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Structural and electronic properties of multifunctional carbon composites of organometal halide perovskite

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Table S1. Structural and energetic properties of composites.

	Graphene/ $MAPbI_3$	GO/ $MAPbI_3$	C_{60}/ $MAPbI_3$	$C_{60}(OH)_{24}$/ $MAPbI_3$
E_{ads} [eV]	– 0.980	– 0.806	– 0.860	– 2.136
d_{eq} [Å]	3.77	2.51	($d_{Pb–(C–C)}$) 3.00	(d_{Pb-O}) 2.77

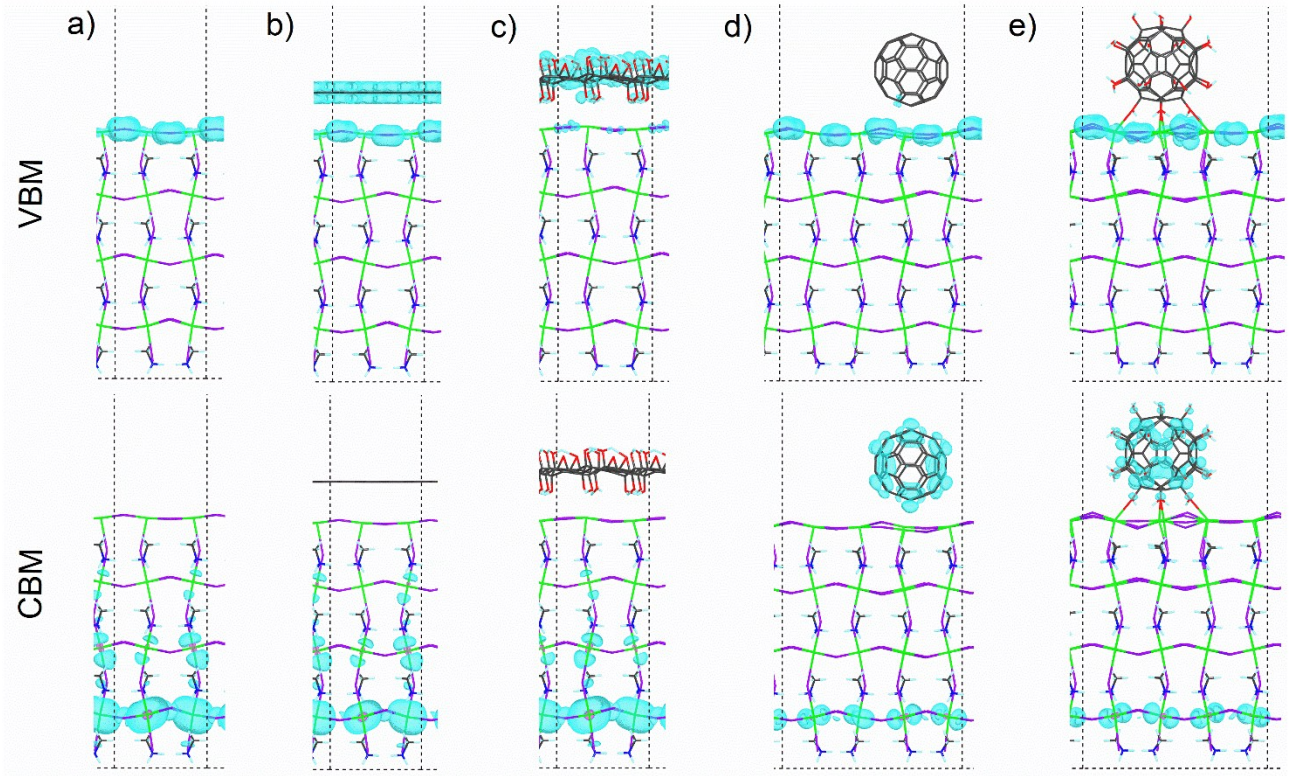
Adsorption energies (E_{ads}) and equivalent distance (d_{eq}) computed with PBE-D3 for PbI_2-rich $MAPbI_3$ (001) surface modified with graphene, graphene oxide, C_{60} and $C_{60}(OH)_{24}$.

Table S2. Density of states analysis.

	Bulk $MAPbI_3$	Slab $MAPbI_3$	Graphene	GO	C_{60}	$C_{60}(OH)_{24}$
E_g^{pure} [eV] PBE-D3/HSE06-D3	1.74/2.27	0.60/1.08 (1×1) 0.62/1.10 (2×2)	0.00/0.00	2.28/3.83	1.64/2.25	2.30/3.83
$E_g^{composite}$ [eV] PBE-D3/HSE06-D3	–	–	0.00/0.00	0.92/1.45	0.66/1.14	0.84/1.34
VBO [eV] PBE-D3	–	–	– 1.01	– 0.10	0.34	0.72

DFT band gap energies computed for pure components (E_g^{pure}) and composite structures ($E_g^{composite}$) according to PBE and HSE06 methods. Valance band offset (VBO) determined from DOS analysis according to PBE-D3 method.

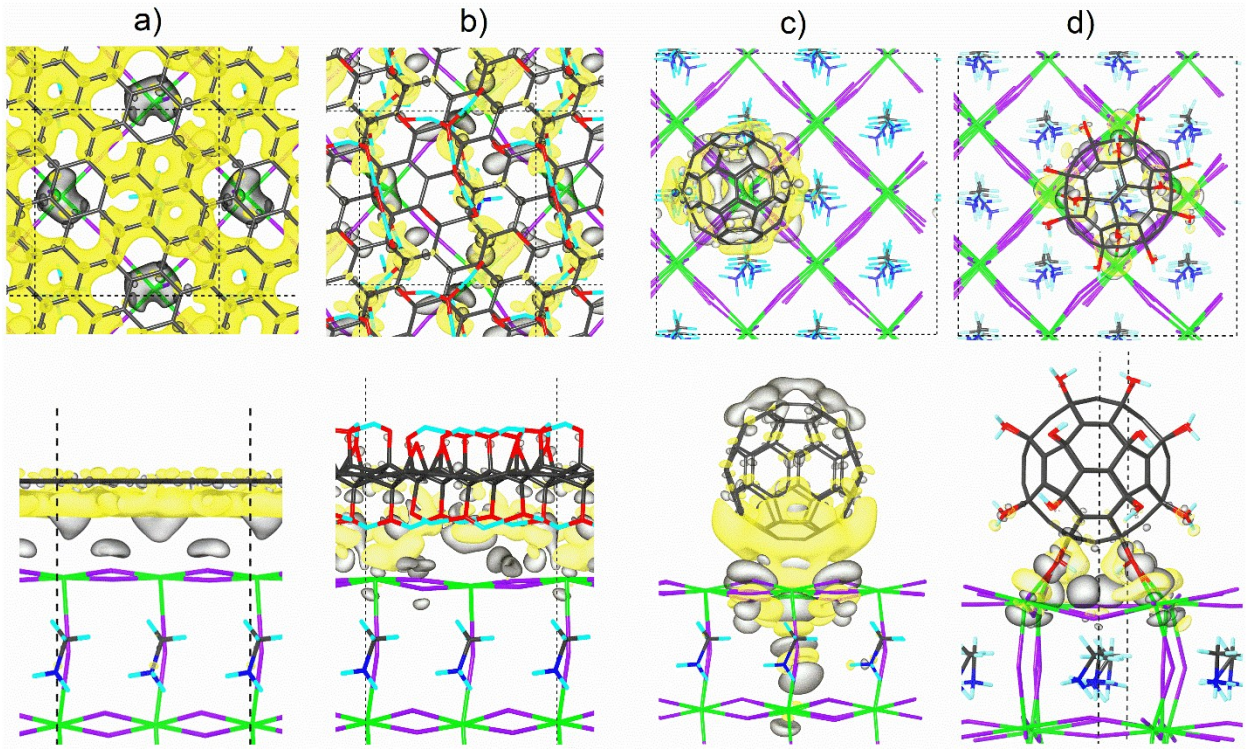
Figure S1 Isosurfaces of band decomposed charge density corresponding to valance band maximum (VBM) and conduction band minimum (CBM) according to PBE-D3 for **a)** $MAPbI_3$, **b)** graphene/ $MAPbI_3$, **c)** GO/ $MAPbI_3$, **d)** C_{60}/ $MAPbI_3$, and **e)** $C_{60}(OH)_{24}$/ $MAPbI_3$.



Colour code: Pb in green, I in violet, C in dark grey, N in blue, H in cyan, O in red.

Figure S2 Top and side view on isosurfaces of electron charge density difference according to PBE-D3 for

a) graphene/ $MAPbI_3$, **b)** GO/ $MAPbI_3$, **c)** C_{60}/ $MAPbI_3$ and **d)** $C_{60}(OH)_{24}$/ $MAPbI_3$ interfaces.



Dark grey (yellow) corresponds to electron charge accumulation (depletion). Colour code: Pb in green, I in violet, C in dark grey, N in blue, H in cyan, O in red.

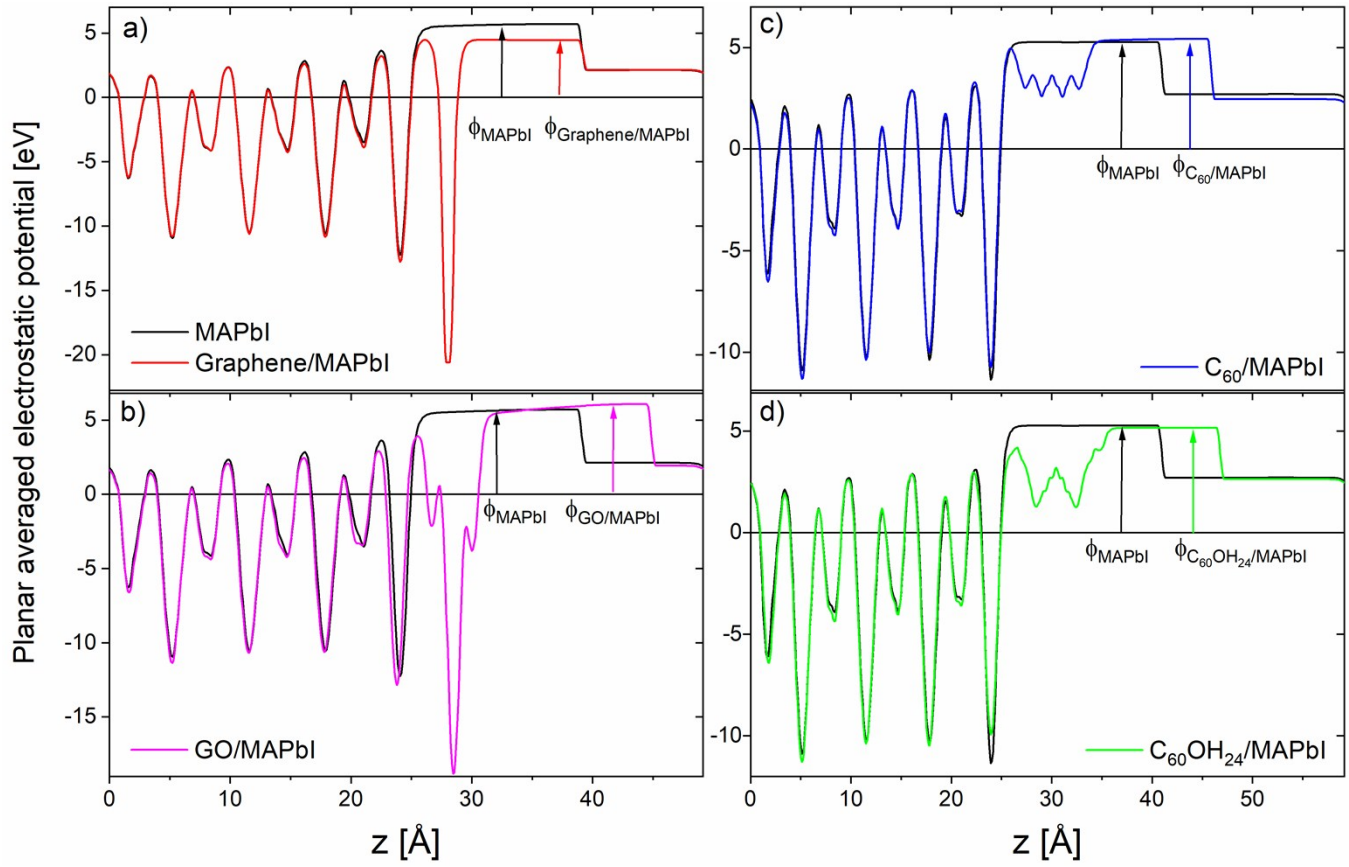
Table S3. Charge density analysis.

	Graphene/ $MAPbI_3$	GO/ $MAPbI_3$	C_{60}/ $MAPbI_3$	$C_{60}(OH)_{24}$/ $MAPbI_3$
Q_{Bader} [*e*]	0.004	0.101	0.000	0.201
$\mu(z)$ [D]	– 1.42	1.58	0.83	– 0.34
$V(z)$ [eV]	– 0.29	0.31	0.15	– 0.08

Charge density analysis according to PBE-D3 of graphene/ $MAPbI_3$, GO/ $MAPbI_3$, C_{60}/ $MAPbI_3$ and $C_{60}(OH)_{24}$/ $MAPbI_3$ interfaces: Bader charges (Q_{Bader}) – positive value corresponds to the

electron charge transfer from $MAPbI_3$ to carbon nanostructure; interface dipole moment ($\mu(z)$); interface electrostatic potential energy ($V(z)$).

Figure S3 Planar avaraged electrostatic (Hartree) potential computed with PBE-D3 for **a)** graphene/ $MAPbI_3$, **b)** GO/ $MAPbI_3$, **c)** C_{60}/ $MAPbI_3$ and **d)** $C_{60}(OH)_{24}$/ $MAPbI_3$ interfaces.



Fermi energy is set to 0 eV.

Table S4. Local electrostatic (Hartree) potential analysis.

	$MAPbI_3$	Graphene (1×1)	GO (1×1)	C_{60} (2×2)	$C_{60}(OH)_{24}$ (2×2)
IE^{pure} [eV]	5.89 (1×1) 5.73 (2×2)	4.20	5.87	5.84	6.17
$IE^{composite}$ [eV]	–	4.45	6.38	5.67	5.57
ϕ^{pure} [eV]	5.59 (1×1) 5.32 (2×2)	4.20	5.04	5.35	5.72
$\phi^{composite}$ [eV]	–	4.45	5.94	5.41	5.14
$\Delta\phi$ [eV]	–	– 1.14	0.35	0.09	– 0.18
CBO [eV]	–	0.00	– 2.48	– 0.16	– 0.24

Ionization energies (IE^{pure}, $IE^{composite}$), work functions (ϕ^{pure}, $\phi^{composite}$) and work function differences ($\Delta\phi$) computed from PBE-D3 local electrostatic (Hartree) potential analysis for pure components and composites of graphene/ $MAPbI_3$, GO/ $MAPbI_3$, C_{60}/ $MAPbI_3$ and $C_{60}(OH)_{24}$/ $MAPbI_3$. The conduction band offsets (CBO) were computed as the difference in electron affinities (EAs) of heterostructure components. EAs were evaluated with the use of HSE06-D3 E_g for $MAPbI_3$, graphene and GO, while PBE-D3 E_g for C_{60} and $C_{60}(OH)_{24}$.