Supplementary information: First principles thermodynamical modeling of the binodal and spinodal curves in lead chalcogenides

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Received Xth XXXXXXXXXX 20XX, Accepted Xth XXXXXXXXX 20XX
First published on the web Xth XXXXXXXXXX 200X
DOI: 10.1039/b000000x

SI Convergence tests

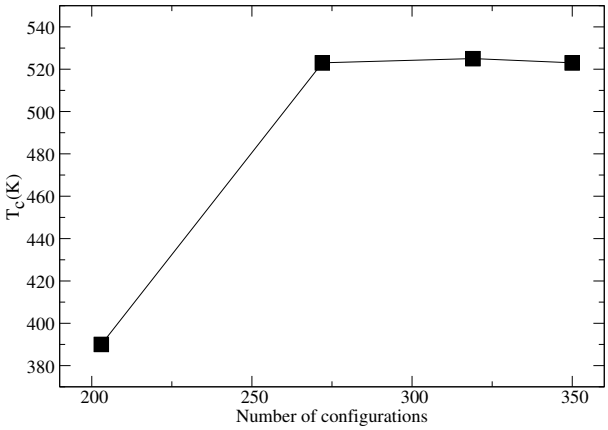

Figure S1 Convergence test for consolute temperature using as variable the number of total explored configurations (DFT+CE) and considering only HSS for the calculation of the interaction parameter.

SII CE influence

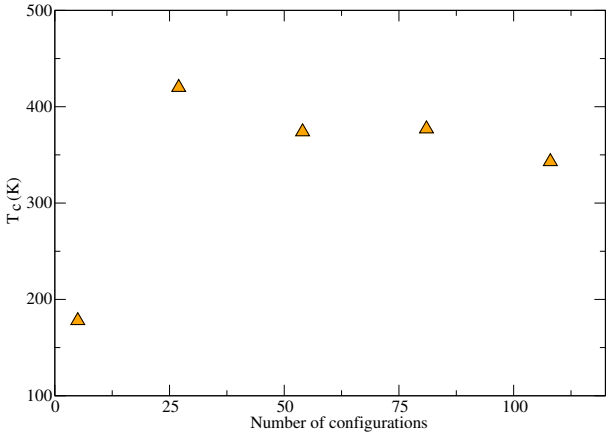

Figure S2 Consolute temperature using as variable the number of DFT energies and considering an average of the whole set of points to obtain the interaction parameter.

SIII Fitting parameters

	L_0(eV/atom)	L_1(eV/atom)	L_2(eV/atom)
$PbSe_{1-x}Te_x$	1.167×10^{-1}	-9.365×10^{-2}	7.162×10^{-2}
$PbS_{1-x}Te_x$	3.118×10^{-1}	-2.600×10^{-1}	1.701×10^{-1}
$PbS_{1-x}Se_x$	4.251×10^{-2}	-2.744×10^{-2}	1.515×10^{-2}

Table S1 Fitting parameters obtained from formation enthalpy values of highly symmetric structures (HSS) to evaluate the interaction parameter of lead chalcogenide pseudobinary systems

SIV Interaction parameter

References

1 H. Liu and L. L. Y. Chang, *Mineral. Mag.*, 1994, **58**, 567–578.
2 N. Boukhris, H. Meradji, S. Ghemid, S. Drablia and F. E. H. Hassan, *Phys. Scripta*, 2011, **83**, 065701–065710.

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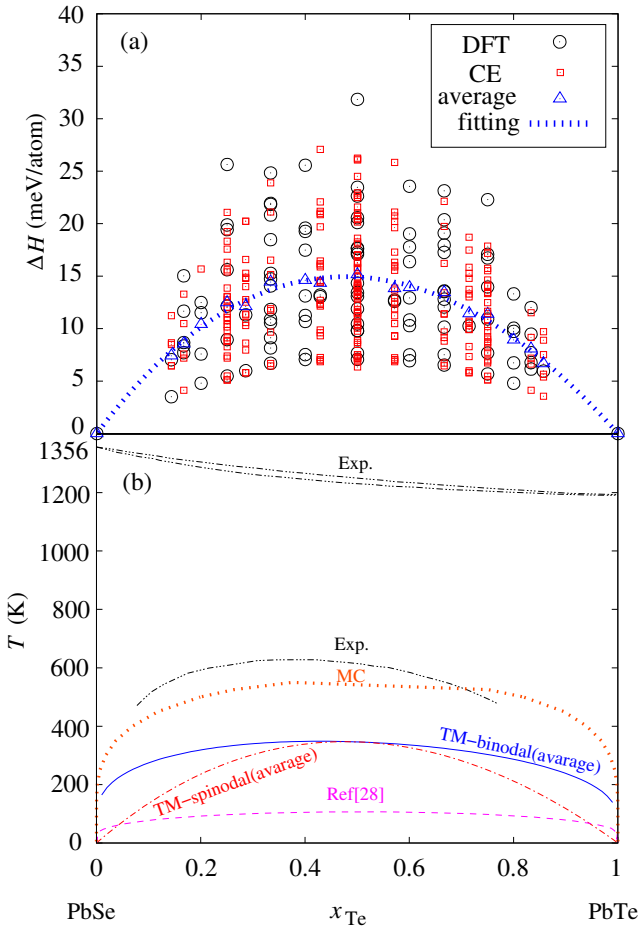

Figure S3 (a) Formation enthalpies of the PbSe-PbTe structures using DFT calculations (◯) and CE technique (□). Average points are represented by △ and the fitting using whole set of points (DFT+CE) to obtain the interaction parameter is plotted with a blue dashed line. (b) Binodal and spinodal curves from TM using average (— and –·–), and MC simulations (···), compared with experimental data [1] (–···–) and the previous theoretical model (- - -) from Boukhris *et al.* [2]