Supplementary materials

*Ab initio* construction of full phase diagram of MgO-CaO eutectic system using neural network interatomic potentials

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Training data for SCAN-NNP and the final model trained on PBE or SCAN functional are available at GitHub (https://github.com/mdil-pung/MgOCaO2022data).

![FIG. S1. Uncertainty of neural network potentials during semigrand ensemble simulations of (a) solid and (b) liquid phases. The committee models have 2 hidden layers and different numbers of hidden nodes, 30, 100, or 150, for example. Note that the uncertainty is < 5 meV/atom for all the trajectories spanning concentrations.](image)

![FIG. S2. Temperature-dependent lattice parameters ($a_0$) of rocksalt (s) and liquid (l) phases of MgO and CaO. Lattice parameters that are directly calculated from NPT simulations are presented as markers.](image)
and the analytical model \( a_0(T) = aT^2 + bT + c \) is fitted to them and shown as lines. Liquid lattice parameters are converted from the volume of rocksalt conventional unit cells.

FIG. S3. (a) Total and partial radial distribution functions \( (g(r)) \) and (b) total angular distribution functions \( (p(\theta)) \) of liquid MgO and CaO calculated within SCAN functional. The numbers in the parentheses indicate the number of atoms in the supercells. As can be seen from 100- and 12,500-atom results, SCAN-NNPs consistently reproduce the liquid structure regardless of supercell size.
### TABLE S1. Hyperparameters of atom-centered symmetry function. Cutoff radius is 7.0 and 4.5 Å for $G^2$ and $G^4$, respectively. Atom type corresponds to 1=Mg, 2=Ca, and 3=O.

| Type of neighbor atoms | $G^2$ | $\eta$ (Å$^{-3}$) | $R_s$ | $G^4$ | $\eta$ (Å$^{-3}$) | $\zeta$ | $\lambda$ |
|------------------------|-------|-------------------|-------|-------|-------------------|-------|-------|
| 1                      | 0.003214 | 0.000357 | 1 |-1 | 1.3 | 0.000357 | 1 |-1 | 2.3 | 0.000357 | 1 |-1 |
| 1                      | 0.357111 | 0.028569 | 1 |-1 | 1.3 | 0.028569 | 1 |-1 | 2.3 | 0.028569 | 1 |-1 |
| 1                      | 0.714211 | 0.089277 | 1 |-1 | 1.3 | 0.089277 | 1 |-1 | 2.3 | 0.089277 | 1 |-1 |
| 1                      | 1.249871 | 0.000357 | 2 |-1 | 1.3 | 0.000357 | 2 |-1 | 2.3 | 0.000357 | 2 |-1 |
| 2                      | 0.214264 | 0.028569 | 2 |-1 | 1.3 | 0.28569 | 2 |-1 | 2.3 | 0.28569 | 2 |-1 |
| 2                      | 0.537106 | 0.089277 | 2 |-1 | 1.3 | 0.089277 | 2 |-1 | 2.3 | 0.089277 | 2 |-1 |
| 1                      | 0.714213 | 0.000357 | 4 |-1 | 1.3 | 0.000357 | 4 |-1 | 2.3 | 0.000357 | 4 |-1 |
| 1                      | 1.248426 | 0.028569 | 4 |-1 | 1.3 | 0.28569 | 4 |-1 | 2.3 | 0.28569 | 4 |-1 |
| 2                      | 0.003214 | 0.089277 | 4 |-1 | 1.3 | 0.089277 | 4 |-1 | 2.3 | 0.089277 | 4 |-1 |
| 1                      | 0.357111 | 0.000357 | 1 | 1 | 1.3 | 0.000357 | 1 | 1 | 2.3 | 0.000357 | 1 | 1 |
| 2                      | 0.714211 | 0.028569 | 1 | 1 | 1.3 | 0.28569 | 1 | 1 | 2.3 | 0.28569 | 1 | 1 |
| 1                      | 1.249871 | 0.089277 | 1 | 1 | 1.3 | 0.089277 | 1 | 1 | 2.3 | 0.089277 | 1 | 1 |
| 2                      | 0.214264 | 0.000357 | 2 | 1 | 1.3 | 0.000357 | 2 | 1 | 2.3 | 0.000357 | 2 | 1 |
| 1                      | 0.537106 | 0.028569 | 2 | 1 | 1.3 | 0.28569 | 2 | 1 | 2.3 | 0.28569 | 2 | 1 |
| 2                      | 0.714213 | 0.089277 | 2 | 1 | 1.3 | 0.089277 | 2 | 1 | 2.3 | 0.089277 | 2 | 1 |
| 1                      | 1.248426 | 0.000357 | 4 | 1 | 1.3 | 0.000357 | 4 | 1 | 2.3 | 0.000357 | 4 | 1 |
| 2                      | 0.003214 | 0.028569 | 4 | 1 | 1.3 | 0.28569 | 4 | 1 | 2.3 | 0.28569 | 4 | 1 |

### TABLE S2. Data set statistics. The same protocol for constructing data sets is used for both PBE-NNP and SCAN-NNP.

| Type               | X (Mg, Ca, 1−O) | Temperature (K) | MD time (ps) | Sampling interval (fs) | Number of structure | Number of atoms |
|--------------------|-----------------|----------------|---------------|------------------------|---------------------|-----------------|
| Strained crystal   | 0, 1            | 0              | -             | -                      | 162                 | 1,296           |
| Random alloys      | 0.08, 0.2, 0.8, 0.92 | 0              | -             | -                      | 8                   | 800             |
| Melting pure rock-salt (NVT, NPT) | 0, 1 | 300, 2000, 4000 | 1 | 40 | 300 | 30,000 |
| Melting random alloy (NVT, NPT) | 0.08, 0.2, 0.8, 0.92 | 300, 2000, 4000 | 1 | 40 | 1,200 | 120,000 |
| Total              |                 |                 |               |                        | 5,670               | 552,096         |