Supporting Information

Accurate prediction of grain boundary structures and energetics in CdTe: A machine-learning potential approach

Tatsuya Yokoi,[a]* Kosuke Adachi[a], Sayuri Iwase[a], Katsuyuki Matsunaga[a,b]

[a]*Department of Materials Physics, Nagoya University, Nagoya 464-8603, Japan*
[b]*Nanostructures Research Laboratory, Japan Fine Ceramics Center, Nagoya, 456-8587, Japan*

Determination of hyperparameters for ANN potential

Tables S1 and S2 summarize the hyperparameters used for two- and three-body symmetry functions, respectively. These values and the total number of symmetry functions were empirically determined so as to distinguish different atomic environments, as well as different elements, with sufficient accuracy.

To find an appropriate cutoff radius for the symmetry functions (R_c), five artificial-neural-network (ANN) potentials were trained using five R_c values, 6, 7, 8, 9 and 10 Å. In this test calculation, the $66 \times 33 \times 33 \times 1$ architecture was used. Single-crystal structures were used as references for a training dataset. This dataset also included vacancies. To cover a wide range of atomic environments, various initial structures were generated by randomly changing their atomic positions and cell dimensions of reference structures. Using the generated structures, total energies and atomic forces were obtained by performing DFT single-point calculations and DFT structural relaxations. Totally, 7990 energies and 112800 atomic forces were used to train the ANN potentials. Table S3 list the MAE values for these ANN potentials. It is found that for $R_c = 7$ Å, the ANN potential shows the lowest MAE values of 2.16 meV/atom and 20.73 meV/Å for the total energies and atomic forces, respectively, MAE values are also found to increase with increasing R_c. Thus R_c was finally chosen to be 7 Å.

In addition, an appropriate number of nodes was also examined by constructing three ANN potentials: $66 \times 44 \times 44 \times 1$, $66 \times 55 \times 55 \times 1$ and $66 \times 66 \times 66 \times 1$ architectures. In this test calculation, $R_c = 7$ Å was used. As a training dataset, single-crystal, surfaces ((001), (110), (111), (112) and (310) planes), $\Sigma 3(112)$ GBs were used as reference structures. Their initial structures were generated in the same way as that mentioned above. In this case, single-point calculations and structural relaxations were performed using DFT calculations to obtain total energies and atomic forces. Totally, 10750 energies and 522810 atomic forces were used to train the ANN potentials. Table S4 summarizes the MAE values for the three ANN potential. The $66 \times 66 \times 66 \times 1$ architecture is found to provide the lowest value of 4.10 meV/atom and 39.43 meV/Å for the total energies and atomic forces, respectively. More nodes in the hidden layers may decrease MAE values in the training dataset, but the difference between the MAEs of the 66

× 55 × 55 × 1 and 66 × 66 × 66 × 1 architectures is small. Thus considering the trade-off between the accuracy and computational cost, the 66 × 66 × 66 × 1 architecture was finally used in the main calculation.

Table S1. Hyperparameters (η and R_s) of two-body symmetry functions, G_i^2 (i = 1, 2, ..., 18).

No.	Two-body pair	η	R_s
1		0.0010	0.000
2		0.0050	0.200
3	Cd-Cd	0.0100	0.500
4		0.0500	1.000
5		0.1000	1.400
6		0.1500	1.800
7		0.0030	0.100
8		0.0070	0.300
9	Te-Te	0.0300	0.700
10		0.0700	1.300
11		0.1300	1.700
12		0.1700	2.100
13		0.0020	0.050
14		0.0060	0.250
15	Cd-Te	0.0200	0.600
16		0.0600	1.150
17		0.1150	1.550
18		0.1600	1.950

Table S2. Hyperparameters (η, φ and ζ) for three-body symmetry functions, G_i^3 (i = 19, 20, ..., 66).

No.	Three-body pair	η	φ	ζ
19		0.0400	0.000	1.000
20		0.0600	0.000	1.500
21	Gd-Gd-Gd	0.0800	0.000	1.000
22		0.1200	0.000	1.500
23		0.1600	0.000	1.000
24		0.2000	0.000	1.500

25		0.0400	180.000	1.000
26		0.0600	180.000	1.500
27		0.0800	180.000	1.000
28		0.1200	180.000	1.500
29		0.1600	180.000	1.000
30		0.2000	180.000	1.500
31		0.0500	30.000	1.300
32		0.0700	30.000	1.700
33		0.0900	30.000	1.300
34		0.1500	30.000	1.700
35		0.1900	30.000	1.300
36	Te-Te-Te	0.2300	30.000	1.700
37		0.0500	120.000	1.300
38		0.0700	120.000	1.700
39		0.0900	120.000	1.300
40		0.1500	120.000	1.700
41		0.1900	120.000	1.300
42		0.2300	120.000	1.700
43		0.0466	20.000	1.200
44		0.0666	20.000	1.633
45		0.0866	20.000	1.200
46		0.1400	20.000	1.633
47		0.1800	20.000	1.200
48	Cd-Te-Te	0.2200	20.000	1.633
49		0.0466	140.000	1.200
50		0.0666	140.000	1.633
51		0.0866	140.000	1.200
52		0.1400	140.000	1.633
53		0.1800	140.000	1.200
54		0.2200	140.000	1.633
55		0.0433	10.000	1.100
56		0.0633	10.000	1.566
57	Cd-Cd-Te	0.0833	10.000	1.100
58		0.1300	10.000	1.566
59		0.1700	10.000	1.100
60		0.2100	10.000	1.566

61	0.0433	160.000	1.100
62	0.0633	160.000	1.566
63	0.0833	160.000	1.100
64	0.1300	160.000	1.566
65	0.1700	160.000	1.100
66	0.2100	160.000	1.566

Table S3. MAE values of the total energies and atomic forces for R_c values of 6, 7, 8, 9 and 10 Å.

R_c [Å]	MAE	
	Energy [meV/atom]	Atomic force [meV/Å]
6	2.37	23.63
7	2.16	20.73
8	2.21	23.24
9	2.51	26.67
10	2.92	30.76

Table S4. MAE values of the total energies and atomic forces for the 66 × 55 × 55 × 1 and 66 × 66 × 66 × 1 architectures.

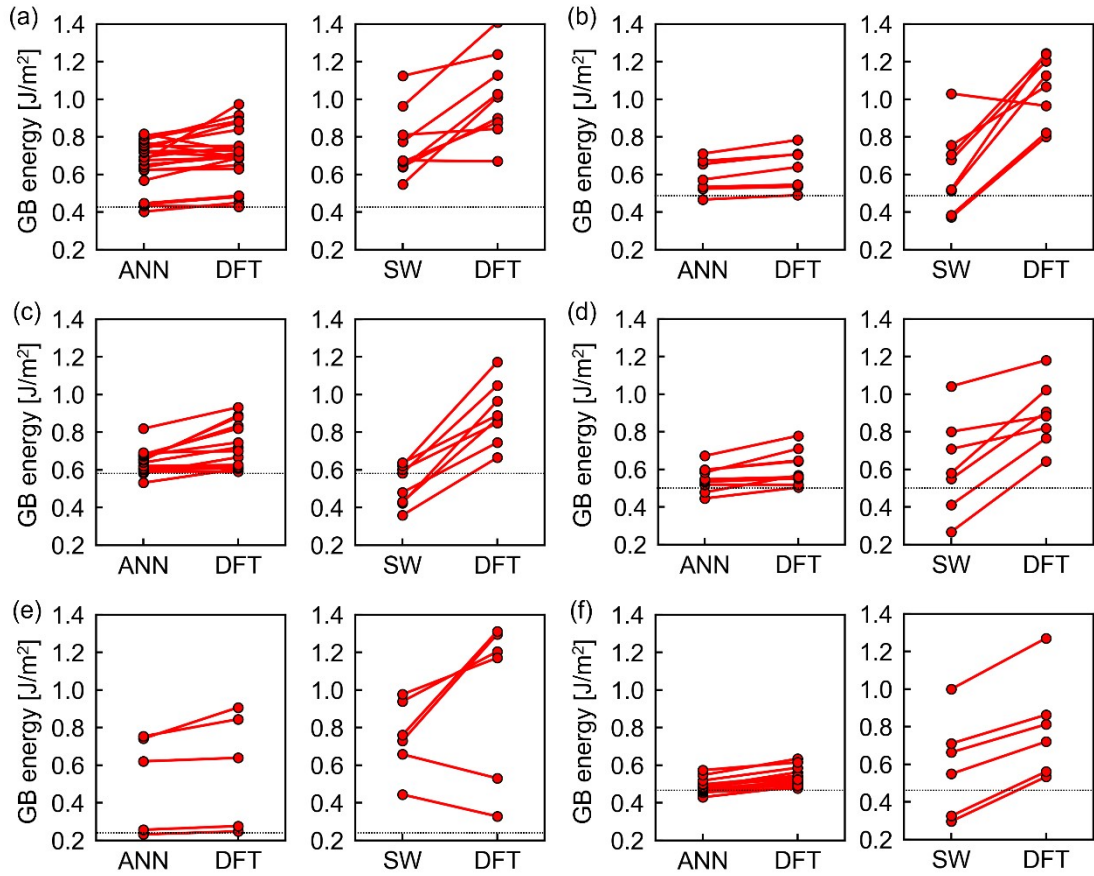| Architecture	MAE	
	Energy [meV/atom]	Atomic force [meV/Å]
66 × 44 × 44 × 1	4.39	41.17
66 × 55 × 55 × 1	4.30	40.34
66 × 66 × 66 × 1	4.10	39.43

Figure S1. Comparison of GB energies of the ANN and SW potentials with DFT values for (a) the $\Sigma3(112)/[112]$, (b) $\Sigma5(210)/[001]$, (c) $\Sigma5(310)/[001]$, (d) $\Sigma9(221)/[221]$, (e) $\Sigma11(113)/[113]$, (f) $\Sigma11(332)/[332]$ GBs. The dashed line indicates the lowest DFT GB energy of the GB structure predicted by the ANN potential. Note that for the $\Sigma3(111)/[111]$ GB, the lowest-energy structure shows a GB energy of ~0.01 J/m^2 for both the ANN and SW potential. Additionally, high-energy structures are absent in both cases. Thus their results are not shown in this figure.