

## Supplemental Information

### Oxygen-assisted monodisperse transition-metal atoms induced graphite phase transformation to diamond: A First-Principles Calculation

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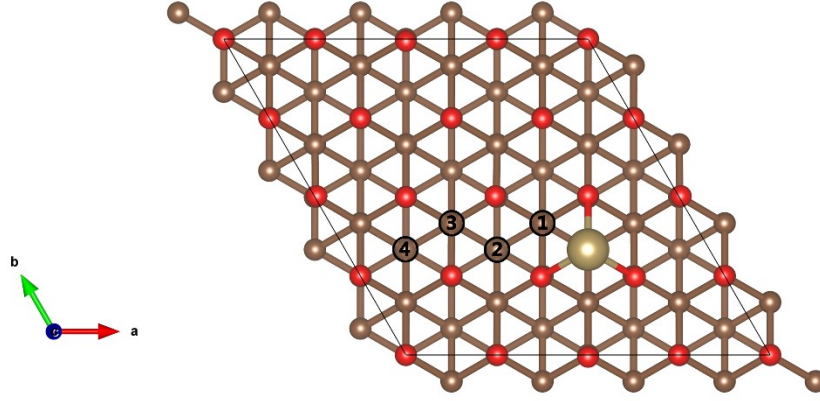
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**Table S1. The bond lengths and bond angles information for Bulk diamond, Graphene, and diamane.**

Systems	$d_1(\text{C-C})/\text{\AA}$	$d_2(\text{C-C})/\text{\AA}$	$d_3(\text{C-C})/\text{\AA}$	$\alpha/^\circ$	$\beta/^\circ$	$\gamma/^\circ$
Bulk diamond	1.540	1.540	1.540	109.5	109.5	109.5
Graphene	1.420	/	1.420	120.0	120.0	120.0
No-metal	1.479	1.704	1.704	117.0	100.1	111.3
Ti	1.529	1.654	1.513	112.3	106.0	112.0
Zr	1.518	1.665	1.526	111.5	107.0	111.6
Hf	1.519	1.669	1.525	110.8	107.6	111.5
Ta	1.535	1.635	1.529	109.5	109.4	111.3
Mo	1.530	1.619	1.525	110.8	109.9	111.1
W	1.555	1.609	1.528	109.4	109.4	111.0
Re	1.547	1.605	1.528	109.9	109.2	111.1
Os	1.540	1.611	1.526	110.2	110.6	111.3
Ir	1.537	1.609	1.531	110.3	111.2	111.3
Ni	1.519	1.621	1.523	106.5	109.7	113.5
Pd	1.527	1.618	1.524	111.3	110.4	113.6
Pt	1.535	1.601	1.524	110.7	111.4	111.3

**Table S2 Adsorption energy of TM atoms adsorbed on the O-terminated diamond surface**

TM atoms	Stable adsorption site	Adsorption energy / eV
Ti	FCC	-13.15
	HCP	-12.96
Zr	FCC	-14.54
	HCP	-14.38
Hf	FCC	-14.76
	HCP	-14.59
Ta	Bridge	-13.59
	FCC	-13.76
	HCP	-13.52
Mo	Bridge	-9.55
	FCC	-9.68
	HCP	-9.47
W	Bridge	-11.13
	FCC	-10.89
	HCP	-10.66
Re	Bridge	-8.97
	FCC	-8.63
	HCP	-8.47
Os	Bridge	-8.02
	FCC	-8.10
	HCP	-8.03
Ir	FCC	-7.22
	HCP	-7.18
Ni	FCC	-7.24
	HCP	-7.17
Pd	FCC	-4.55
	HCP	-4.51
Pt	FCC	-5.43
	HCP	-5.34



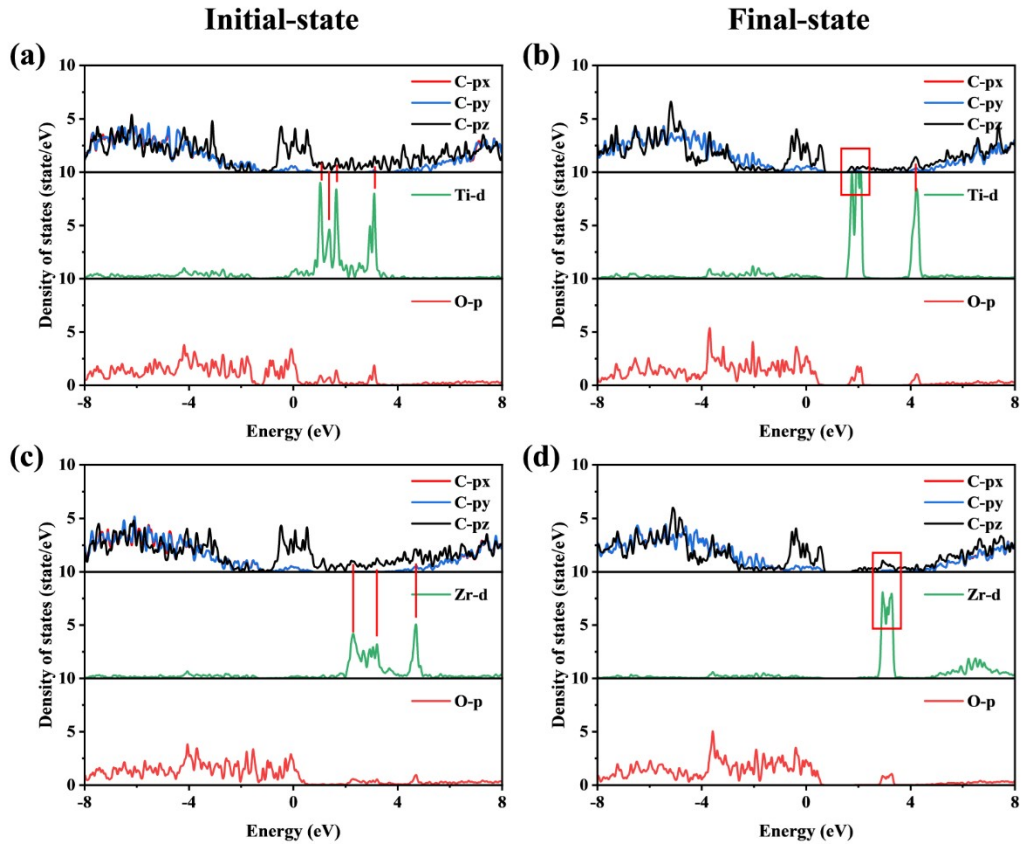
**Fig. S1 The adsorption sites for the second Ta atom on the O-terminated diamond (111) substrate**

Considering that TM atoms approach the surface along the vertical direction, using Ta as an example, we calculated the stable adsorption model for the adsorption of a second Ta atom on the O-terminated diamond (111) substrate where one Ta atom has already been adsorbed. The adsorption energy was calculated as

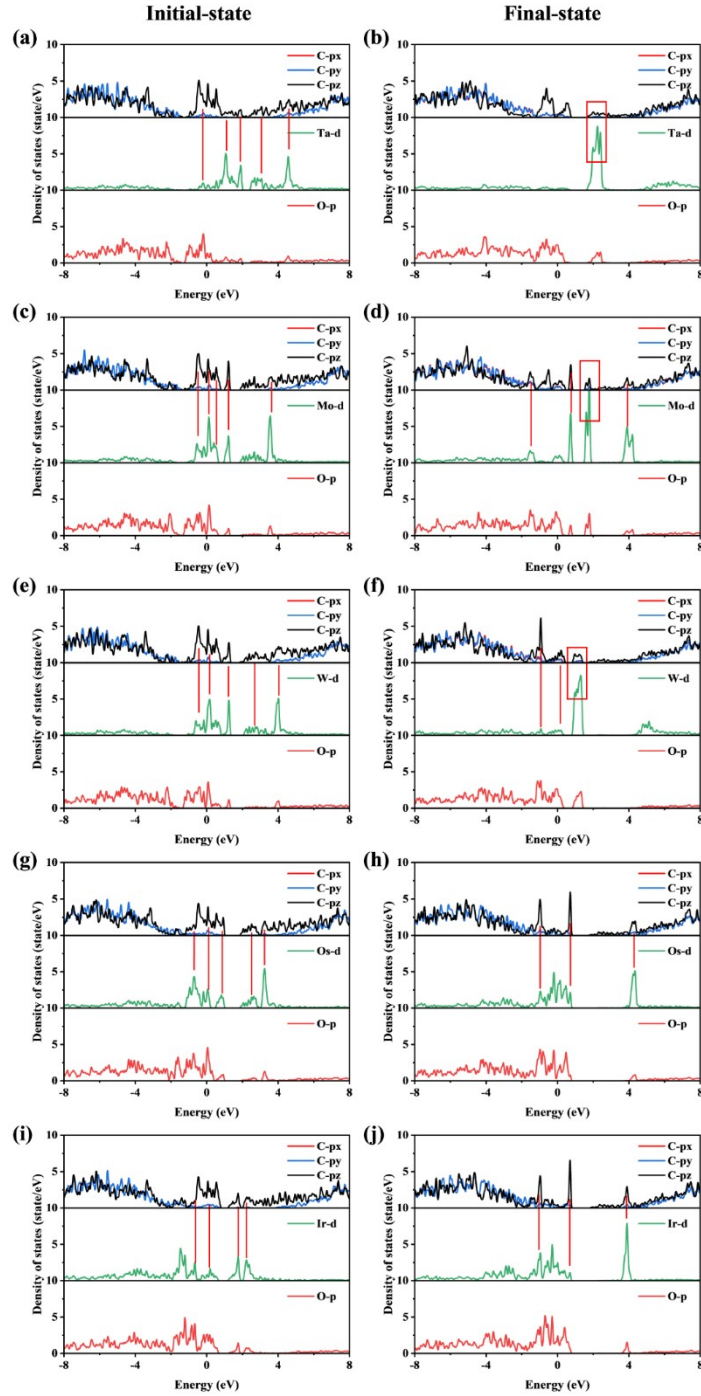
$$E_{ads} = \frac{(E_{surf+nTM} - E_{surf} - nE_{TM})}{n}$$

where  $E_{surf+nTM}$  is the total energy of the adsorption model,  $E_{surf}$  is the energy of O-terminated diamond (111) substrate,  $E_{TM}$  is the energy of a single TM atom, and  $n$  is number of adsorbed TM atom.

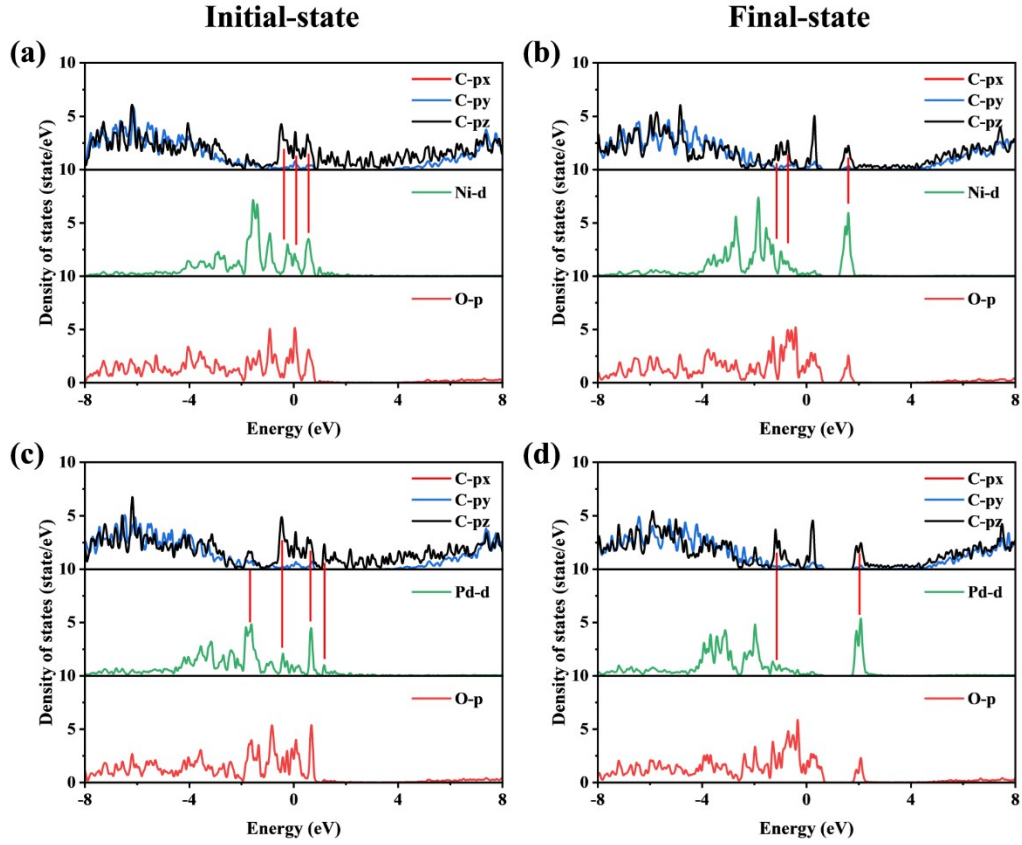
As shown in **Fig. S1**, there are four adsorption sites for the second Ta atom on the O-terminated diamond (111) substrate. We placed the second Ta atom above these adsorption sites for structure optimization to obtain stable adsorption structures. There are three stable adsorption structures. The  $E_{ads}$  at position 2 is -11.92 eV, the  $E_{ads}$  at position 3 is -12.62 eV, and the  $E_{ads}$  at position 4 is the most negative (-12.83 eV). This suggests that the second TM atom is more stable at position 4 and the falling TM atoms will be adsorbed farther away from the adsorbed TM atom when TM atoms are vertically close to O-terminated diamond (111) substrate. Thus, the O-terminated diamond (111) substrate is more effective in suppressing the aggregation of TM atoms more effectively.



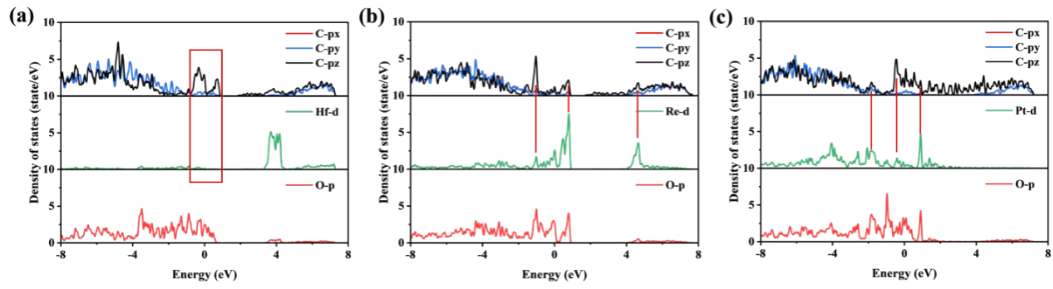
**Fig. S2.** The partial density of states (PDOS) for initial-state and final-state structures of other Type I systems with the outer valence electron state of  $d^2s^2$  for Ti (a, b) and Zr (c, d). C represents the carbon atoms of bottom graphene. (The short vertical red line indicates the position of the overlapping peaks, and the Fermi level position is set to 0 eV)



**Fig. S3.** The partial density of states (PDOS) for initial-state and final-state structures of other Type II systems with the outer valence electron state of  $d^3s^2-d^7s^2$  for Ta (a, b), Mo (c, d), W (e, f), Os (g, h), and Ir (i, j). C represents the carbon atoms of bottom graphene. (The short vertical red line indicates the position of the overlapping peaks, and the Fermi level position is set to 0 eV)

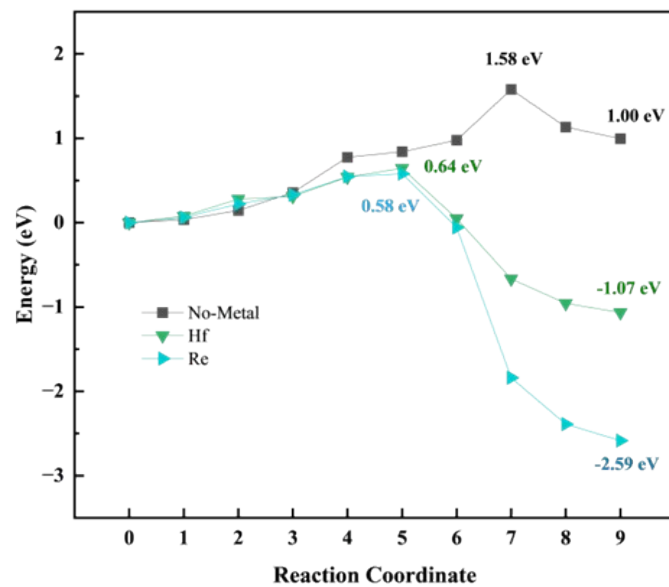


**Fig. S4.** The partial density of states (PDOS) for initial-state and final-state structures of other Type III systems with the outer valence electron state of  $d^9s^1$  for Ni (a, b) and Pd (c, d). C represents the carbon atoms of bottom graphene. (The short vertical red line indicates the position of the overlapping peaks, and the Fermi level position is set to 0 eV)

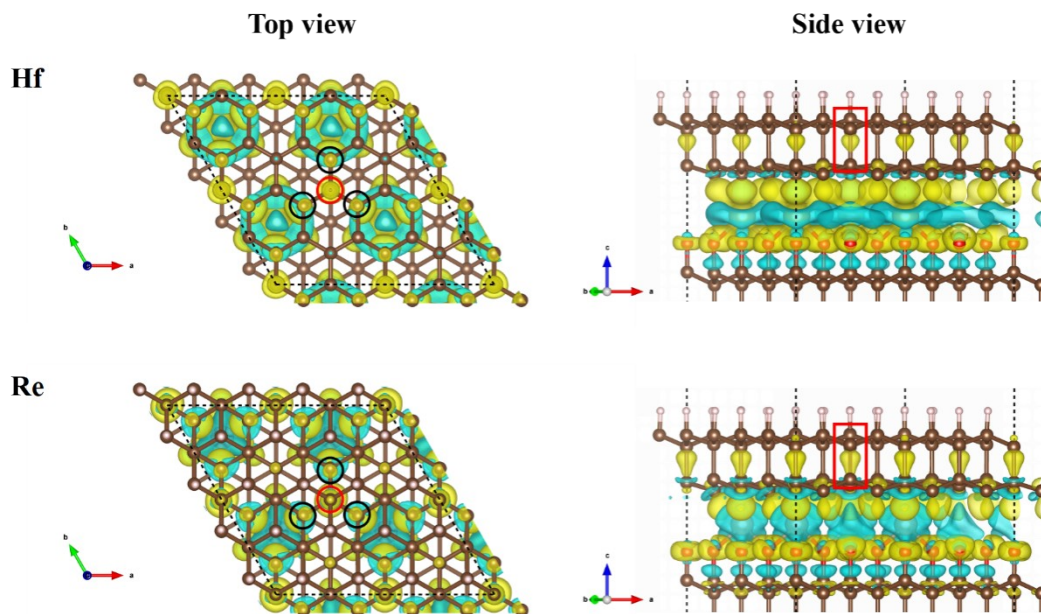


**Fig. S5.** The partial density of state (PDOS) of final-state structures for Hf, Re, and Pt systems calculated using the DFT+U theory ( $U_{\text{Hf}} = 2.71$ ,  $U_{\text{Re}} = 2.32$ , and  $U_{\text{Pt}} = 2.42$ ). C represents the carbon atoms of bottom graphene. The Fermi level position is set to 0 eV.

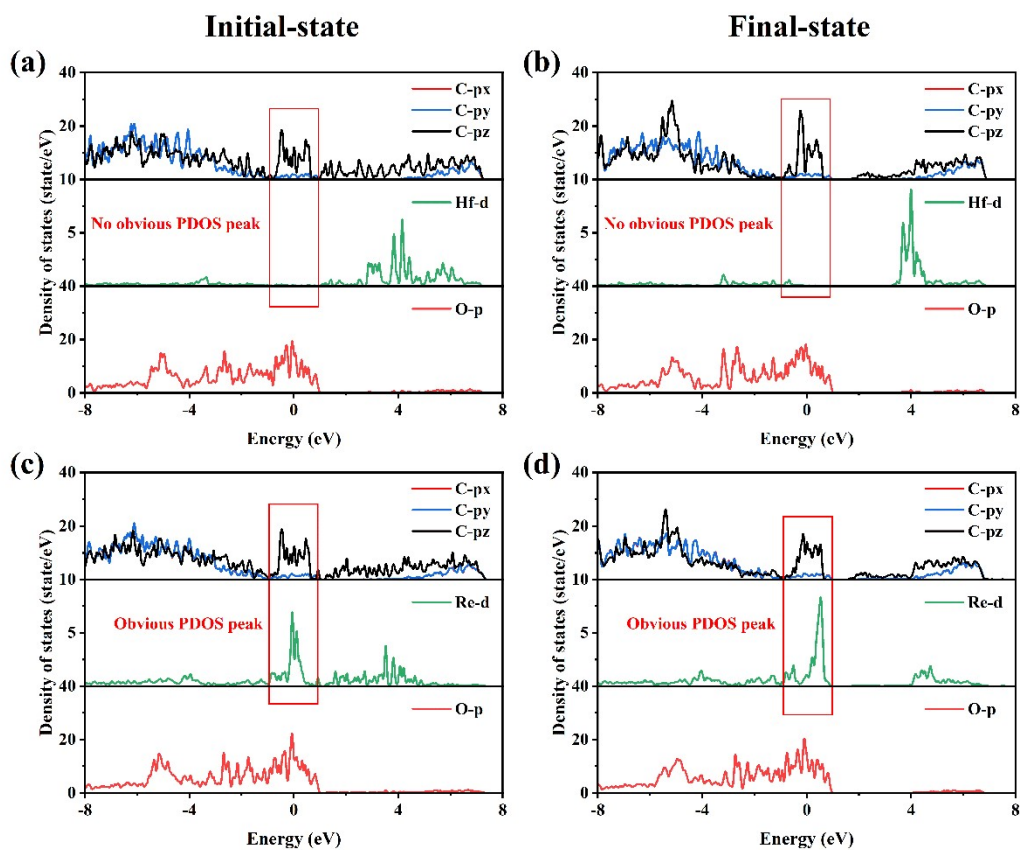




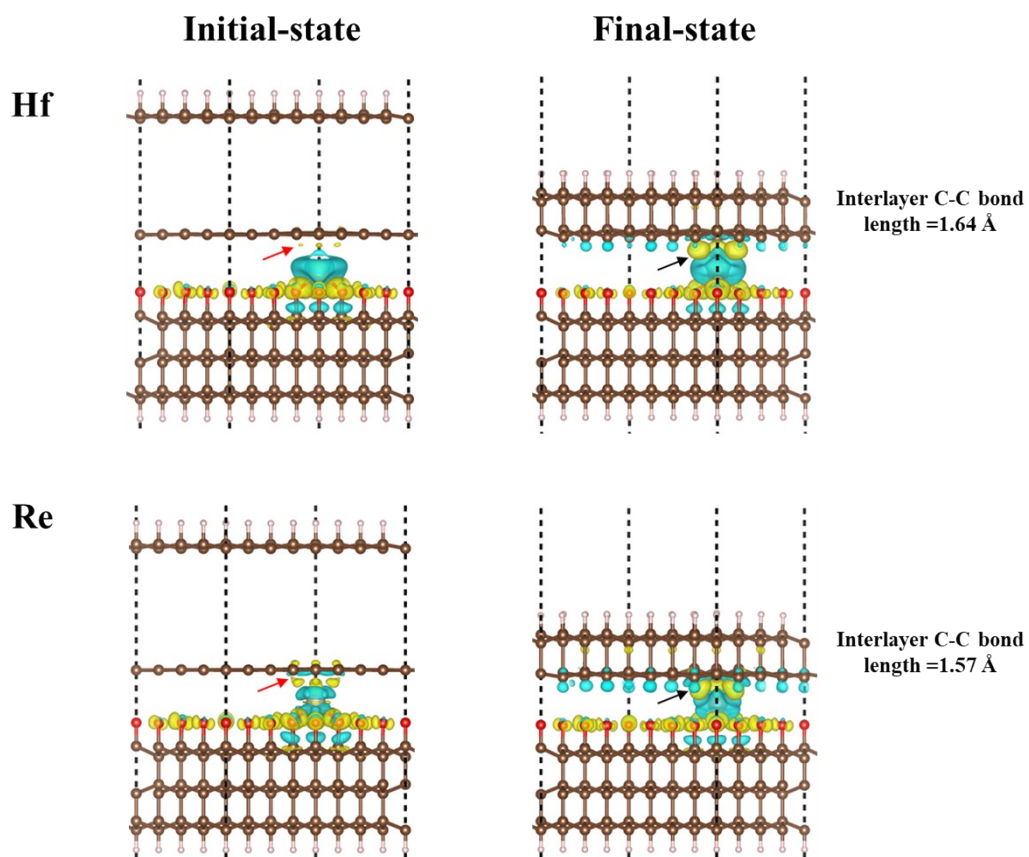
**Fig. S6 Energy changes of phase transformation from 3-layer graphene to diamond in the No-Metal, Hf, and Re systems.**



**Fig. S7** Differential charge density of final-state structures for Hf and Re systems calculated using 4×4 supercells including four TM atoms (The lateral dimensions of approximately is 10.08 Å and TM coverage is 25%). The isosurface level is set to 0.006 e/ Bohr<sup>3</sup>.



**Fig. S8** Partial density of state (PDOS) of initial- and final-state structures for Hf (a, b) and Re (c, d) systems calculated using  $4\times 4$  supercells including one TM atom (The lateral dimensions of approximately is  $10.08 \text{ \AA}$  and TM coverage is 6.25%). C represents the carbon atoms of bottom graphene. The Fermi level position is set to 0 eV.



**Fig. S9** Differential charge density of initial- and final-state structures for Hf and Re systems calculated using  $4 \times 4$  supercells including one TM atom (The lateral dimensions of approximately is 10.08 Å and TM coverage is 6.25%). The isosurface level is set to  $0.006 \text{ e} / \text{Bohr}^3$ .