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

Supporting information – First-principles study on the chemical stability of inorganic perovskite solid solutions $Cs_{1-x}Rb_xPbI_3$ at finite temperature and pressure

Un-Gi Jong[a,b], Chol-Jun Yu[a*], Yun-Hyok Kye[a], Yun-Sim Kim[a], Chol-Ho Kim[b], Son-Guk Ri[c]

[a]*Chair of Computational Materials Design, Faculty of Materials Science, Kim Il Sung University, Ryongnam-Dong, Taesong District, Pyongyang, Democratic People's Republic of Korea*

[b]*Natural Science Centre, Kim Il Sung University, Ryongnam-Dong, Taesong District, Pyongyang, Democratic People's Republic of Korea*

[c]*Chair of Semiconductor Engineering, Faculty of Materials Science, Kim Il Sung University, Ryongnam-Dong, Taesong District, Pyongyang, Democratic People's Republic of Korea*

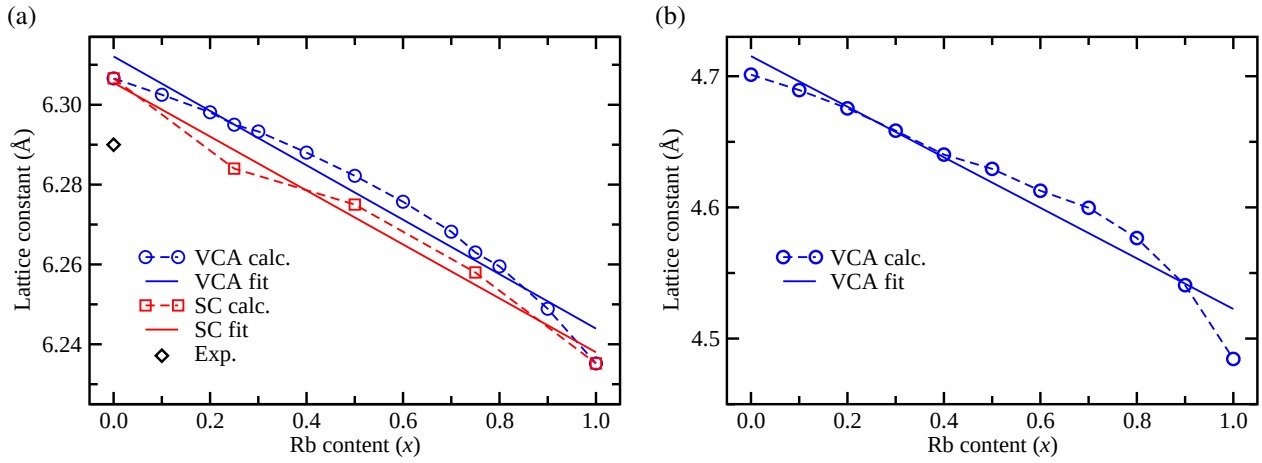

Figure S1. The lattice constants calculated by virtual crystal approximation (VCA) and supercell (SC) methods with their linear fitting lines as a function of Rb content x for (a) $Cs_{1-x}Rb_xPbI_3$ and (b) $Cs_{1-x}Rb_xI$.

Table S1. Atomic charge of atoms in $CsPbI_3$ and $RbPbI_3$, calculated by using the Hirshfeld method. C_R, C_H, and C_N are reference, Hirshfield, and net charges, respectively. The sum of atomic charges of iodine atoms is also presented. Positive value indicates the loss of electron, while negative value means the gain of electron.

Atom	C_R	$CsPbI_3$		$RbPbI_3$	
		C_H	C_N	C_H	C_N
Pb	14.000	13.705	0.295	13.717	0.283
I	7.000	7.181	-0.181	7.170	-0.170
	7.000	7.181	-0.181	7.170	-0.170
	7.000	7.181	-0.181	7.170	-0.170
I_3			-0.543		-0.510
Cs (Rb)	1.000	0.752	0.248	0.772	0.228

*Corresponding author: Chol-Jun Yu, Email: cj.yu@ryongnamsan.edu.kp

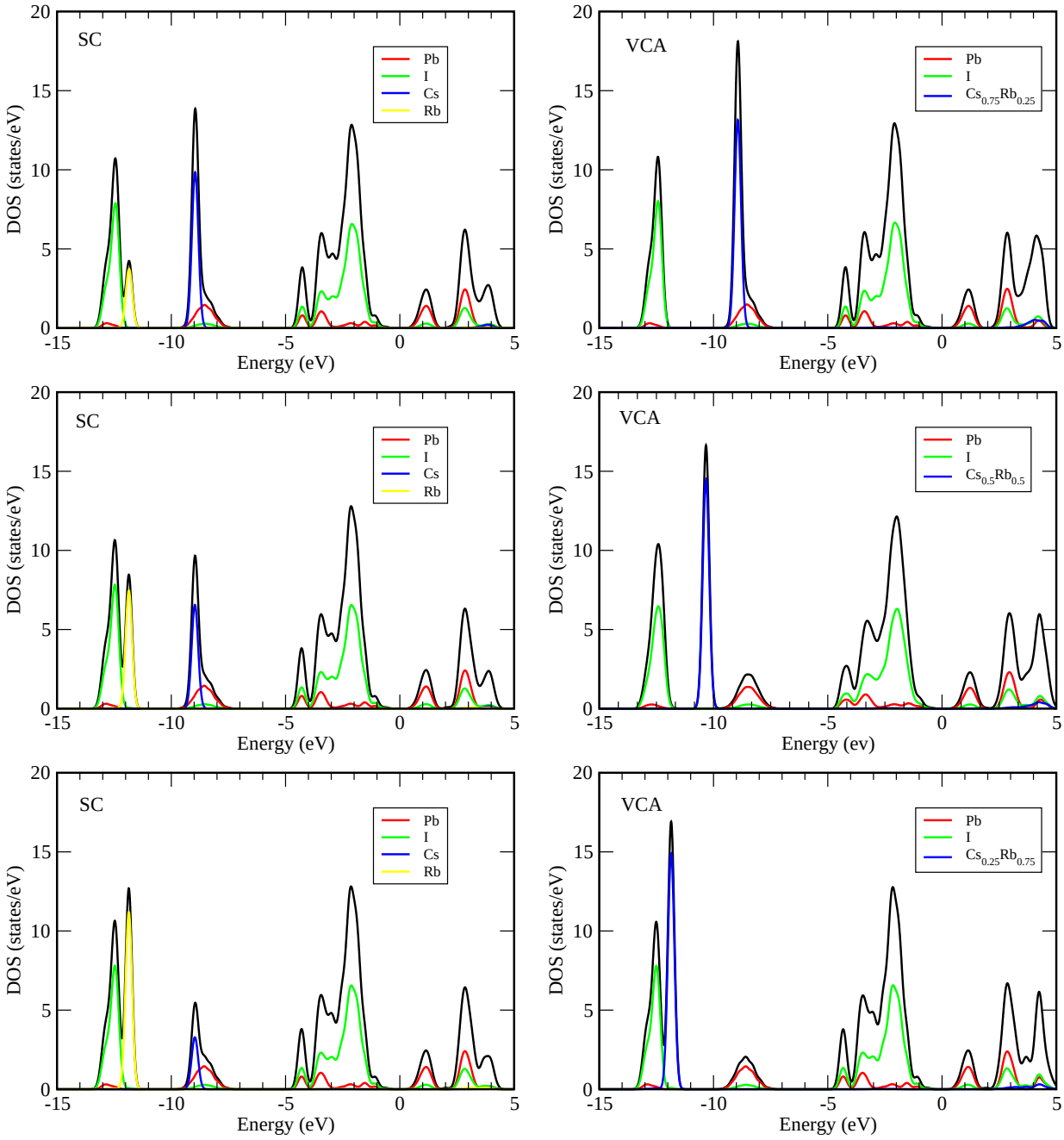

Figure S2. Density of states for $Cs_{1-x}Rb_xPbI_3$ calculated by SC (left panel) and VCA (right panel) methods. To coincide the scale of DOS, the values of DOS calculated by SC method are multiplied by 0.25 due to the use of (2×2×1) supercell.

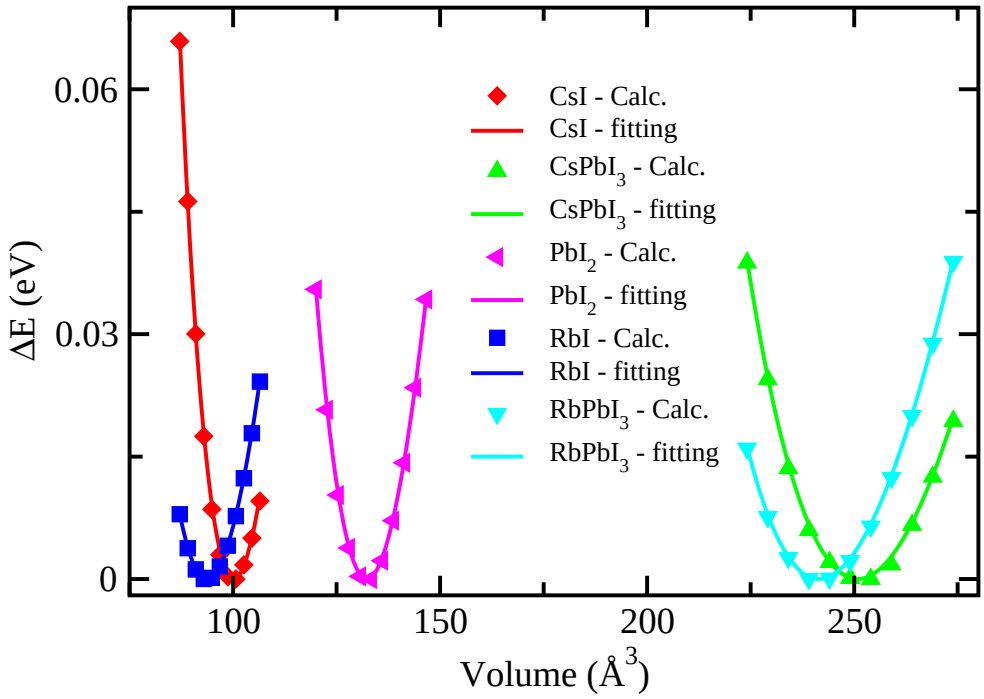

Figure S3. $E - V$ curves obtained by fitting into the equation of state for Cs(Rb)PbI$_3$, PbI$_2$, and Cs(Rb)I.

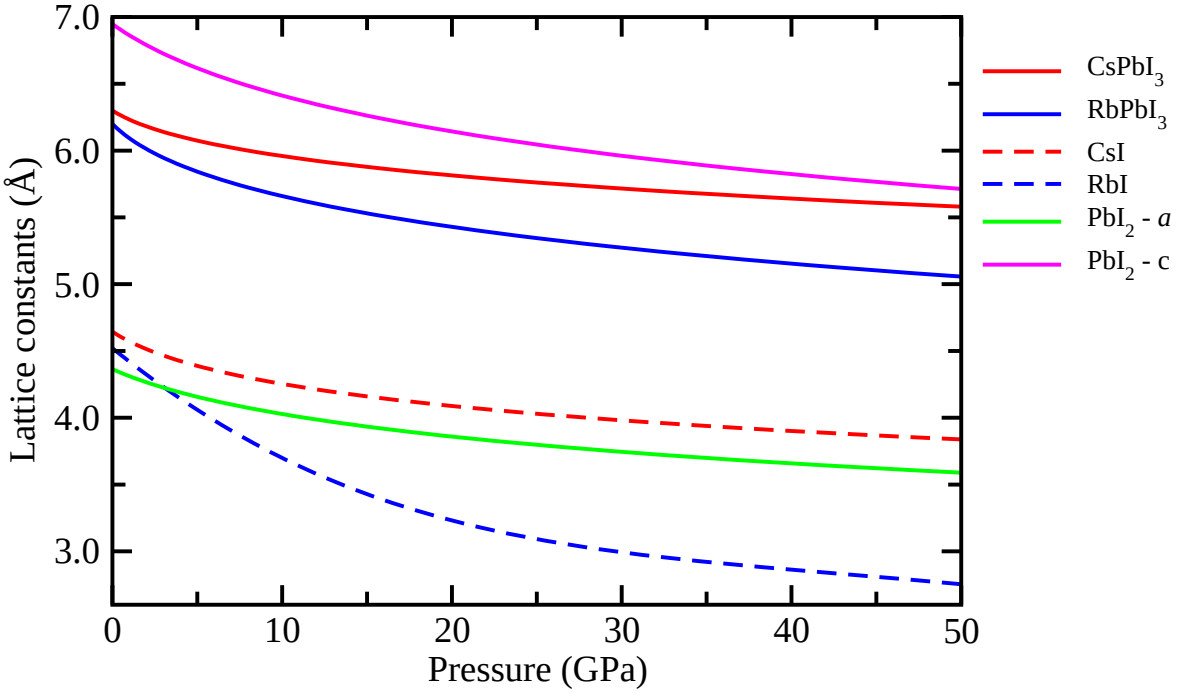

Figure S4. Lattice constant vs. pressure curves for Cs(Rb)PbI$_3$, PbI$_2$, and Cs(Rb)I.

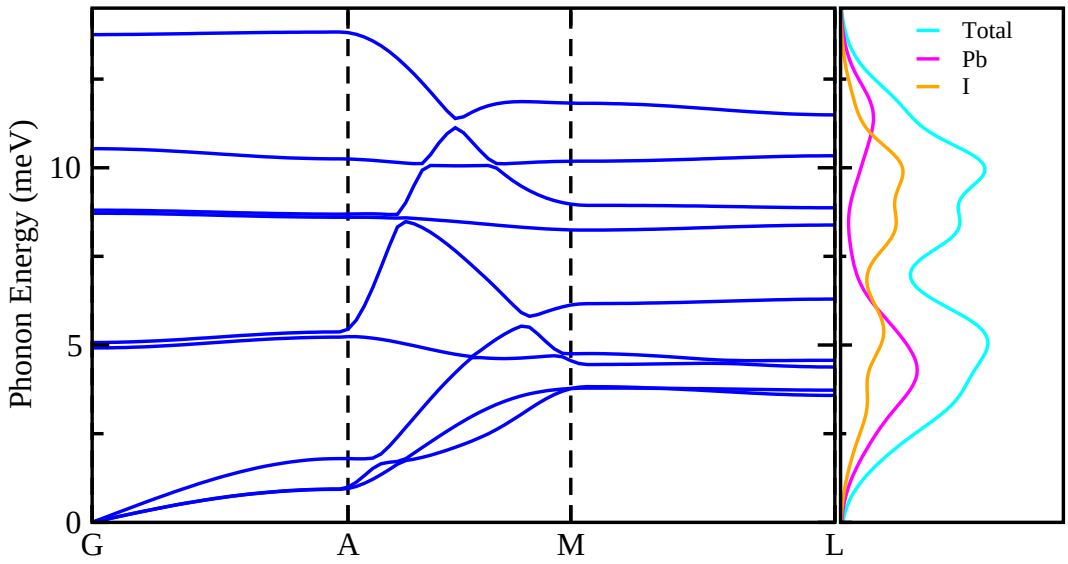

Figure S5. Phonon dispersion curve and DOS of PbI_2.

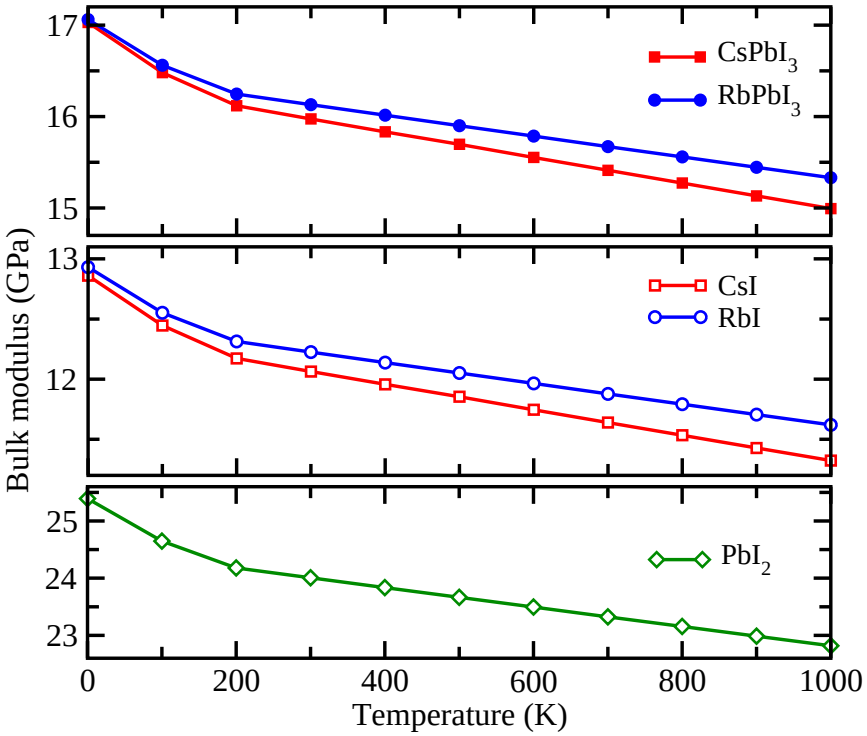

Figure S6. Bulk modulus of $CsPbI_3$, $RbPbI_3$, CsI, RbI and PbI_2 as the temperature is rising to 1000 K.