | 14.3 | 7.1 | 7.1 | 14.3 | 720 | * |
| 144 | 42.9 | 7.1 | 14.3 | 7.1 | 14.3 | 14.3 | 694 | * |
| 145 | 7.0 | 0.0 | 23.3 | 0.0 | 23.3 | 46.5 | 149 | * |
| 146 | 11.1 | 0.0 | 22.2 | 0.0 | 22.2 | 44.4 | 229 | * |
| 147 | 13.0 | 0.0 | 28.0 | 22.0 | 6.0 | 31.0 | 459 | * |
| 148 | 12.0 | 0.0 | 31.0 | 21.0 | 5.0 | 31.0 | 469 | * |
| 149 | 12.0 | 0.0 | 31.0 | 20.0 | 5.0 | 32.0 | 483 | * |
| 150 | 10.0 | 0.0 | 35.0 | 25.0 | 5.0 | 25.0 | 472 | * |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 151 | 10.0 | 0.0 | 35.0 | 26.0 | 5.0 | 24.0 | 454 | * |
| 152 | 10.0 | 0.0 | 35.0 | 24.0 | 5.0 | 26.0 | 441 | * |
| 153 | 11.0 | 0.0 | 29.0 | 29.0 | 5.0 | 26.0 | 495 | * |
| 154 | 11.0 | 0.0 | 28.0 | 29.0 | 7.0 | 25.0 | 477 | * |
| 155 | 11.0 | 0.0 | 28.0 | 27.0 | 6.0 | 28.0 | 469 | * |

**Notes:** "*" represents the experimental data of HEAs from our laboratory.

**Table.S5. The deleted data during data cleaning**

| Alloy | Elemental content, at% | | | | | | Hardness, HV | Ref. |
|---|---|---|---|---|---|---|---|---|
| No. | Al | Co | Cr | Cu | Fe | Ni | | |
| 156 | 0.0 | 22.2 | 22.2 | 11.1 | 22.2 | 22.2 | 134/174 | 16+29 |
| 157 | 7.0 | 23.3 | 23.3 | 0.0 | 23.3 | 23.3 | 125/149 | 11+30 |
| 158 | 22.2 | 22.2 | 22.2 | 11.1 | 0.0 | 22.2 | 496/564 | 16+31 |
| 159 | 25.0 | 0.0 | 25.0 | 0.0 | 25.0 | 25.0 | 472/558 | 9+32 |
| 160 | 27.3 | 18.2 | 18.2 | 0.0 | 18.2 | 18.2 | 482/575 | 11+21 |
| 161 | 10.0 | 20.0 | 20.0 | 10.0 | 20.0 | 20.0 | 204/290 | 16+25 |
| 162 | 33.3 | 16.7 | 16.7 | 0.0 | 16.7 | 16.7 | 510/610 | 11+21 |
| 163 | 18.2 | 18.2 | 18.2 | 9.1 | 18.2 | 18.2 | 458/563/665 | 7+16+25 |
| 164 | 11.1 | 22.2 | 22.2 | 0.0 | 22.2 | 22.2 | 160/198/247 | 11+13+33 |
| 165 | 16.7 | 16.7 | 16.7 | 16.7 | 16.7 | 16.7 | 364/405/410/416 /420/463/470/515 | 7+10+22+23 24+34+35+36 |

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