Supplementary information to *Machine-learning informed prediction of high-entropy solid solution formation: Beyond the Hume-Rothery rules*

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1. Input data for machine learning

The input data consist of two parts. One is the 85 elemental properties for all the elements in the Periodic Table of Elements, taken from Ref. [1]. The other is the tabulated information of 1252 multicomponent alloys on whether they are solid solutions or not. We attach a file to elaborate the information for the 1252 alloys.

2. Prediction by the new rule

We use the new rule $\gamma \geq 1$ to predict solid solution alloys with ≥ 4 components for the three 9-element blocks (see main text for the blocks). Here is a list of all the combinations predicted to be solid solutions. The alloys are arranged by the crystal structures and their numbers of components.

FCC(5)

Ni1Ir1Au1Pd1Pt1 Ni1Ir1Rh1Pd1Pt1 Ni1Cu1Au1Pd1Pt1 Ni1Ir1Pd1Pt1Cu1

Au1Ir1Co1Pd1Pt1 Rh1Ir1Co1Pd1Pt1 Au1Cu1Co1Pd1Pt1 Ir1Co1Pd1Pt1Cu1
Ni1Pd1Co1Au1Pt1 Ni1Ir1Co1Pd1Pt1 Ni1Cu1Co1Pd1Pt1 Ni1Ir1Co1Cu1Pd1

BCC(5)

Cr1Nb1Mo1W1Ta1 Nb1Ta1Mo1W1V1 Nb1Ta1Hf1Zr1V1 Cr1Mo1Ta1W1V1
Cr1Nb1Ta1W1V1 Cr1Nb1Mo1W1V1 Cr1Nb1Ta1Mo1V1 Nb1Ta1Mo1W1Ti1
Nb1Ta1Hf1Zr1Ti1 Cr1Mo1Ta1W1Ti1 Cr1Nb1Ta1W1Ti1 Cr1Nb1Mo1W1Ti1
Cr1Nb1Ta1Mo1Ti1 Mo1V1Ta1W1Ti1 Nb1V1Ta1W1Ti1 Nb1V1Mo1W1Ti1
Nb1V1Ta1Mo1Ti1 Cr1V1Ta1W1Ti1 Cr1Mo1V1W1Ti1 Cr1Mo1V1Ta1Ti1
Cr1Nb1V1W1Ti1 Cr1Nb1V1Ta1Ti1 Cr1Nb1V1Mo1Ti1

HCP(5)

Ru1Ir1Os1Rh1Re1 Re1Ir1Os1Tc1Rh1 Ru1Ir1Os1Tc1Re1 Ru1Ir1Os1Tc1Rh1
Ru1Ir1Rh1Tc1Re1 Ru1Os1Tc1Rh1Re1 Os1Ir1Co1Rh1Re1 Os1Ru1Ir1Co1Re1
Os1Ru1Ir1Co1Rh1 Ru1Ir1Co1Rh1Re1 Os1Ru1Co1Rh1Re1 Re1Ir1Os1Fe1Rh1
Ru1Ir1Os1Fe1Re1 Ru1Ir1Os1Fe1Rh1 Ru1Ir1Rh1Fe1Re1 Ru1Os1Fe1Rh1Re1
Re1Ir1Os1Fe1Tc1 Rh1Ir1Os1Fe1Tc1 Re1Ir1Rh1Fe1Tc1 Re1Os1Fe1Rh1Tc1
Ru1Ir1Os1Fe1Tc1 Tc1Ru1Ir1Fe1Re1 Tc1Ru1Os1Fe1Re1 Ru1Ir1Rh1Fe1Tc1
Ru1Os1Fe1Rh1Tc1 Tc1Ru1Rh1Fe1Re1 Ru1Ir1Co1Fe1Re1 Ru1Ir1Co1Fe1Rh1
Os1Ru1Co1Fe1Rh1 Re1Rh1Ir1Os1Mn1 Ru1Ir1Os1Mn1Re1 Ru1Rh1Ir1Os1Mn1
Ru1Ir1Rh1Mn1Re1 Re1Ir1Os1Tc1Mn1 Rh1Ir1Os1Tc1Mn1 Re1Ir1Rh1Tc1Mn1
Re1Mn1Os1Tc1Rh1 Ru1Ir1Os1Tc1Mn1 Ru1Ir1Tc1Mn1Re1 Ru1Mn1Os1Tc1Re1
Ru1Ir1Rh1Tc1Mn1 Ru1Mn1Os1Tc1Rh1 Ru1Mn1Rh1Tc1Re1 Ru1Ir1Co1Mn1Re1
Ru1Rh1Ir1Co1Mn1 Ru1Ir1Fe1Mn1Re1 Ru1Ir1Rh1Fe1Mn1 Ru1Mn1Os1Fe1Rh1
Ru1Ir1Fe1Mn1Tc1 Ru1Mn1Rh1Fe1Tc1 Ru1Ir1Co1Fe1Mn1 Os1Ru1Mn1Co1Fe1
Ru1Mn1Co1Fe1Re1 Ru1Mn1Co1Fe1Rh1

FCC(6)

Ni1Co1Pt1Ir1Au1Pd1 Ni1Co1Pt1Ir1Pd1Rh1 Ni1Co1Pt1Au1Pd1Cu1 Ni1Co1Pt1Ir1Pd1Cu1

BCC(6)

Nb1W1V1Cr1Mo1Ta1 Nb1W1Ti1Cr1Mo1Ta1 Nb1Ti1W1V1Mo1Ta1 Mo1Ti1W1V1Cr1Ta1

Nb1Ti1W1V1Cr1Ta1 Nb1Ti1W1V1Cr1Mo1 Nb1Ti1V1Cr1Mo1Ta1

HCP(6)

Ru1Rh1Ir1Re1Os1Tc1 Ru1Co1Rh1Ir1Re1Os1 Ru1Rh1Ir1Re1Fe1Os1 Rh1Ir1Re1Fe1Os1Tc1

Ru1Ir1Re1Fe1Os1Tc1 Ru1Rh1Ir1Fe1Os1Tc1 Ru1Ir1Re1Fe1Rh1Tc1 Ru1Rh1Re1Fe1Os1Tc1

Ru1Co1Ir1Re1Fe1Os1 Ru1Co1Rh1Ir1Fe1Os1 Ru1Co1Ir1Re1Fe1Rh1 Ru1Mn1Os1Ir1Re1Rh1

Mn1Os1Ir1Re1Rh1Tc1 Ru1Mn1Ir1Re1Os1Tc1 Ru1Mn1Os1Ir1Rh1Tc1 Ru1Mn1Ir1Re1Rh1Tc1

Ru1Rh1Mn1Re1Os1Tc1 Ru1Mn1Co1Rh1Ir1Os1 Ru1Mn1Co1Ir1Re1Rh1 Ru1Mn1Os1Ir1Fe1Rh1

Ru1Mn1Ir1Re1Fe1Rh1 Ru1Mn1Ir1Fe1Os1Tc1 Ru1Mn1Ir1Re1Fe1Tc1 Ru1Mn1Re1Fe1Os1Tc1

Ru1Mn1Ir1Fe1Rh1Tc1 Ru1Rh1Mn1Fe1Os1Tc1 Ru1Mn1Re1Fe1Rh1Tc1 Ru1Mn1Co1Ir1Fe1Os1

Ru1Mn1Co1Ir1Re1Fe1 Ru1Mn1Co1Ir1Fe1Rh1 Ru1Co1Rh1Mn1Fe1Os1 Ru1Co1Mn1Re1Fe1Rh1

FCC(7)

Ni1Co1Pt1Ir1Pd1Rh1Cu1

BCC(7)

Nb1Ti1W1V1Cr1Mo1Ta1

HCP(7)

Ru1Co1Rh1Ir1Re1Os1Tc1 Ru1Rh1Ir1Re1Fe1Os1Tc1 Ru1Co1Rh1Ir1Re1Fe1Os1

Ru1Mn1Os1Ir1Re1Rh1Tc1 Ru1Mn1Co1Rh1Ir1Re1Os1 Ru1Mn1Rh1Ir1Re1Fe1Os1

Mn1Os1Ir1Re1Fe1Rh1Tc1 Ru1Mn1Ir1Re1Fe1Os1Tc1 Ru1Mn1Os1Ir1Fe1Rh1Tc1

Ru1Mn1Ir1Re1Fe1Rh1Tc1 Ru1Rh1Mn1Re1Fe1Os1Tc1 Ru1Mn1Co1Rh1Ir1Fe1Os1

Ru1Mn1Co1Ir1Re1Fe1Rh1

FCC(8,9)

None

BCC(8,9)

None

HCP(8,9)

Ru1Co1Rh1Ir1Re1Fe1Os1Tc1 Ru1Mn1Rh1Ir1Re1Fe1Os1Tc1 Ru1Mn1Co1Rh1Ir1Re1Fe1Os1

3. Validation of the new rule by CALPHAD

CALPHAD (acronym of Calculation of Phase Diagrams) calculations were carried out using TCNI8 thermodynamic database provided by ThermoCalc^{TM} [2]. The database covers the whole composition ranges of all constituent binaries and limited ternaries. The TCNI8 database does not contain elements Os, Tc, Rh, Ir, Au and Ag, and hence only HEA compositions that do not contain these 6 elements are considered. This limits our validation to 77 equimolar solid solution HEAs, comprising 4, 5 or 6 components. Among these 77 HEAs, 72 are successfully validated by CALPHAD calculations comprising 5 FCC and 66 BCC HEAs, with a consistency as high as 94%.

Among those equimolar alloys predicted by our new thermodynamic rule, some are already confirmed experimentally, including NbTaMoWV [3], MoNbTaTiVW [4], CrMoNbTaVW [5]. Septenary CrMoNbTaTiVW was previously suggested by Gao et al. [6] using CALPHAD method. As supplementary information, we give seven examples to demonstrate the successful predictions in Figure S1. These alloys are CoCuNiPdPt, CrMoNbTiV, MoNbTiVW, CrMoNbTaTiW, CrNbTaTiVW, CrMoNbTiVW and CrMoNbTaTiV. All these alloys show a temperature range of 600-2000 °C for solid solution phase. 72 out of the total 77 alloys have the similar feature.

References

[1] http://periodictable.com., Accessed: 2018-03-10.

[2] B. Sundman, B. Jansson, J.-O. Andersson, The thermo-calc databank system, Calphad 9 (1985) 153 – 190.

[3] O. Senkov, G. Wilks, D. Miracle, C. Chuang, P. Liaw, Refractory high-entropy alloys, Intermetallics 18 (2010) 1758 – 1765.

[4] B. Zhang, M. C. Gao, Y. Zhang, S. Yang, S. M. Guo, Senary refractory high entropy alloy MoNbTaTiVW, Materials Science and Technology 31 (2015) 1207–1213.

[5] B. Zhang, M. Gao, Y. Zhang, S. Guo, Senary refractory high-entropy alloy CrxMoNbTaVW, Calphad 51 (2015) 193 – 201.

[6] M. C. Gao, P. Gao, J. A. Hawk, L. Ouyang, D. E. Alman, M. Widom, Computational modeling of high-entropy alloys: Structures, thermodynamics and elasticity, Journal of Materials Research 32 (2017) 36273641.

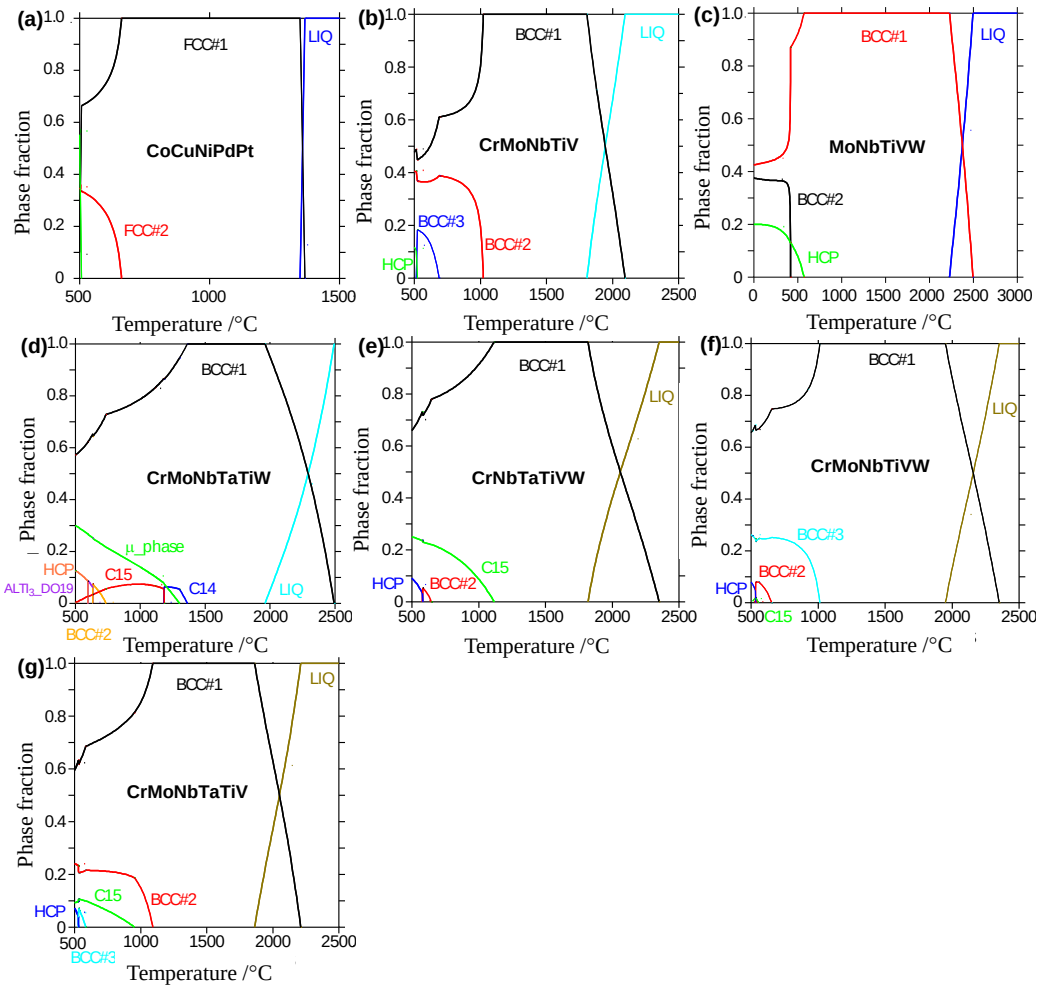

Figure S1: Seven single-phase examples predicted by the new rule and validated by the CALPHAD method.