Supplementary Information

Effect of Ta addition on the structural, thermodynamic and mechanical properties of CoCrFeNi high entropy alloys

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Table S1 SQSs of CoCrFeNiTa<sub>x</sub> (<i>x</i> = 0.0, 0.2, 0.4, 0.6, 0.8 and 1.0) structures containing 20/21/22/23/24/25 atoms, respectively. Lattice vectors and atomic positions are given in Cartesian coordinates in Å. Atomic positions represents the ideal, unrelaxed sites.

|                  | CoCrFeNi     | CoCrFeNiTa<sub>0.2</sub> |
|------------------|--------------|----------------------------|
|                  | bcc          | fcc                        | bcc                   | fcc                        |
| Lattice vectors  |              |                            |                       |                            |
| (1.5, 1.5, -0.5) | (1.5, 0.5, 0.0) | (1.5, -0.5, -0.5) | (-2.0, 1.5, -0.5)    |
| (1.0, -2.0, 0.0) | (-0.5, 1.5, 0.0) | (0.0, -2.0, 1.0) | (1.5, -2.0, -0.5)    |
| (-1.0, 0.0, -2.0) | (0.0, 0.0, 2.0) | (-0.5, -1.5, -2.5) | (-1.0, -1.0, 1.0)    |
|                  |              |                            |                       |                            |
| Atomic positions |              |                            |                       |                            |
| (0.0, -1.0, -2.0) Ni | (1.0, 2.0, 1.0) Ni | (0.5, -3.5, -1.5) Cr | (-2.0, 0.0, -0.0) Ni |  |
| (1.0, 1.0, -2.0) Cr | (0.0, 0.5, 1.5) Ni | (0.0, -3.0, -1.0) Co | (-1.5, -0.5, 0.0) Ni |  |
| (0.5, -1.5, -1.5) Co | (1.0, 2.0, 2.0) Co | (0.0, -2.0, -2.0) Ni | (-1.0, -1.0, 0.0) Fe |  |
| (1.5, 0.5, -1.5) Ni | (0.0, 0.5, 0.5) Cr | (1.0, -3.0, -2.0) Fe | (-1.0, -0.5, 0.5) Fe |  |
| (2.0, 0.0, -1.0) Fe | (0.0, 1.0, 1.0) Cr | (0.5, -2.5, -1.5) Co | (-0.5, -1.5, 0.0) Ni |  |
| (0.0, 0.0, -2.0) Fe | (0.0, 1.5, 1.5) Co | (0.0, -2.0, -1.0) Ta | (-0.5, -1.0, 0.5) Ta |  |
| (1.0, -1.0, -2.0) Cr | (0.5, 0.5, 1.0) Fe | (1.0, -3.0, -1.0) Fe | (0.0, -2.0, 0.0) Fe |  |
| (0.5, -0.5, -1.5) Ni | (0.5, 1.0, 1.5) Ni | (0.5, -2.5, -0.5) Cr | (-1.5, 0.5, 0.0) Co |  |
| (0.0, 0.0, -1.0) Co | (1.0, 0.5, 1.5) Fe | (0.0, -2.0, -0.0) Ni | (-1.0, -0.5, -0.5) Ni |  |
| (1.0, -1.0, -1.0) Cr | (0.0, 1.0, 2.0) Cr | (1.0, -2.0, -2.0) Ni | (-1.0, 0.0, 0.0) Cr |  |
| (0.5, -0.5, -0.5) Fe | (0.0, 1.5, 0.5) Fe | (0.5, -1.5, -1.5) Ni | (-0.5, -1.0, -0.5) Cr |  |
| (1.5, -0.5, -2.5) Cr | (0.5, 0.5, 2.0) Cr | (0.0, -1.0, -1.0) Co | (-0.5, -0.5, 0.0) Co |  |
| (1.0, 0.0, -2.0) Ni | (0.5, 1.0, 0.5) Co | (1.0, -2.0, -1.0) Ni | (0.0, -1.0, 0.0) Fe |  |
| (0.5, 0.5, -1.5) Fe | (0.5, 1.5, 1.0) Fe | (0.5, -1.5, -0.5) Cr | (0.5, -1.5, 0.0) Co |  |
| (1.5, -0.5, -1.5) Ni | (1.0, 0.5, 0.5) Co | (0.0, -1.0, -0.0) Cr | (-2.5, 0.0, 0.5) Cr |  |
| Lattice vectors | bcc       | fcc       | bcc       | fcc       |
|----------------|-----------|-----------|-----------|-----------|
| (-1.5, -1.5, 0.5) | (0.5, 0.0, -1.5) | (-1.5, 0.5, -1.5) | (-0.5, 1.5, 0.0) |
| (1.5, -0.5, 1.5) | (-1.0, 1.0, -1.0) | (0.0, -2.0, -1.0) | (0.0, 0.5, -1.5) |
| (-1.0, 2.0, 1.0) | (1.0, 2.0, 0.0) | (-2.0, -1.0, 1.0) | (-2.5, 0.0, -0.5) |

| Atomic positions | bcc       | fcc       | bcc       | fcc       |
|------------------|-----------|-----------|-----------|-----------|
| (-1.0, -1.0, 1.0) Ni | (0.5, 3.0, -2.5) Cr | (-3.0, -2.0, -1.0) Fe | (-2.5, 0.5, -1.0) Co |
| (-1.5, -0.5, 1.5) Co | (0.0, 0.5, -0.5) Co | (-2.5, -2.5, -0.5) Co | (-2.5, 1.0, -0.5) Co |
| (-0.5, -1.5, 1.5) Ni | (-0.5, 1.0, -1.5) Cr | (-2.5, -1.5, -1.5) Ni | (-2.0, 0.5, -0.5) Fe |
| (-0.5, -0.5, 0.5) Cr | (-0.5, 1.5, -1.0) Co | (-3.0, -1.0, -1.0) Ta | (-2.5, 1.0, -1.5) Fe |
| (-1.0, 0.0, 1.0) Cr | (0.0, 0.5, -1.5) Cr | (-2.0, -2.0, -1.0) Ni | (-2.5, 1.5, -1.0) Co |
| (-1.5, 0.5, 1.5) Cr | (0.0, 1.0, -1.0) Ni | (-2.5, -1.5, -0.5) Ni | (-2.0, 0.5, -1.5) Ni |
| (-1.0, 1.0, 2.0) Fe | (0.5, 1.5, -2.0) Cr | (-1.5, -2.5, -0.5) Co | (-2.0, 1.0, -1.0) Fe |
| (-0.5, -0.5, 1.5) Fe | (0.5, 0.5, -1.0) Co | (-2.0, -2.0, -0.0) Fe | (-2.0, 1.5, -0.5) Cr |
| (-1.0, 0.0, 2.0) Fe | (0.5, 1.0, -0.5) Ni | (-1.5, -1.5, -1.5) Ta | (-1.5, 0.5, -1.0) Ta |
| (0.0, -1.0, 2.0) Ta | (0.0, 1.0, -2.0) Fe | (-2.0, -1.0, -1.0) Co | (-1.5, 1.0, -0.5) Ni |
| (-0.5, -0.5, 2.5) Ni | (0.0, 1.5, -1.5) Ni | (-2.5, -0.5, -0.5) Cr | (-1.0, 0.5, -0.5) Ta |
| Lattice vectors | CoCrFeNiTa<sub>0.8</sub> | CoCrFeNiTa<sub>1.0</sub> | CoCrFeNiTa<sub>0.8</sub> | CoCrFeNiTa<sub>1.0</sub> |
|----------------|------------------------|------------------------|------------------------|------------------------|
| (-1.0, 0.0, 3.0) Cr | (0.0, 2.0, -1.0) Fe | (-1.0, -2.0, -1.0) Ni | (-3.0, 2.0, -2.0) Cr |
| (-0.5, 0.5, 0.5) Ni | (0.5, 1.0, -1.5) Ta | (-1.5, 1.5, -0.5) Fe | (-2.0, 1.5, -1.5) Fe |
| (-1.0, 1.0, 1.0) Co | (0.5, 1.5, -1.0) Fe | (-2.0, -1.0, -0.0) Co | (-1.5, 1.0, -1.5) Fe |
| (0.0, 0.0, 1.0) Ni | (0.5, 2.0, -0.5) Cr | (-1.5, -0.5, -1.5) Fe | (-1.5, 1.5, -1.0) Cr |
| (-0.5, 0.5, 1.5) Co | (0.0, 2.0, -2.0) Fe | (-2.0, 0.0, -1.0) Cr | (-1.0, 0.5, -1.5) Ni |
| (0.5, -0.5, 1.5) Fe | (0.0, 2.5, -1.5) Fe | (-1.0, -1.0, -1.0) Cr | (-1.0, 1.0, -1.0) Cr |
| (0.0, 0.0, 2.0) Fe | (0.5, 2.0, -1.5) Co | (-1.5, -0.5, -0.5) Co | (-1.0, 1.5, -0.5) Ni |
| (-0.5, 0.5, 2.5) Co | (0.5, 2.5, -1.0) Ta | (-0.5, -1.5, -0.5) Ta | (-0.5, 0.5, -1.0) Ni |
| (-0.5, 1.5, 1.5) Ta | (1.0, 1.5, -1.5) Ni | (-1.0, -1.0, -0.0) Fe | (-0.5, 1.0, -0.5) Co |
| (0.5, 0.5, 1.5) Cr | (1.0, 2.0, -1.0) Co | (-1.0, 0.0, -1.0) Ta | (-1.0, 1.5, -1.5) Co |
| (0.0, 1.0, 2.0) Co | (0.5, 2.5, -2) Ni | (-0.5, -0.5, -0.5) Ni | (-0.5, 1.0, -1.5) Cr |
|                  | (-3.5, -2.5, -1.5)  | (-0.5, 1.5, -1.0) Ta  |               |

| Atomic positions | CoCrFeNiTa<sub>0.8</sub> | CoCrFeNiTa<sub>1.0</sub> | CoCrFeNiTa<sub>0.8</sub> | CoCrFeNiTa<sub>1.0</sub> |
|------------------|------------------------|------------------------|------------------------|------------------------|
| (-0.5, -0.5, -1.5) Co | (0.0, -1.0, 0.0) Fe | (-1.5, -0.5, 1.5) Cr | (1.5, -2.5, 1.0) Ta |
| (1.0, 1.0, -2.0) Ta | (0.0, -1.0, -1.0) Ni | (-2.0, 0.0, 2.0) Ta | (0.0, -1.5, 0.5) Co |
| (0.0, 0.0, -2.0) Co | (0.0, -0.5, -0.5) Fe | (-1.0, -1.0, 2.0) Ni | (0.5, -2.5, 0.0) Cr |
| (-0.5, 0.5, -1.50) Ni | (1.5, -1.5, -2.0) Fe | (-1.0, 0.0, 1.0) Fe | (0.5, -2.0, 0.5) Co |
| (-1.0, 1.0, -1.0) Cr | (0.5, -1.0, -0.5) Co | (-1.5, 0.5, 1.5) Cr | (1.0, -3.0, 0.0) Co |
|                  |                  |                  |                  |
|-----------------|-----------------|-----------------|-----------------|
| (0.0, 0.0, -1.0) Fe | (0.5, -0.5, -0.0) Cr | (-2.0, 1.0, 2.0) Ta | (1.0, -2.5, 0.5) Ta |
| (-0.5, 0.5, -0.5) Fe | (1.0, -1.0, -0.0) Ni | (-0.5, -0.5, 1.5) Ta | (1.0, -2.0, 1.0) Cr |
| (1.0, 2.0, -1.0) Fe | (0.5, -1.0, -1.5) Ta | (-1.0, 0.0, 2.0) Ni | (1.5, -3.0, 0.5) Cr |
| (0.0, 2.0, -2.0) Co | (0.5, -0.5, -1.0) Co | (-1.5, 0.5, 2.5) Co | (1.5, -1.5, 1.0) Co |
| (0.5, 0.5, -2.5) Ni | (0.0, -1.5, -0.5) Ta | (0.0, -1.0, 2.0) Co | (2.0, -2.5, 0.5) Fe |
| (0.0, 1.0, -2.0) Co | (1.0, -1.0, -1.0) Cr | (-0.5, -0.5, 2.5) Fe | (0.5, -1.5, 0.0) Cr |
| (-0.5, 1.5, -1.5) Ta | (0.5, -0.5, -2.0) Ni | (-1.0, 0.0, 3.0) Cr | (0.5, -1.0, 0.5) Ni |
| (0.5, 0.5, -1.5) Ni | (0.0, -1.5, -1.5) Ni | (-1.5, 0.5, 3.5) Ni | (1.0, -2.0, 0.0) Fe |
| (-0.0, 1.0, -1.0) Fe | (1.0, -1.0, -2.0) Co | (-0.5, 0.5, 0.5) Fe | (1.0, -1.5, 0.5) Ni |
| (-0.5, 1.5, -0.5) Cr | (1.0, -0.5, -1.5) Ta | (-1.0, 1.0, 1.0) Ni | (1.5, -2.5, 0.0) Fe |
| (0.5, 0.5, -0.5) Co | (0.5, -1.5, -1.0) Fe | (-1.5, 1.5, 1.5) Ni | (1.5, -2.0, 0.5) Ni |
| (0.0, 1.0, 0.0) Ni | (1.5, -1.0, -1.5) Co | (0.0, 0.0, 1.0) Fe | (2.0, -3.5, 0.5) Cr |
| (-0.5, 1.5, 0.5) Cr | (1.0, -0.5, -1.5) Ta | (-0.5, 0.5, 1.5) Ta | (2.0, -1.5, 0.5) Ta |
| (0.5, 1.5, -1.5) Ta | (0.5, -1.5, -1.0) Fe | (-1.0, 1.0, 2.0) Co | (0.5, -0.5, 0.0) Fe |
| (0.0, 2.0, -1.0) Ni | (1.5, -0.5, -1.0) Co | (0.5, -0.5, 1.5 Co | (1.0, -1.5, 0.0) Co |
| (1.0, 1.0, -1.0) Fe | (1.0, -1.5, -0.5) Fe | (0.0, 0.0, 2.0) Ta | (1.0, -1.0, 0.0) Fe |
| (0.5, 1.5, -0.5) Ta | (0.5, -1.5, -2.0) Cr | (-0.5, 0.5, 2.5) Cr | (1.0, -0.5, 0.5) Ni |
| (0.0, 2.0, 0.0) Cr | (1.5, -0.5, -2.0) Ni | (-0.5, 1.5, 1.5) Cr | (1.5, -1.5, 0.0) Ni |
| (0.5, 2.5, -0.5) Cr | (1.0, -1.5, -1.5) Cr | (0.5, 0.5, 1.5 Co | (1.5, -1.0, 0.5) Ta |
|                  |                  |                  |                  |
|                  |                  |                  |                  |
**Fig. S1** Partial pair distribution function of fcc CoCrFeNi alloy.
Fig. S2 Partial pair distribution function of bcc CoCrFeNi alloy.
Fig. S3 Partial pair distribution function of fcc CoCrFeNiTa₀.₂ alloy.
Fig. S4 Partial pair distribution function of bcc CoCrFeNiTa_{0.2} alloy.
Fig. S5 Partial pair distribution function of fcc CoCrFeNiTa$_{0.4}$ alloy.
Fig. S6 Partial pair distribution function of bcc CoCrFeNiTa<sub>0.4</sub> alloy.
Fig. S7 Partial pair distribution function of fcc CoCrFeNiTa_{0.6} alloy.
Fig. S8 Partial pair distribution function of \( bcc \) CoCrFeNiTa\(_{0.6}\) alloy.
Fig. S9 Partial pair distribution function of fcc CoCrFeNiTa$_{0.8}$ alloy.
Fig. S10 Partial pair distribution function of \(bce\) CoCrFeNiTa\(_{0.8}\) alloy.
Fig. S11 Partial pair distribution function of $fcc$ CoCrFeNiTa$_{1.0}$ alloy.
Fig. S12 Partial pair distribution function of $bcc$ CoCrFeNiTa$_{1.0}$ alloy.
**Fig. S13** Phonon density of states for *fcc* (left) and *bcc* (right) structures. $x$ represents the Ta content in CoCrFeNiTa$_x$ alloys.
Fig. S14 Band structures of fcc CoCrFeNiTa$_x$. 
Fig. S15 Band structures of $bcc$ CoCrFeNiTa.
Fig. S16 Density of states of fcc and bcc CoCrFeNiTa.
Appendix:

According to Boltzmann's hypothesis,\textsuperscript{1,2} the configurational entropy of an n-element regular solution is as follows:

\[ \Delta S_{\text{con}} = -R \sum_{i=1}^{n} (C_i \ln C_i) \]

(1)

where \( C_i \) is mole percent of element \( i \), \( i = 1 \) to \( n \), and \( R \) is the ideal gas constant.

1 R.A. Swalin, \textit{Wiley}, New York, 1972, 35-41.

2 X. Yang and Y. Zhang, \textit{Mater. Chem. Phys.}, 2012, 132, 233-238.