Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary materials

Revealing the crystallization dynamics of Sb-Te phase change materials by large-scale simulations

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1. The energy-volume curves and melting points (T_m) in elementary Sb and Te

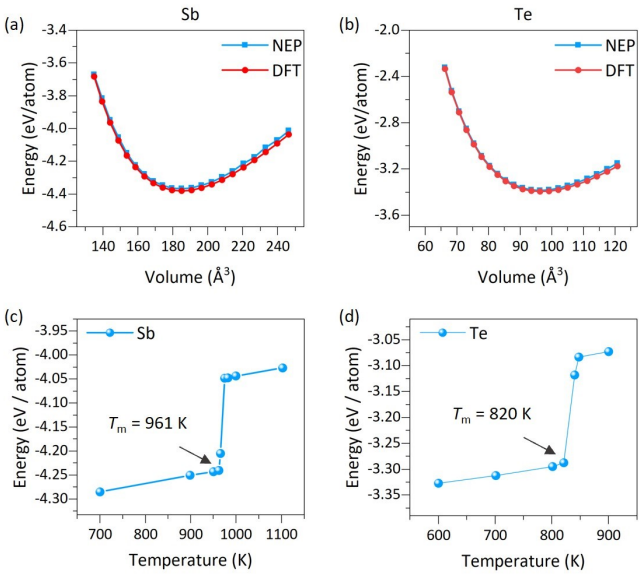

Fig. S1. **(a)** and **(b)** The energy-volume (E-V) curves for elementary Sb and Te. **(c)** and **(d)** The calculated T_m for elementary Sb and Te by solid-liquid coexistence method[1].

2. The energy-volume curves and melting points (T_m) in Sb_4Te_3, Sb_8Te_3 and $Sb_{16}Te_3$

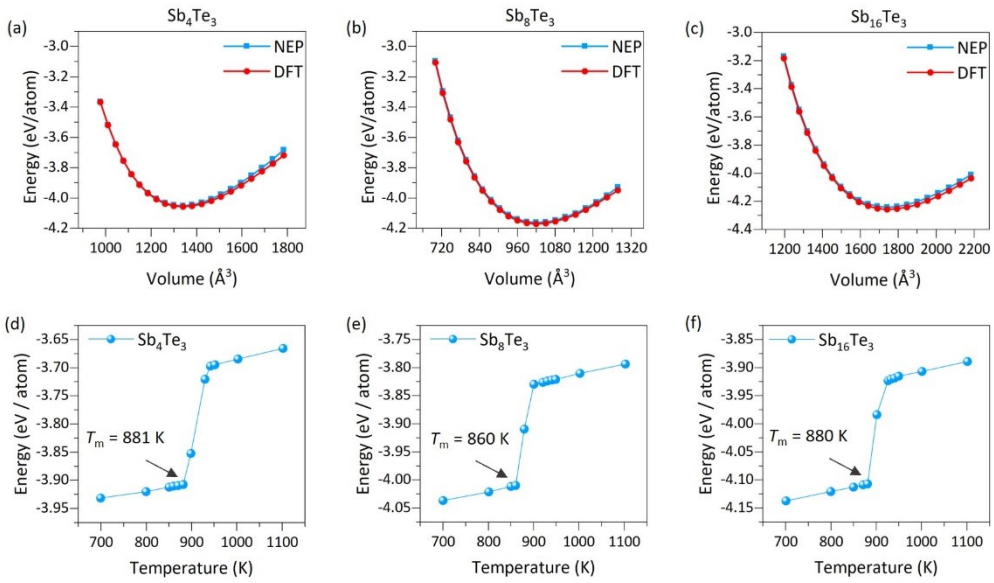

Fig. S2. **(a-c)** The energy-volume (E-V) curve for Sb_4Te_3, Sb_8Te_3 and $Sb_{16}Te_3$ compositions. **(d-f)** The calculated melting point T_m for Sb_4Te_3, Sb_8Te_3 and $Sb_{16}Te_3$ compositions.

The corresponding lattice parameters fitted from energy-volume curves are listed in Table S1.

Table S1. Equilibrium volume and lattice parameters of Sb_4Te_3, Sb_8Te_3 and $Sb_{16}Te_3$. The DFT results are calculated at the PBE-D3(BJ) functional.

Component	Space Group	DFT Prediction		NEP Calculation	
		Volume (Å^3)	Lattice parameters (Å)	Volume (Å^3)	Lattice parameters (Å)
Sb_4Te_3	*R-3m*	1339.94	a = 4.341, c = 82.109	1334.56	a = 4.362, c = 80.993
Sb_8Te_3	*R-3m*	1023.98	a = 4.350, c = 62.484	1023.343	a = 4.398, c = 61.086
$Sb_{16}Te_3$	*R-3m*	1750.64	a = 4.350, c = 106.810	1744.47	a = 4.394, c = 104.325

3. The radial distribution function (RDF) of elementary Sb and Te

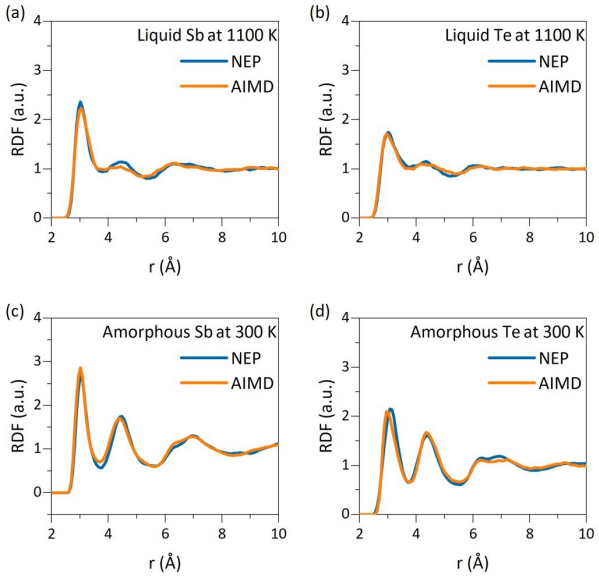

Fig. S3. The radial distribution function (RDF) of **(a)** liquid Sb, **(b)** liquid Te, **(c)** amorphous Sb and **(d)** amorphous Te.

4. The partial RDF of amorphous Sb_2Te_3 phase

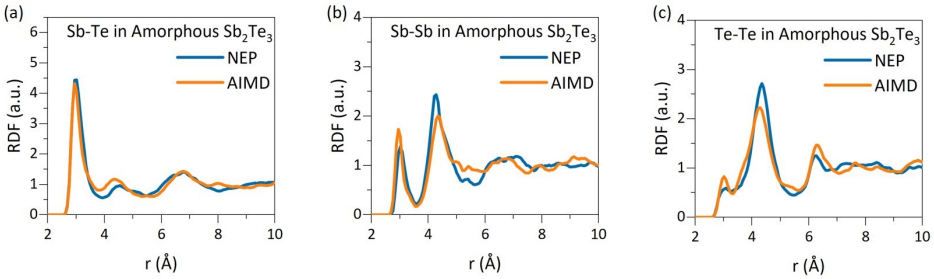

Fig. S4. The partial RDF of amorphous Sb_2Te_3 for **(a)** Sb-Te, **(b)** Sb-Sb and **(c)** Te-Te pairs.

5. The RDF of large size supercells in Sb_2Te and Sb_2Te_2 components

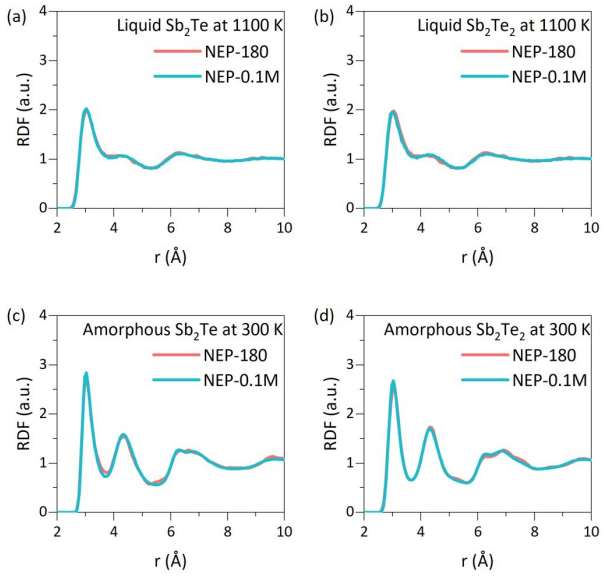

Fig. S5. The RDF under different supercell sizes for **(a)** liquid Sb_2Te, **(b)** liquid Sb_2Te_2, **(c)** amorphous Sb_2Te and **(d)** amorphous Sb_2Te_2.

6. The effect of quenching-rate in generating Sb_2Te_3 amrophous phase

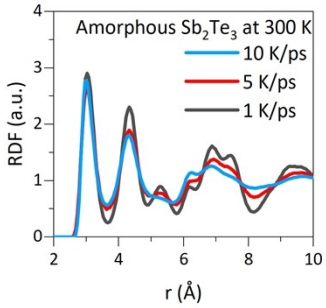

Fig. S6. The RDF of Sb_2Te_3 amorphous phase under different quenchin-rate of 1 K/ps, 5 K/ps and 10 K/ps.

7. The RDF of Sb_2Te, Sb_2Te_2 and Sb_2Te_3 compounds after a 2 ns crystallization simulations at different temperatures

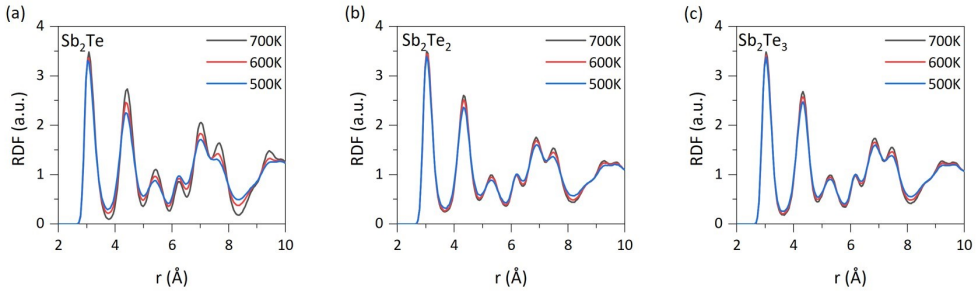

Fig. S7. **(a-c)** The RDF after crystallization process at different temperatures for Sb_2Te, Sb_2Te_2 and Sb_2Te_3.

8. The analysis on amorphous phase before crystallization using Polyhedral Template Matching

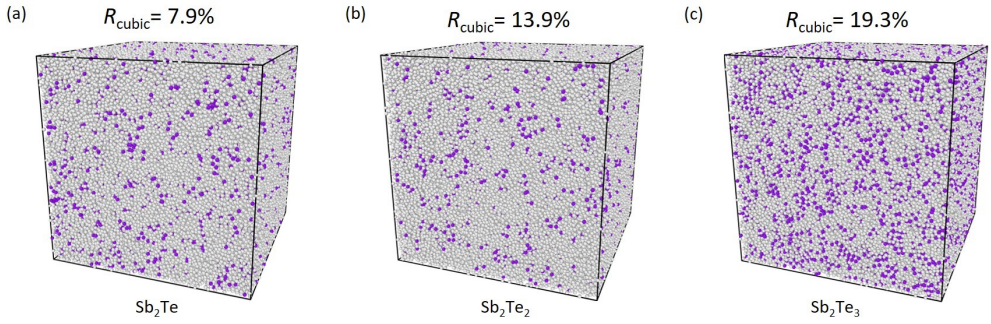

Fig. S8. **(a-c)** The snapshots of initial amorphous Sb_2Te, Sb_2Te_2 and Sb_2Te_3 compounds before crystallization. The atoms possessed a cubic-type order are colored by purple and the disorder atoms are colored by light gray. The atom types are identified by the polyhedral template matching method[2] and the RMSE cutoff is 0.15. These models are rendered using OVITO software[3]. R_{cubic} indicates the ratio of cubic-type atoms.

9. The construction of dataset for fitting NEP potential

Table S2. Summary of the dataset for training NEP potential.

		Cells in training set	**Cells in testing set**
Isolated atom	Sb / Te	2	0
Dimer	Sb-Sb / Sb-Te / Te-Te	105	0
Crystalline phase	Sb / Te / Sb_2Te / SbTe / Sb_2Te_3	695	231
Defect structure	Sb_2Te / SbTe / Sb_2Te_3	81	0
Molten phase in 3000 K	Sb / Te / Sb_2Te / SbTe / Sb_2Te_3	78	23
Liquid phase in 2000 K and 1000 K	Sb / Te / Sb_2Te / SbTe / Sb_2Te_3	205	153
Amorphous phase	Sb / Te / Sb_2Te / SbTe / Sb_2Te_3	403	245
Perturbed structure	Sb / Te / Sb_2Te / SbTe / Sb_2Te_3	137	10
Total		1706	662

References

1. J. Zhu, Q. Wang and H. Wang, *Acta Metall. Sin.*, 2017, **53**, 1018-1024.
2. P. M. Larsen, S. Schmidt and J. Schiøtz, *Model. Simul. Mat. Sci. Eng.*, 2016, **24**, 055007.
3. A. Stukowski, *Model. Simul. Mat. Sci. Eng.*, 2009, **18**, 015012.