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).

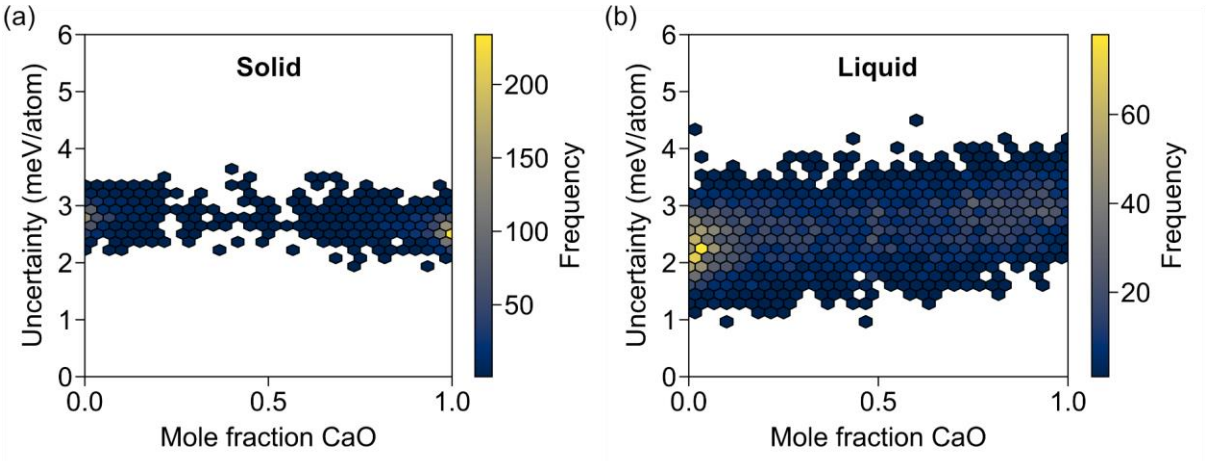

FIG. S1. Uncertainty of neural network potentials during semigrand ensemble simulations of (a) solid and (b) liquid phases. The committee models have 2 hidden layers and different numbers of hidden nodes, 30, 100, or 150, for example. Note that the uncertainty is < 5 meV/atom for all the trajectories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

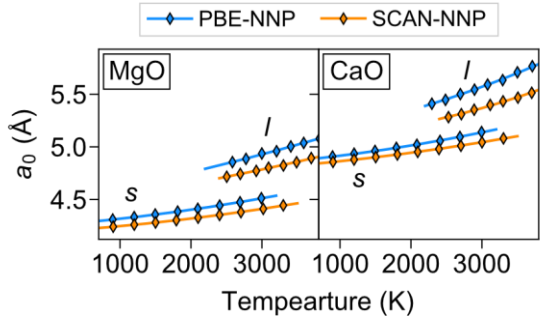

FIG. S2. Temperature-dependent lattice parameters (a_0) of rocksalt (*s*) and liquid (*l*) phases of MgO and CaO. Lattice parameters that are directly calculated from NPT simulations are presented as markers,

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.

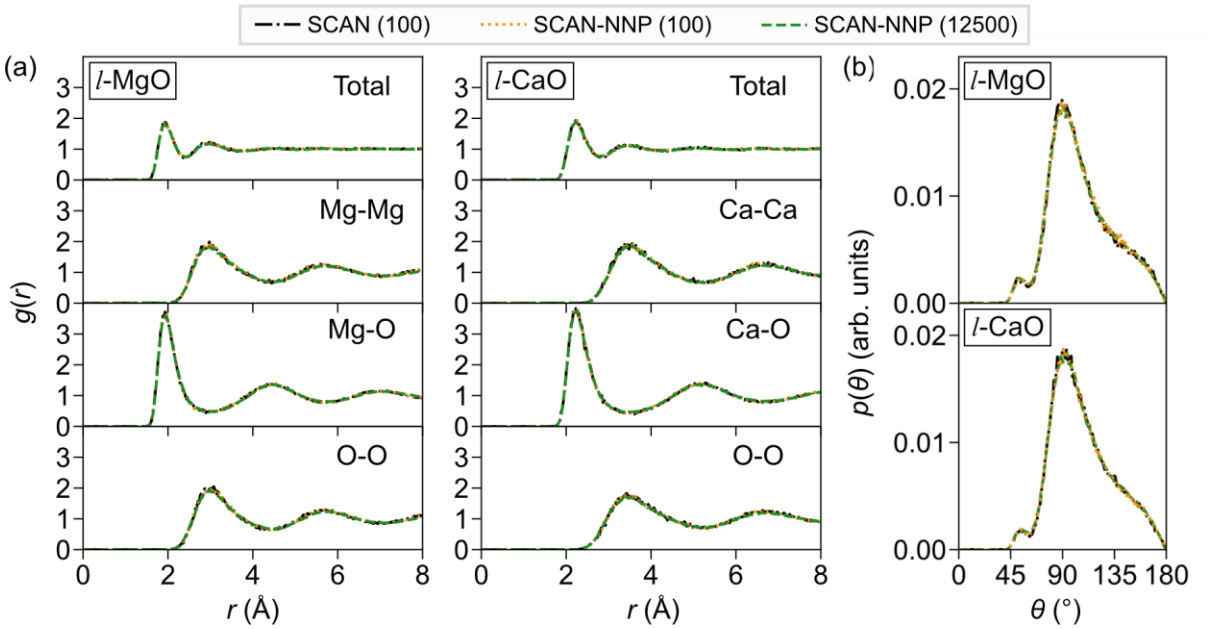

FIG. S3. (a) Total and partial radial distribution functions ($g(r)$) and (b) total angular distribution functions ($p(\theta)$) of liquid MgO and CaO calculated within SCAN functional. The numbers in the parentheses indicate the number of atoms in the supercells. As can be seen from 100- and 12,500-atom 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.

G^2			G^4														
Type of neighbor atoms	η (Å^{-2})	R_s	Type of neighbor atoms		η (Å^{-2})	ζ	λ	Type of neighbor atoms		η (Å^{-2})	ζ	λ	Type of neighbor atoms		η (Å^{-2})	ζ	λ
1	0.003214	0	1	1	0.000357	1	-1	1	3	0.000357	1	-1	2	3	0.000357	1	-1
1	0.035711	0	1	1	0.028569	1	-1	1	3	0.028569	1	-1	2	3	0.028569	1	-1
1	0.071421	0	1	1	0.089277	1	-1	1	3	0.089277	1	-1	2	3	0.089277	1	-1
1	0.124987	0	1	1	0.000357	2	-1	1	3	0.000357	2	-1	2	3	0.000357	2	-1
1	0.214264	0	1	1	0.028569	2	-1	1	3	0.028569	2	-1	2	3	0.028569	2	-1
1	0.357106	0	1	1	0.089277	2	-1	1	3	0.089277	2	-1	2	3	0.089277	2	-1
1	0.714213	0	1	1	0.000357	4	-1	1	3	0.000357	4	-1	2	3	0.000357	4	-1
1	1.428426	0	1	1	0.028569	4	-1	1	3	0.028569	4	-1	2	3	0.028569	4	-1
2	0.003214	0	1	1	0.089277	4	-1	1	3	0.089277	4	-1	2	3	0.089277	4	-1
2	0.035711	0	1	1	0.000357	1	1	1	3	0.000357	1	1	2	3	0.000357	1	1
2	0.071421	0	1	1	0.028569	1	1	1	3	0.028569	1	1	2	3	0.028569	1	1
2	0.124987	0	1	1	0.089277	1	1	1	3	0.089277	1	1	2	3	0.089277	1	1
2	0.214264	0	1	1	0.000357	2	1	1	3	0.000357	2	1	2	3	0.000357	2	1
2	0.357106	0	1	1	0.028569	2	1	1	3	0.028569	2	1	2	3	0.028569	2	1
2	0.714213	0	1	1	0.089277	2	1	1	3	0.089277	2	1	2	3	0.089277	2	1
2	1.428426	0	1	1	0.000357	4	1	1	3	0.000357	4	1	2	3	0.000357	4	1
3	0.003214	0	1	1	0.028569	4	1	1	3	0.028569	4	1	2	3	0.028569	4	1
3	0.035711	0	1	1	0.089277	4	1	1	3	0.089277	4	1	2	3	0.089277	4	1
3	0.071421	0	1	2	0.000357	1	-1	2	2	0.000357	1	-1	3	3	0.000357	1	-1
3	0.124987	0	1	2	0.028569	1	-1	2	2	0.028569	1	-1	3	3	0.028569	1	-1
3	0.214264	0	1	2	0.089277	1	-1	2	2	0.089277	1	-1	3	3	0.089277	1	-1
3	0.357106	0	1	2	0.000357	2	-1	2	2	0.000357	2	-1	3	3	0.000357	2	-1
3	0.714213	0	1	2	0.028569	2	-1	2	2	0.028569	2	-1	3	3	0.028569	2	-1
3	1.428426	0	1	2	0.089277	2	-1	2	2	0.089277	2	-1	3	3	0.089277	2	-1
			1	2	0.000357	4	-1	2	2	0.000357	4	-1	3	3	0.000357	4	-1
			1	2	0.028569	4	-1	2	2	0.028569	4	-1	3	3	0.028569	4	-1
			1	2	0.089277	4	-1	2	2	0.089277	4	-1	3	3	0.089277	4	-1
			1	2	0.000357	1	1	2	2	0.000357	1	1	3	3	0.000357	1	1
			1	2	0.028569	1	1	2	2	0.028569	1	1	3	3	0.028569	1	1
			1	2	0.089277	1	1	2	2	0.089277	1	1	3	3	0.089277	1	1
			1	2	0.000357	2	1	2	2	0.000357	2	1	3	3	0.000357	2	1
			1	2	0.028569	2	1	2	2	0.028569	2	1	3	3	0.028569	2	1
			1	2	0.089277	2	1	2	2	0.089277	2	1	3	3	0.089277	2	1
			1	2	0.000357	4	1	2	2	0.000357	4	1	3	3	0.000357	4	1
			1	2	0.028569	4	1	2	2	0.028569	4	1	3	3	0.028569	4	1
			1	2	0.089277	4	1	2	2	0.089277	4	1	3	3	0.089277	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 ($Mg_xCa_{1-x}O$)	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
		6000, 8000	1	10	800	80,000
Melting random alloy (NVT, NPT)	0.08, 0.2, 0.8, 0.92	300, 2000, 4000	1	40	1,200	120,000
		6000, 8000	1	10	3,200	320,000
Total					5,670	552,096