Supplementary Material for

# DFT study of the hexagonal high-entropy alloy fission product system

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Table S1

## Supplementary Methods

Five Mo and three Ru were removed from the 54 atom compositions previously studied to simulate the segregation of the $Mo_5Ru_3$ σ phase in ~15 % increments. The resulting stoichiometries were then normalised to maintain 54 atoms and the Gibbs free energy was calculated through a weighted sum of the formation enthalpies (obtained by DFT) of the σ phase and normalised solid solution (accounting for the configuration entropy at 1973 K).

**Table S1**. The compositions of the simulated dual phase σ + solid solution.

| [Mo] (at. %) | σ phase (vol. frac.) | Number of atoms in supercell | | | | | ΔG at 1973 K (eV/atom) |
|---|---|---|---|---|---|---|---|
| | | **Mo** | **Pd** | **Rh** | **Ru** | **Tc** | |
| 15 | 0 | 8 | 7 | 7 | 32 | - | $-1.87\times10^{-1}$ |
| | 0.15 | 4 | 8 | 8 | 34 | - | $-1.37\times10^{-1}$ |
| 26 | 0 | 14 | 6 | 6 | 28 | - | $-2.00\times10^{-1}$ |
| | 0.15 | 11 | 7 | 7 | 29 | - | $-2.13\times10^{-1}$ |
| | 0.30 | 6 | 9 | 8 | 31 | - | $-1.60\times10^{-1}$ |
| 37 | 0 | 20 | 5 | 5 | 24 | - | $-1.83\times10^{-1}$ |
| | 0.15 | 18 | 6 | 5 | 25 | - | $-1.96\times10^{-1}$ |
| | 0.30 | 14 | 7 | 7 | 26 | - | $-1.98\times10^{-1}$ |
| | 0.44 | 9 | 9 | 9 | 27 | - | $-2.07\times10^{-1}$ |
| 52 | 0 | 28 | 4 | 4 | 18 | - | $-1.11\times10^{-1}$ |
| | 0.15 | 27 | 5 | 4 | 18 | - | $-1.25\times10^{-1}$ |
| | 0.30 | 26 | 6 | 5 | 17 | - | $-1.47\times10^{-1}$ |
| | 0.44 | 23 | 7 | 8 | 16 | - | $-2.17\times10^{-1}$ |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | 0.59 | 20 | 10 | 9 | 15 | - | $-2.40\times10^{-1}$ |
| | 0.74 | 12 | 15 | 15 | 12 | - | $-2.47\times10^{-1}$ |
| 57 | 0 | 34 | 3 | 3 | 14 | - | $-4.03\times10^{-2}$ |
| | 0.15 | 34 | 4 | 3 | 13 | - | $-4.16\times10^{-2}$ |
| | 0.30 | 34 | 4 | 5 | 11 | - | $-5.60\times10^{-2}$ |
| | 0.44 | 34 | 5 | 6 | 9 | - | $-8.00\times10^{-2}$ |
| | 0.59 | 34 | 7 | 8 | 5 | - | $-1.00\times10^{-1}$ |
| 76 | 0 | 41 | 2 | 2 | 9 | - | $8.69\times10^{-3}$ |
| | 0.15 | 42 | 2 | 3 | 7 | - | $4.72\times10^{-2}$ |
| | 0.30 | 44 | 3 | 3 | 4 | - | $-8.35\times10^{-2}$ |
| | 0.44 | 47 | 4 | 3 | 0 | - | $-2.72\times10^{-2}$ |
| 87 | 0 | 47 | 1 | 1 | 5 | - | $6.03\times10^{-2}$ |
| | 0.15 | 49 | 1 | 2 | 2 | - | $-1.57\times10^{-2}$ |
| 10 | 0 | 6 | 7 | 7 | 23 | 11 | $-2.77\times10^{-1}$ |
| | 0.15 | 1 | 8 | 9 | 23 | 13 | $-2.01\times10^{-1}$ |
| 15 | 0 | 8 | 7 | 7 | 21 | 11 | $-2.80\times10^{-1}$ |
| | 0.15 | 4 | 8 | 8 | 21 | 13 | $-2.17\times10^{-1}$ |
| 25 | 0 | 14 | 6 | 6 | 19 | 9 | $-2.56\times10^{-1}$ |
| | 0.15 | 11 | 7 | 7 | 19 | 10 | $-2.15\times10^{-1}$ |
| | 0.30 | 6 | 9 | 9 | 18 | 12 | $-1.97\times10^{-1}$ |
| 37 | 0 | 20 | 5 | 5 | 16 | 8 | $-2.22\times10^{-1}$ |
| | 0.15 | 18 | 6 | 6 | 15 | 9 | $-2.24\times10^{-1}$ |
| | 0.30 | 14 | 7 | 8 | 14 | 11 | $-1.95\times10^{-1}$ |
| | 0.44 | 9 | 9 | 9 | 13 | 14 | $-1.49\times10^{-1}$ |
| | 0.59 | 0 | 12 | 12 | 10 | 20 | $-9.24\times10^{-2}$ |
| 52 | 0 | 28 | 4 | 4 | 12 | 6 | $-1.39\times10^{-1}$ |
| | 0.15 | 27 | 5 | 5 | 11 | 6 | $-1.39\times10^{-1}$ |
| | 0.30 | 26 | 5 | 5 | 9 | 9 | $-1.23\times10^{-1}$ |
| | 0.44 | 23 | 7 | 7 | 5 | 12 | $-1.14\times10^{-1}$ |
| | 0.59 | 20 | 10 | 9 | 0 | 15 | $-7.31\times10^{-1}$ |
| 59 | 0 | 32 | 3 | 3 | 11 | 5 | $-8.09\times10^{-2}$ |
| | 0.15 | 32 | 4 | 3 | 9 | 6 | $-8.01\times10^{-2}$ |
| | 0.30 | 31 | 4 | 4 | 7 | 8 | $-6.32\times10^{-2}$ |
| | 0.44 | 31 | 5 | 5 | 4 | 9 | $-5.18\times10^{-2}$ |
| 72 | 0 | 39 | 2 | 2 | 7 | 4 | $1.487\times10^{-2}$ |
| | 0.15 | 40 | 2 | 2 | 5 | 5 | $4.37\times10^{-2}$ |
| | 0.30 | 41 | 3 | 3 | 1 | 6 | $1.98\times10^{-2}$ |
| 85 | 0 | 46 | 1 | 1 | 4 | 2 | $8.54\times10^{-3}$ |
| | 0.15 | 48 | 1 | 1 | 1 | 3 | $-3.53\times10^{-2}$ |