Supporting Information

High-throughput Computational Design of Organic-inorganic Hybrid Halide Semiconductors Beyond Perovskites for Optoelectronics

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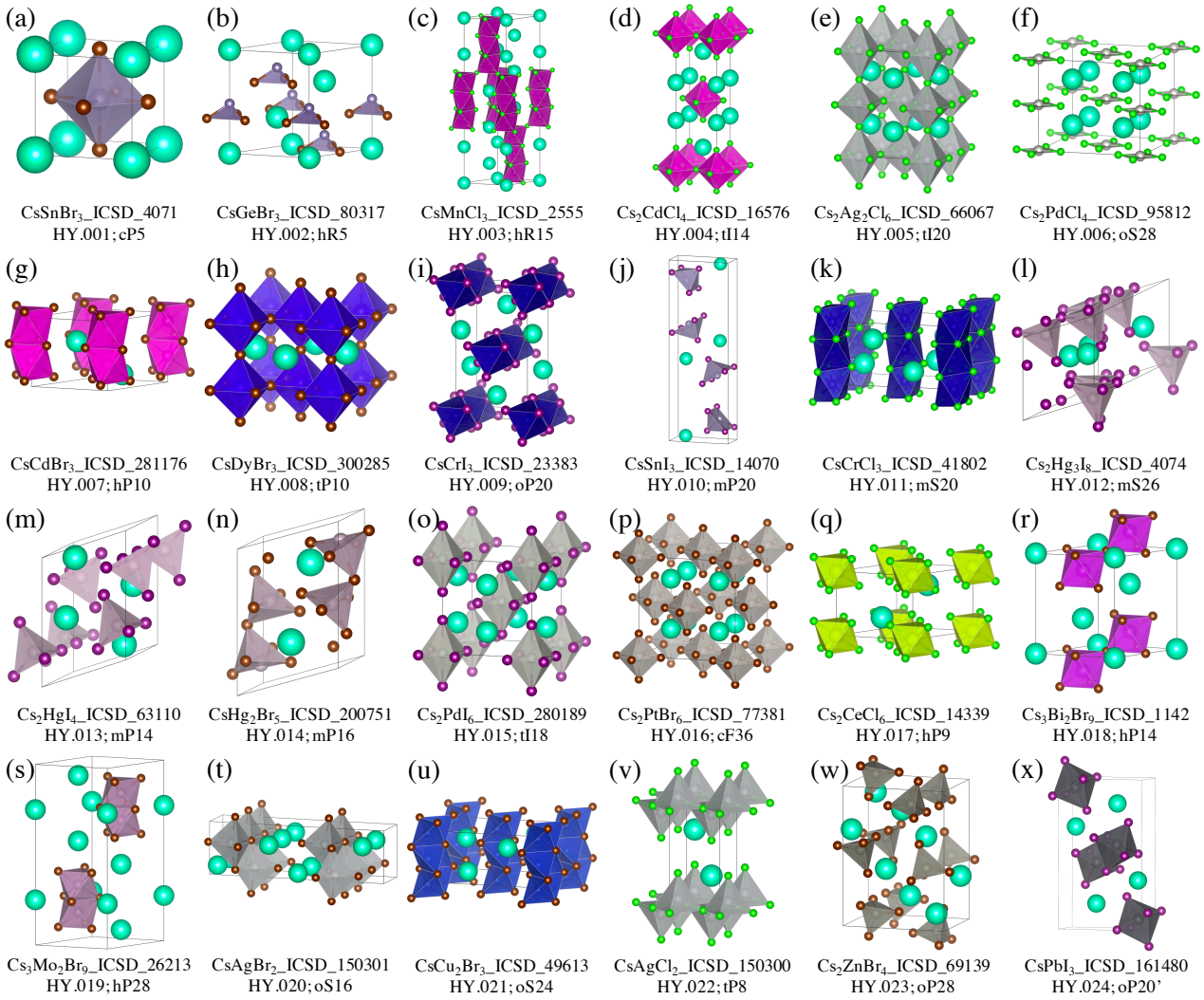
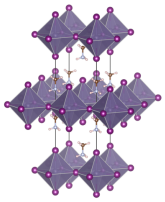
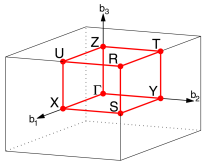
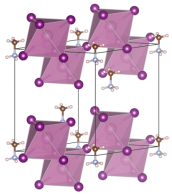
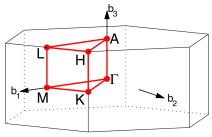
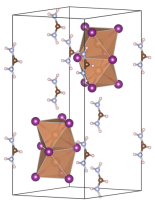
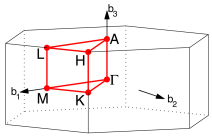
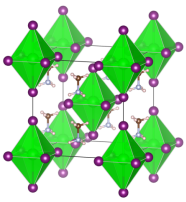
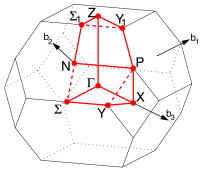


FIG. S1: Structures of the 24 perovskite-derived inorganic metal halide compounds ($Cs_xB_yX_z$) used as structural prototypes (HY.001-HY.024) to build the hybrid metal halide compound repository. The conventional-cell structures are shown in polyhedral style from the standard orientation of crystal shape using VESTA.[1] ICSD number and Pearson symbol of each inorganic compound are given. Note that $CsCrI_3$_ICSD_23383 and $CsPbI_3$_ICSD_161480 share the same Pearson symbol of oP20, and to distinguish them, we denote the former with oP20, and the later with oP20'.

TABLE S1: Calculated properties for the reference compound $MASnBr_3$ (cP5) using different functionals: lattice parameters (in Å), electron (hole) effective masses m_e^* (m_h^*) (in m_0) near the CBM (VBM), reduced effective mass (μ) (in m_0), and decomposition enthalpy (ΔH_d, in eV/f.u.). m_0 refers to the free electron rest mass. The experimentally measured lattice parameters are a = 5.837 Å and c = 5.853 Å.[2]

Functional	Lattice Parameters			m_e^*	m_h^*	μ	ΔH_d
	a	b	c				
PBE	5.9538	5.9144	6.0039	0.31	0.18	0.11	0.26
vdW.D3	5.8446	5.8208	5.8533	0.32	0.12	0.09	0.22
optB86b	5.8289	5.8059	5.8473	0.32	0.14	0.10	0.21

TABLE S2: Calculated electron and hole effective masses along major directions for the anisotropic tI14, hP14, hP28, and tI18 candidates. † denotes effective masses calculated with spin-orbit coupling.

Prototype Structure	Brillouin Zone	Candidates	Γ - Z		Γ - X	
			m_e^*	m_h^*	m_e^*	m_h^*
 tI14		$(MA)_2GeBr_4$	0.89	2.04	0.11	0.23
		$(MA)_2GeI_4$	0.80	38.73	0.10	0.23
		$(MA)_2SnCl_4$	3.37	3.20	0.14	0.23
		$(MA)_2SnBr_4$	4.36	2.29	0.10	0.16
		$(MA)_2SnI_4$	4.12	8.38	0.09	0.15
		$(FA)_2SnBr_4$	2.41	1.76	0.12	0.18
		$(AD)_2GeI_4$	0.82	1.72	0.10	0.56
		$(AD)_2SnBr_4$	1.07	1.89	0.13	0.26
		$(AD)_2SnI_4$	1.17	1.19	0.10	0.22
Prototype Structure	Brillouin Zone	Candidates	Γ - A		M - K	
			m_e^*	m_h^*	m_e^*	m_h^*
 hP14		$(MA)_3In_2I_9$	0.46	12.74	0.99	0.61
		$(MA)_3Sb_2Br_9$	0.33	0.41	0.52	2.66
		$(MA)_3Sb_2I_9$	0.38	0.31	0.33	0.31
		$(MA)_3Bi_2I_9$	0.41†	1.21†	0.63†	0.82†
		$(FA)_3Ga_2I_9$	2.37	50.87	0.76	1.17
		$(FA)_3In_2Br_9$	0.57	6.17	2.02	0.23
		$(FA)_3In_2I_9$	0.52	9.03	1.68	0.57
		$(FA)_3Bi_2I_9$	1.06†	10.46†	0.26†	0.75†
		$(AD)_3Sb_2Br_9$	0.50	0.43	0.65	1.65
		$(AD)_3Sb_2I_9$	0.36	0.31	0.40	2.94
		$(AD)_3Bi_2I_9$	0.51	0.66	0.46	1.26
Prototype Structure	Brillouin Zone	Candidates	Γ - A		M - K	
			m_e^*	m_h^*	m_e^*	m_h^*
 hP28		$(FA)_3Sb_2I_9$	2.00	6.96	0.39	1.90
		$(AD)_3In_2I_9$	0.92	1.63	1.27	1.40
Prototype Structure	Brillouin Zone	Candidates	Γ - Z		Γ - X	
			m_e^*	m_h^*	m_e^*	m_h^*
 tI18		$(MA)_2ZrI_6$	0.80	1.59	8.46	0.98
		$(AD)_2HfI_6$	3.20†	0.80†	2.54†	1.73†
		$(AD)_2SnBr_6$	0.51	1.24	0.52	1.76
		$(AD)_2TeBr_6$	1.69	2.17	2.15	1.03

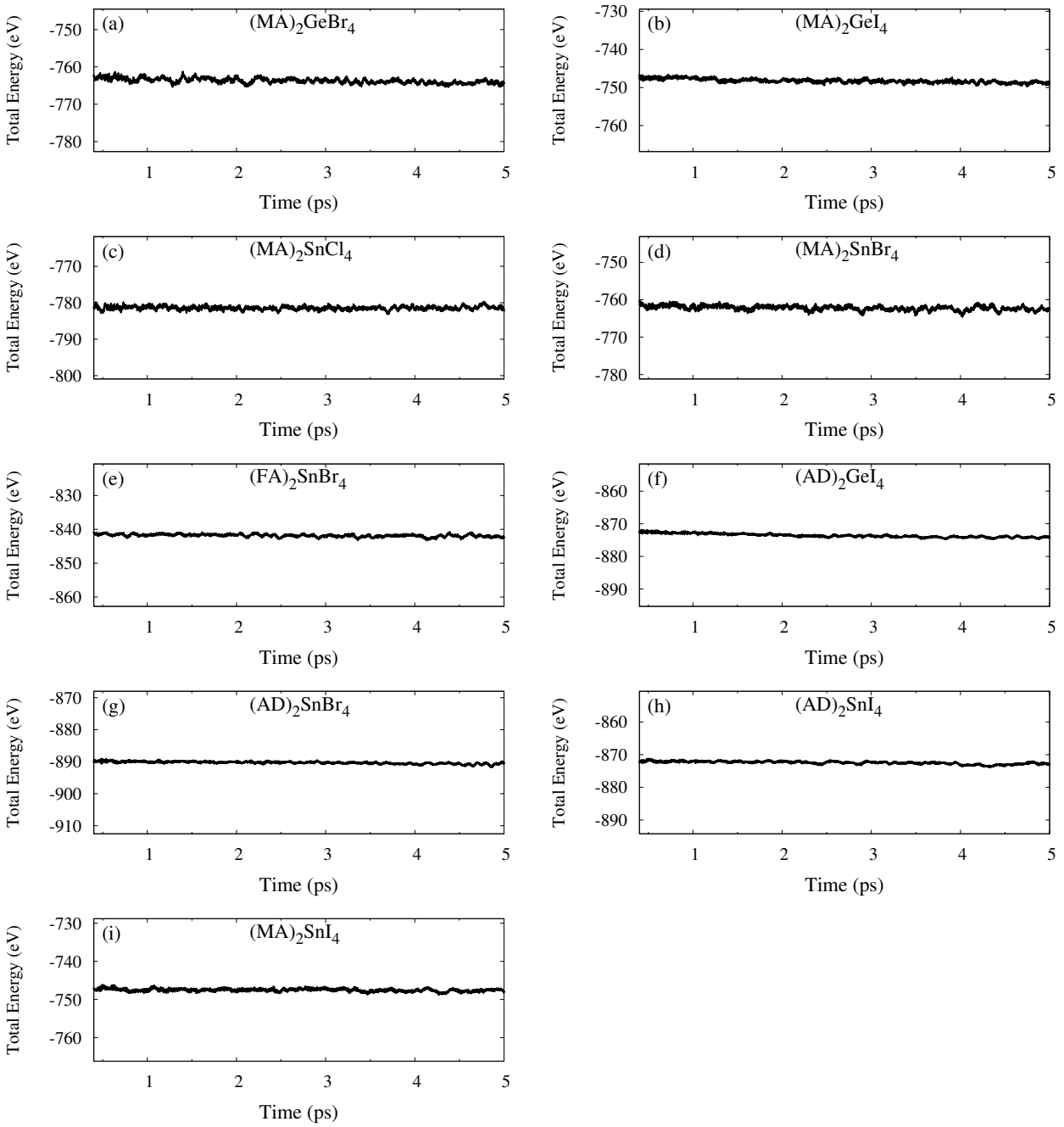


FIG. S2: Total energy during 5ps *ab-initio* molecular dynamics (AIMD) simulations at 300K for all the screened "tI14" compounds. In this and subsequent figures, the Pearson symbols of analogue inorganic compounds including tI14, hP14, tI18, cF36, and hP28 are used to distinguish the prototypes of the screened hybrid inorganic-organic materials.

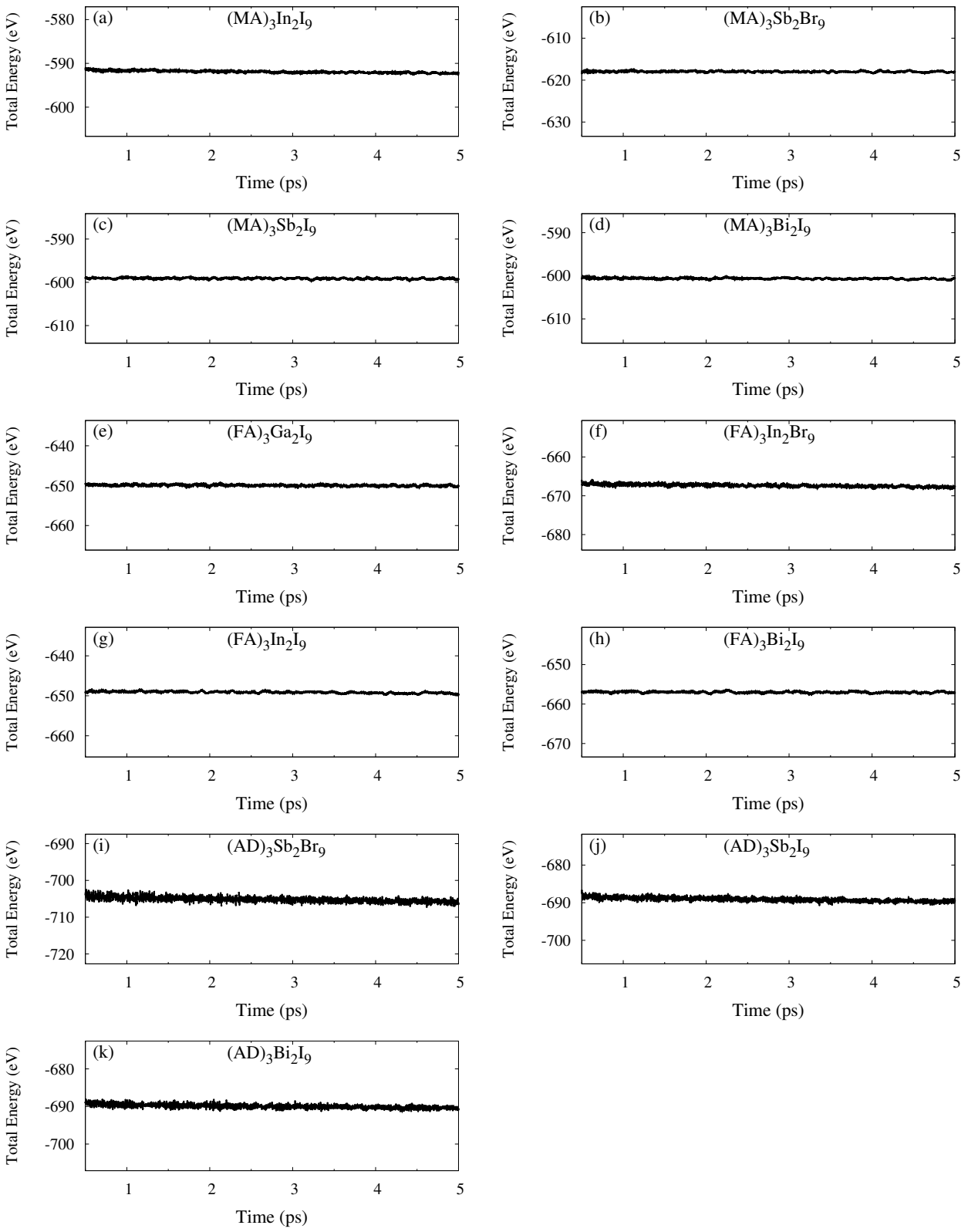


FIG. S3: Total energy during 5ps *ab-initio* molecular dynamics (AIMD) simulations at 300K for all the screened "hP14" compounds.

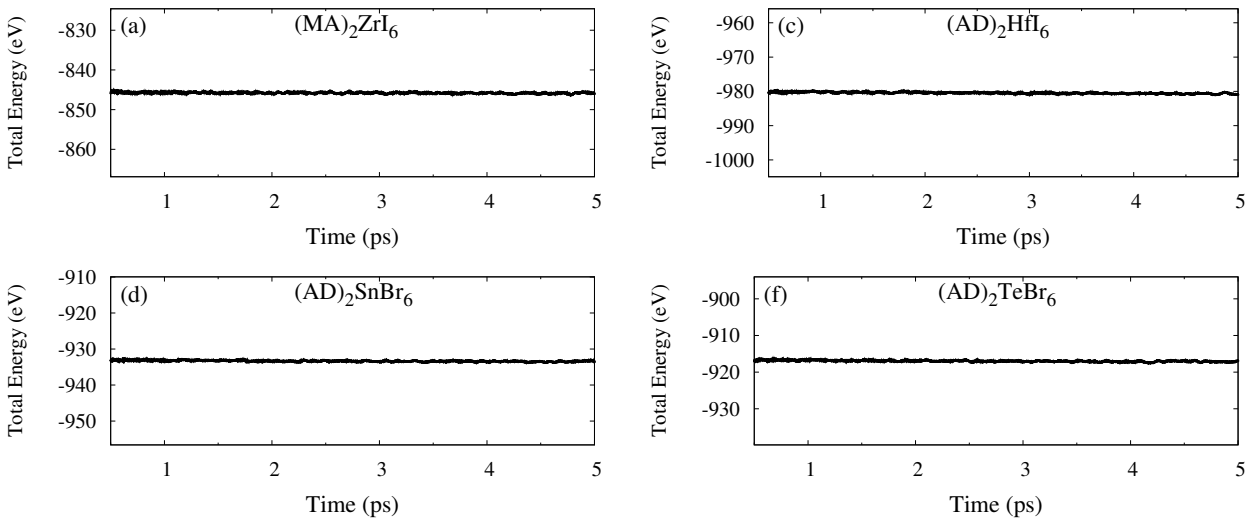


FIG. S4: Total energy during 5ps *ab-initio* molecular dynamics (AIMD) simulations at 300K for all the screened "tI18" compounds.

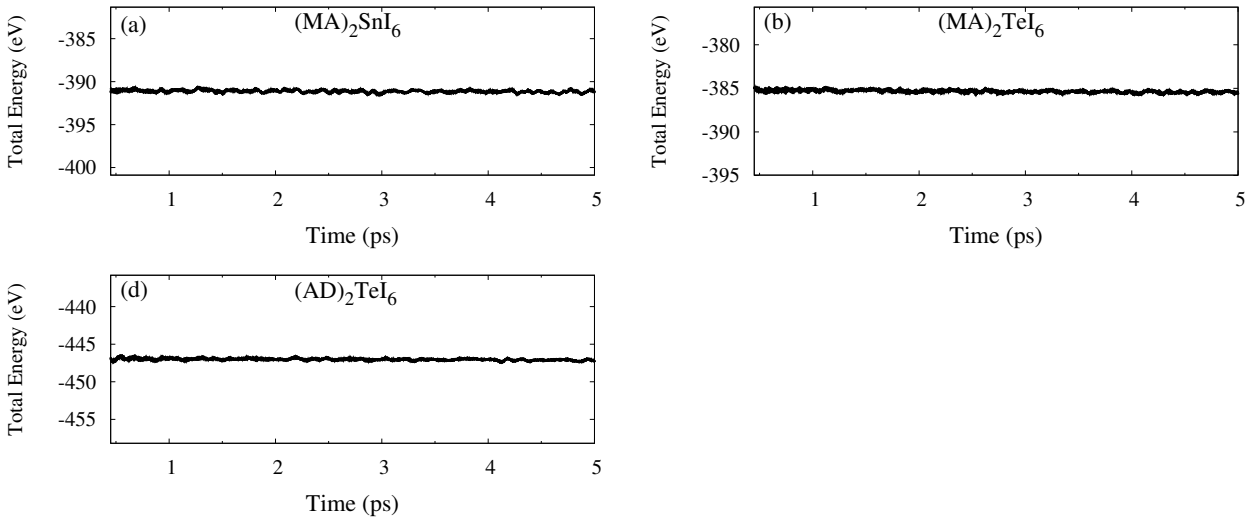


FIG. S5: Total energy during 5ps *ab-initio* molecular dynamics (AIMD) simulations at 300K for all the screened "cF36" compounds.

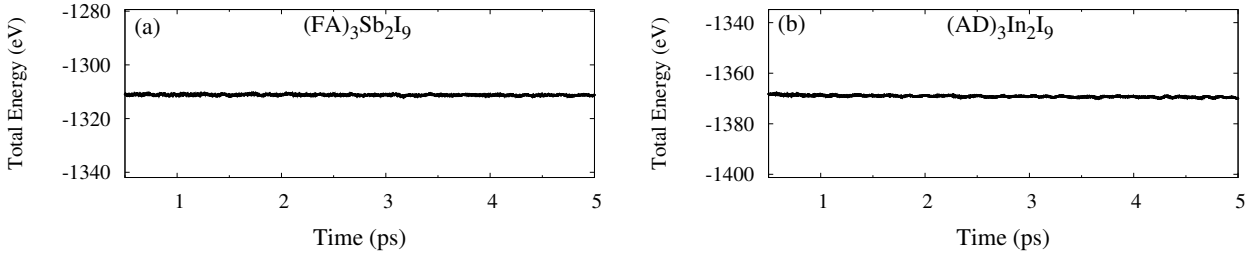


FIG. S6: Total energy during 5ps *ab-initio* molecular dynamics (AIMD) simulations at 300K for all the screened "hP28" compounds.

[1] K. Momma and F. Izumi, *VESTA3 for Three-Dimensional Visualization of Crystal, Volumetric and Morphology Data*, J. Appl. Crystallogr. **44**, 1272–1276 (2011).
[2] F. Hao, C. C. Stoumpos, D. H. Cao, R. P. Chang, and M. G. Kanatzidis, *Lead-Free Solid-State Organic–Inorganic Halide Perovskite Solar Cells*, Nat. Photon. **8**, 489 (2014).