Electronic Supporting Information

Molybdenum Carbide Supported Metal Catalysts (M_n/Mo_xC; M = Co, Ni, Cu, Pd, Pt) – Metal and Surface Dependent Structure and Stability

Fan Wang,[a] Teng Li,[b] Yun Shi[c] and Haijun Jiao[a*]*

a) Leibniz-Institut für Katalyse e.V. an der Universität Rostock, Albert-Einstein-Str. 29a, 18059 Rostock, Germany. b) State Key Laboratory of Coal Conversion, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan, 030001, China. c) School of Chemistry & Chemical Engineering, Linyi University, Linyi 276000, China.

E-mail: shiyun@lyu.edu.cn and haijun.jiao@catalysis.des

Content

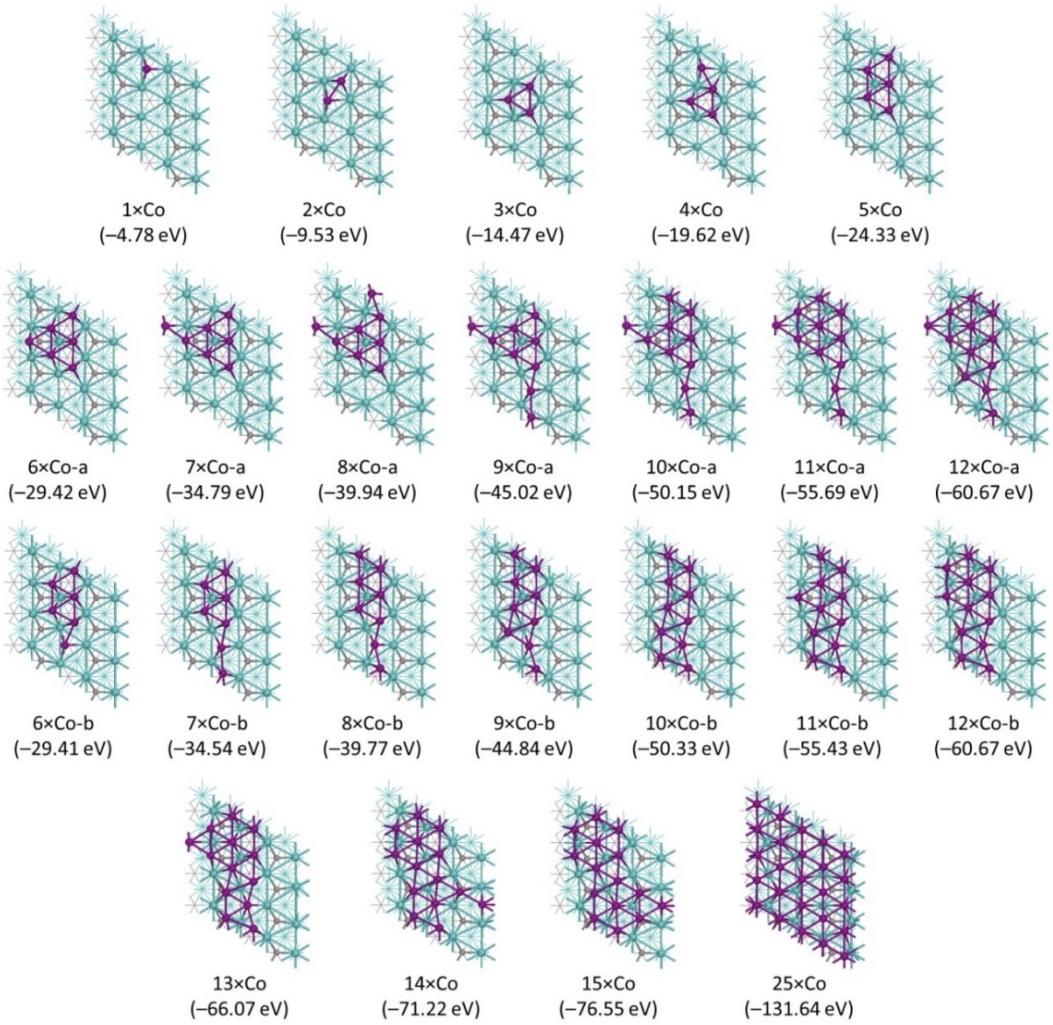

Figure S1. Structures and adsorption energies of aggregation mode for Co_n (n = 1-15, 25) on Mo_2C(001) (Co/purple, C/gray, Mo/cyan).

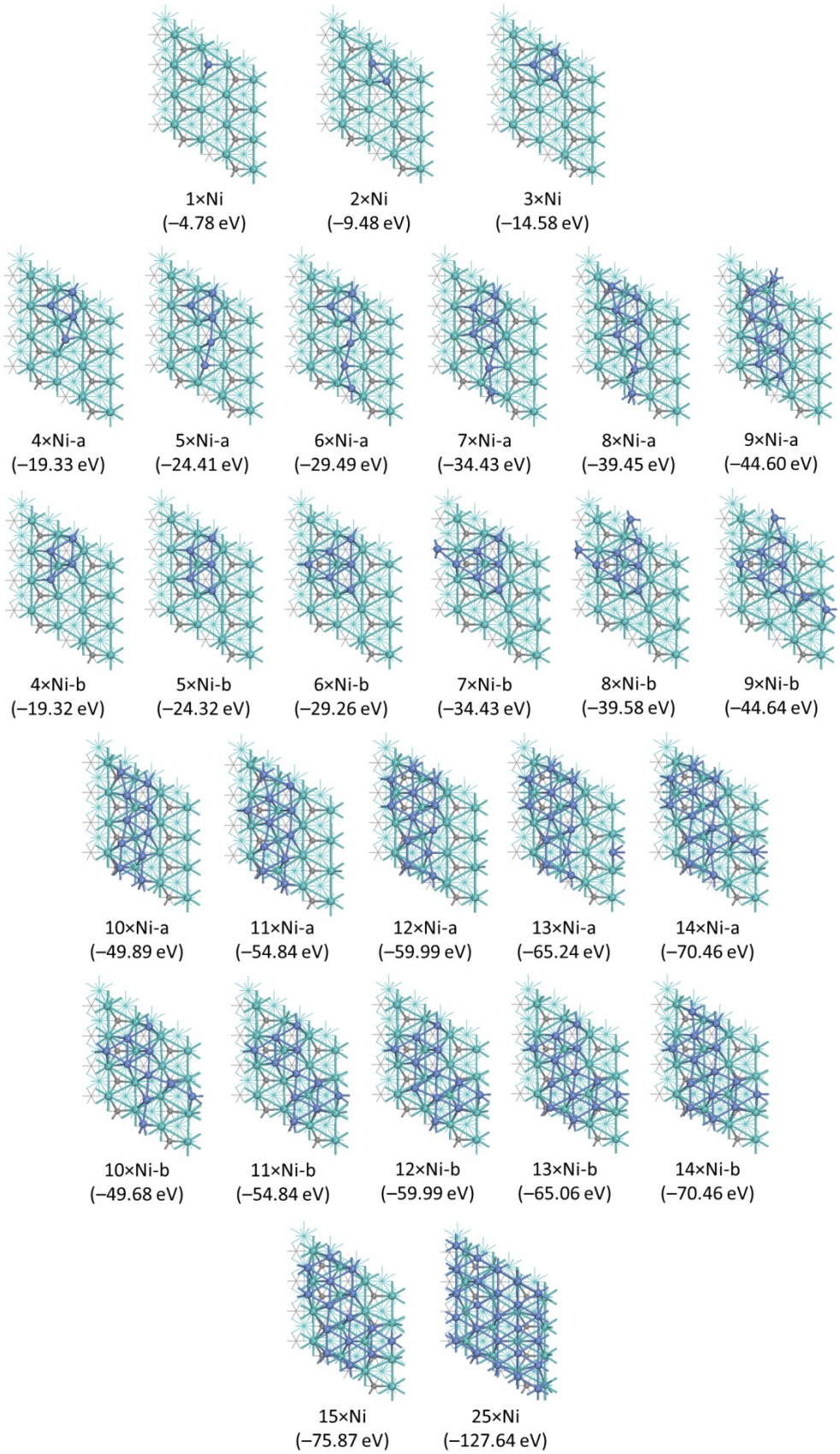

Figure S2. Structures and adsorption energies of aggregation mode for Ni_n (n = 1-15, 25) on Mo_2C(001) (Ni/gray blue, C/gray, Mo/cyan).

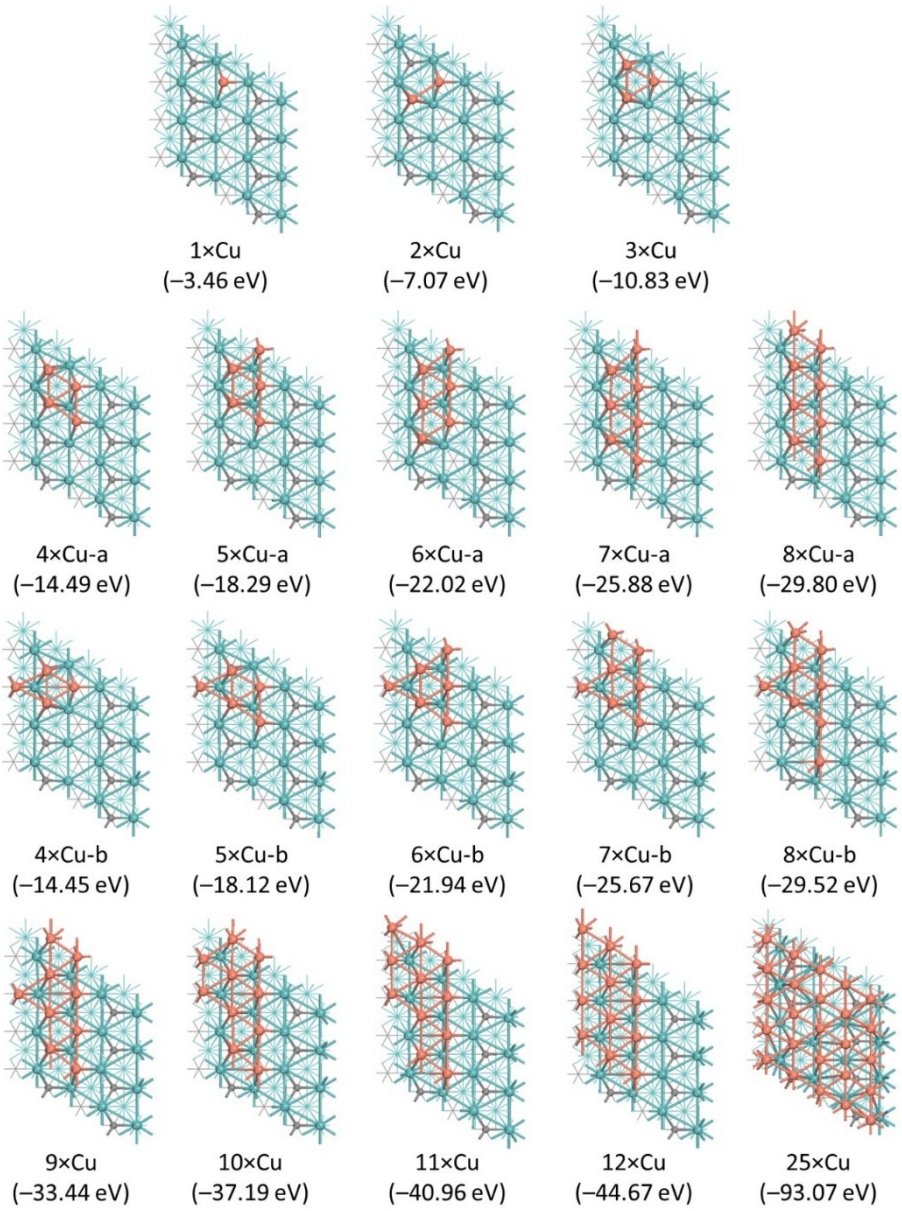

Figure S3. Structures and adsorption energies of aggregation mode for Cu_n (n = 1-12, 25) on Mo_2C(001) (Cu/orange, C/gray, Mo/cyan).

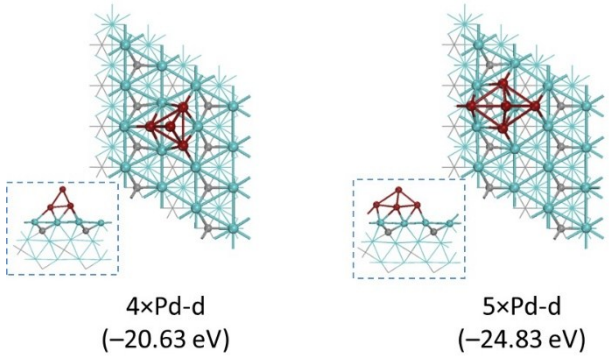

Figure S4. Most stable adsorption configurations and energies of 3D (side views in square) mode for Pd_n (n = 4, 5) on Mo_2C(001) (Pd/brown, C/gray, Mo/cyan).

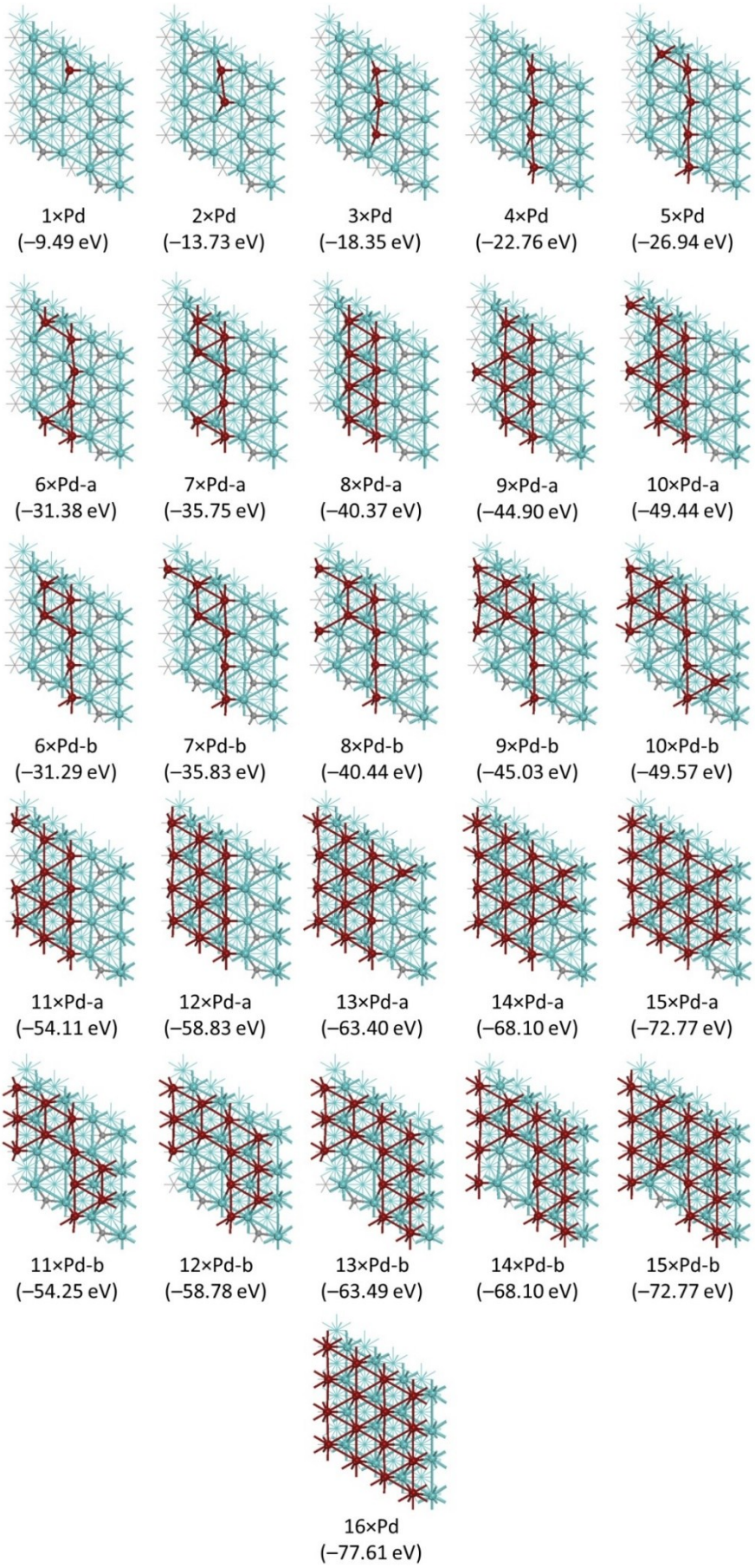

Figure S5. Structures and adsorption energies of aggregation mode for Pd_n (n = 1-16) on Mo_2C(001) (Pd/brown, C/gray, Mo/cyan).

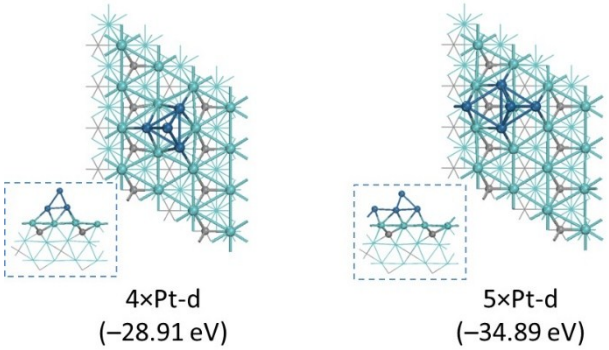

Figure S6. Most stable adsorption configurations and energies of 3D (side views in square) mode for Pt_n (n = 4, 5) on Mo_2C(001) (Pt/blue, C/gray, Mo/cyan).

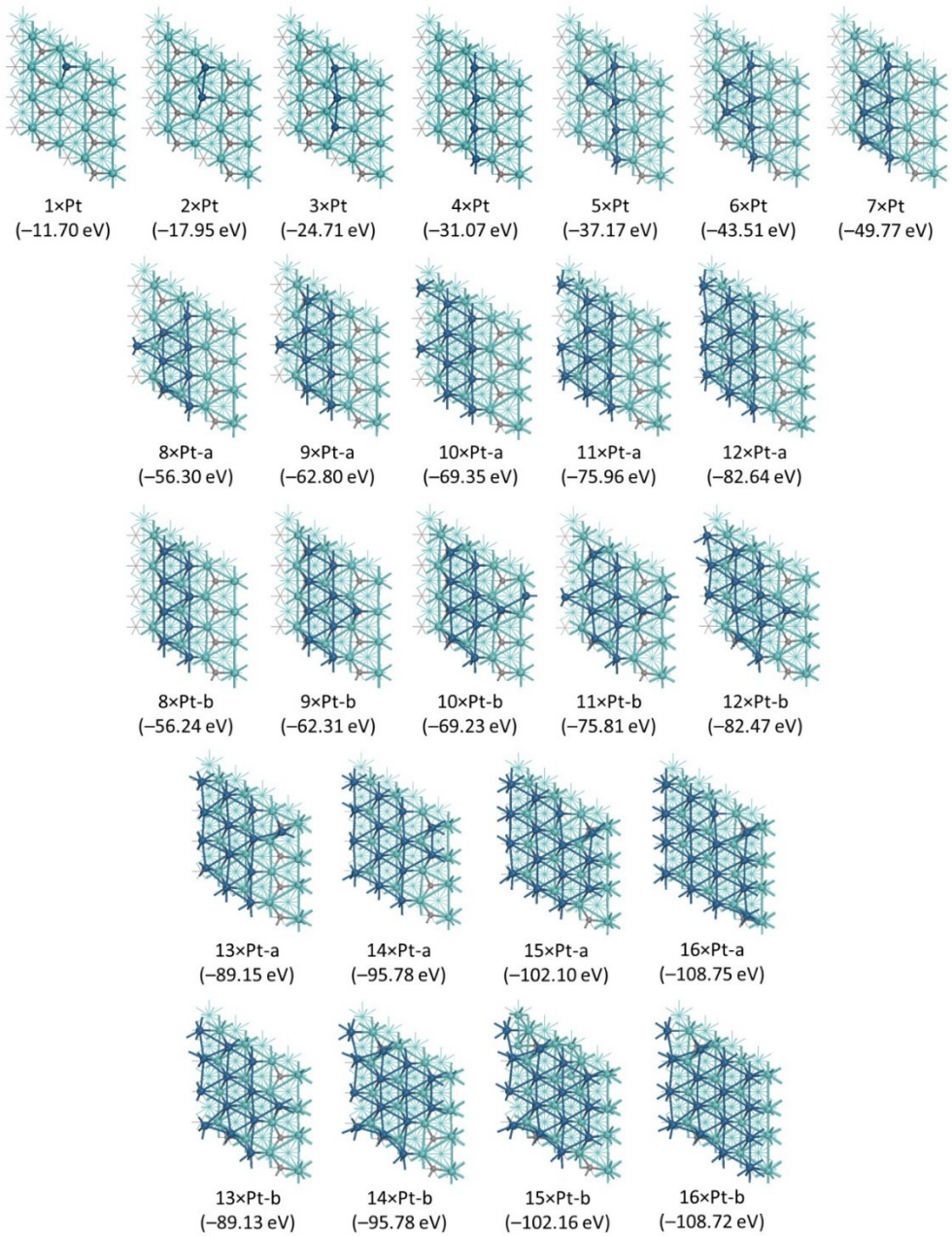

Figure S7. Structures and adsorption energies of aggregation mode for Pt_n (n = 1-16) on Mo_2C(001) (Pt/blue, C/gray, Mo/cyan).

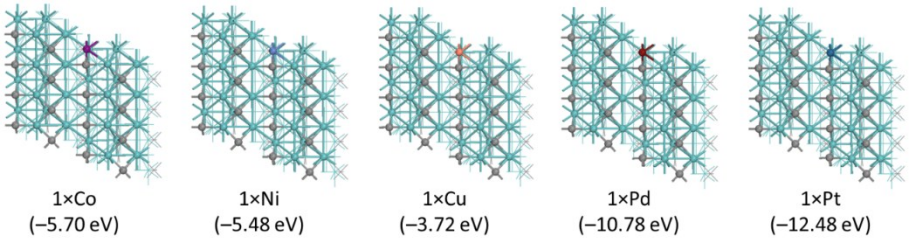

Figure S8. Most stable adsorption configurations and energies for single Co, Ni, Cu, Pd, Pt atoms on Mo_2C(101) (Co/purple, Ni/gray blue, Cu/orange, Pd/brown, Pt/blue, C/gray, Mo/cyan).

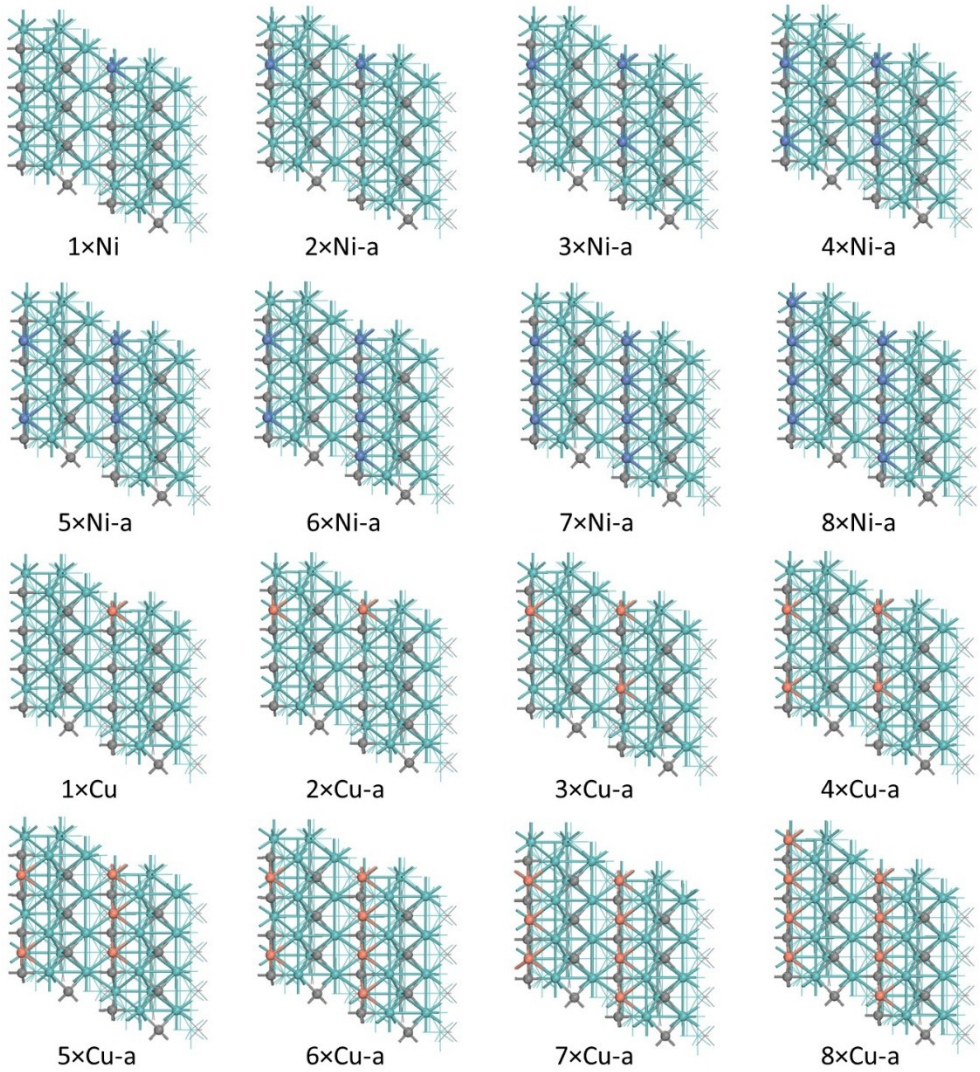

Figure S9. Structures of dispersion mode for Ni_n and Cu_n (n = 1-8) on Mo_2C(101) (Ni/gray blue, Cu/orange, C/gray, Mo/cyan).

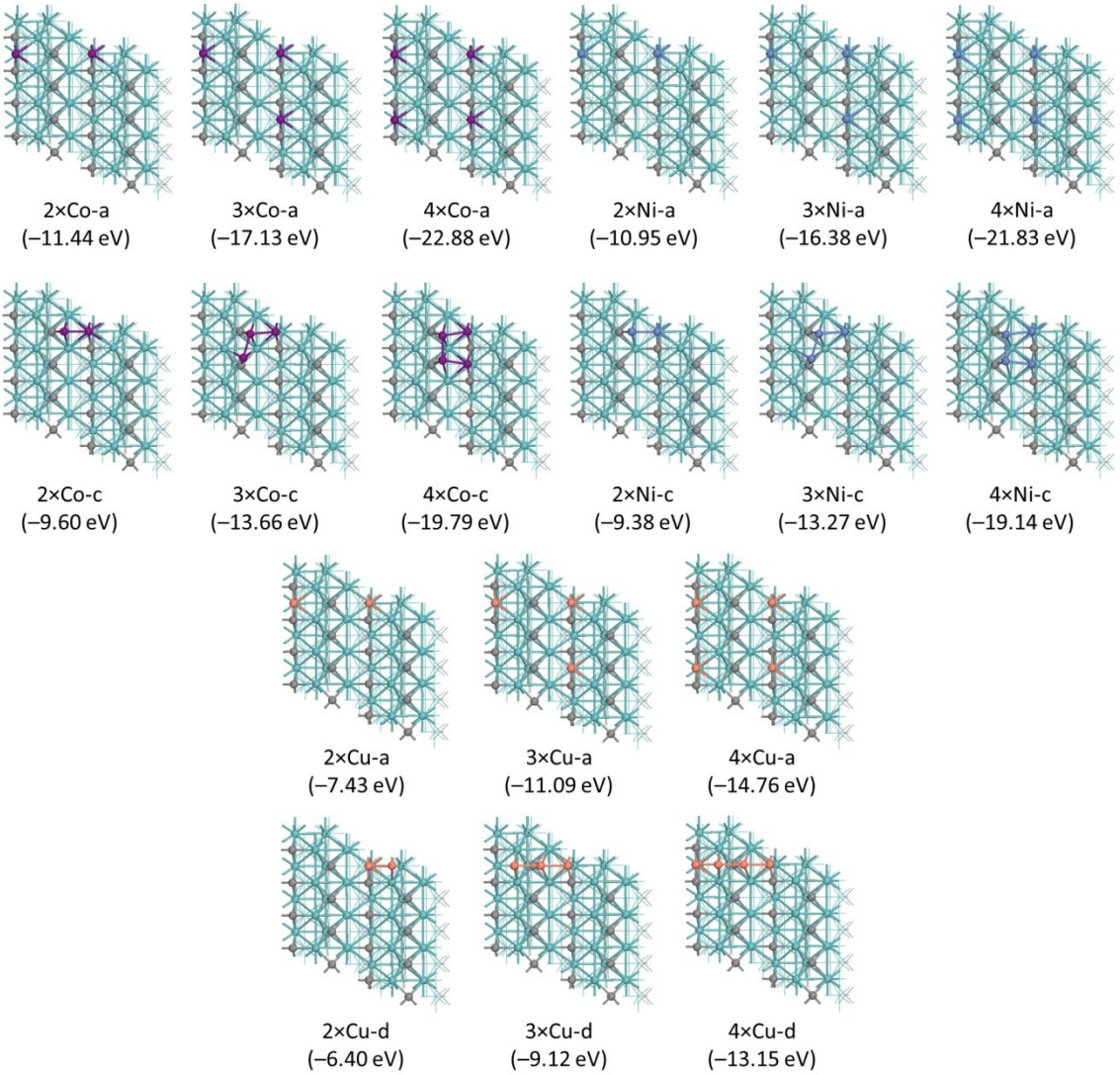

Figure S10. Structures and adsorption energies of dispersion and aggregation mode for Co_n, Ni_n, and Cu_n, (n = 2-4) on Mo_2C(101) (Co/purple, Ni/gray blue, Cu/orange, C/gray, Mo/cyan).

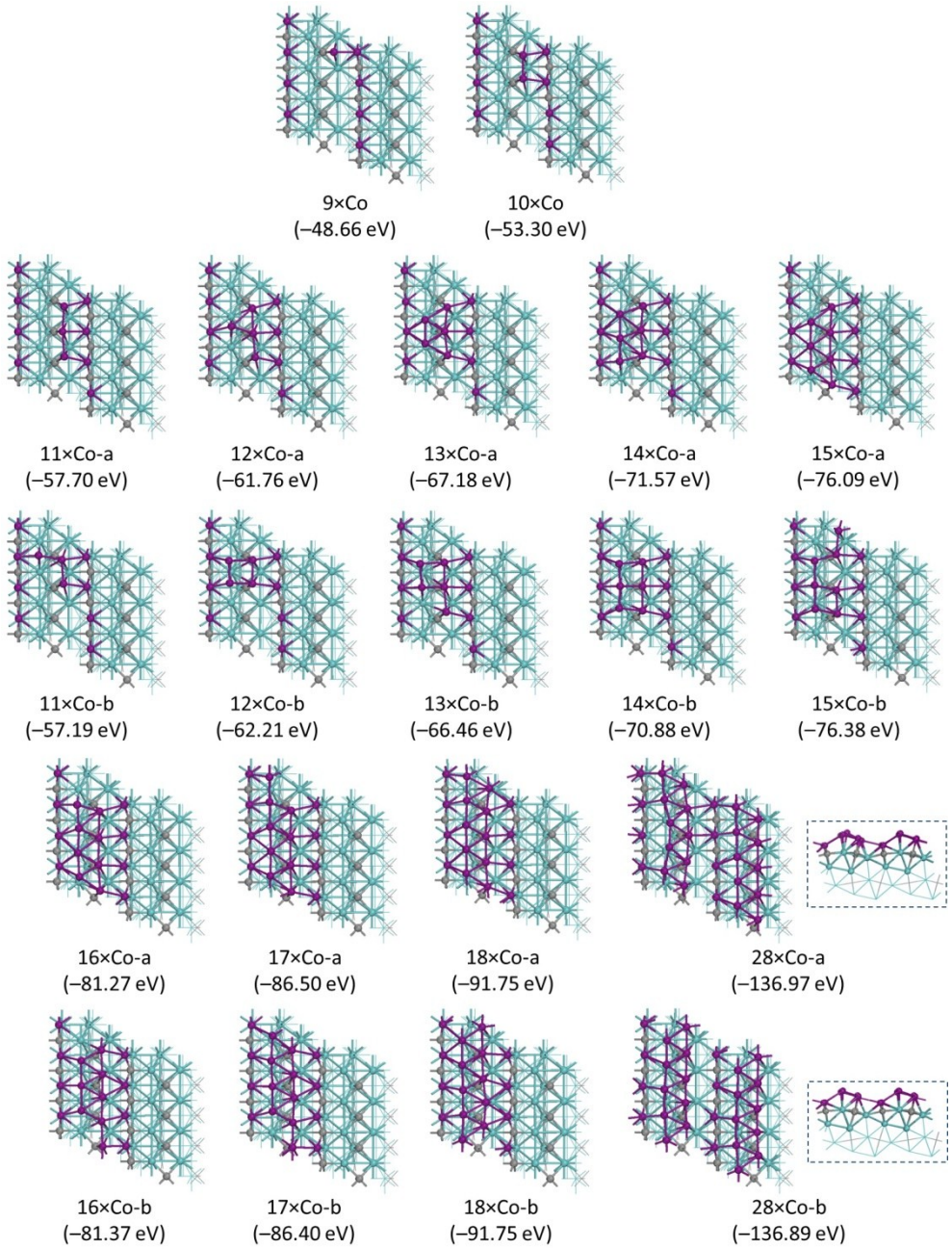

Figure S11. Structures (side views in square) and adsorption energies of aggregation mode for Co_n (n = 9-18, 28) on Mo_2C(101) (Co/purple, C/gray, Mo/cyan).

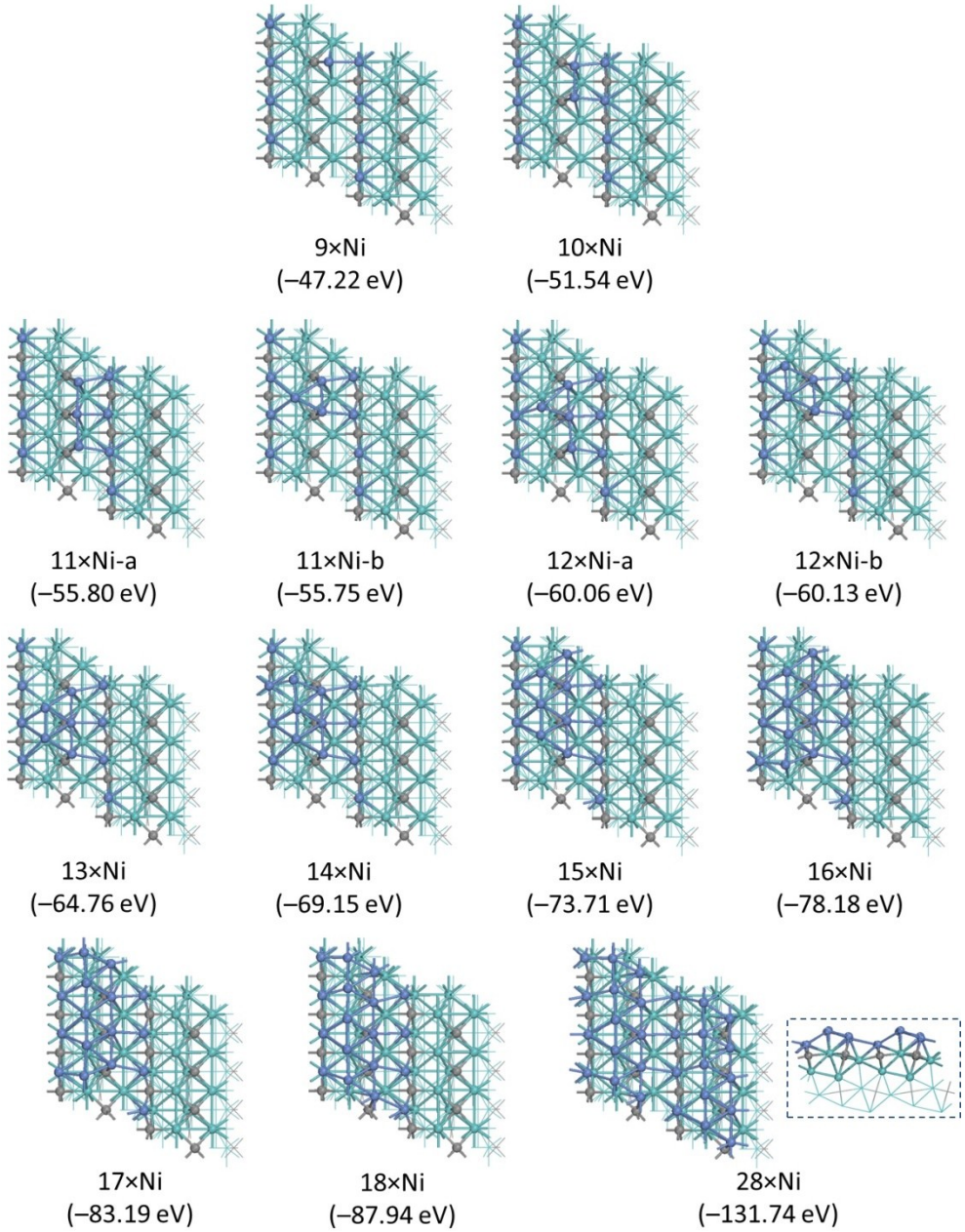

Figure S12. Structures (side views in square) and adsorption energies of aggregation mode for Ni_n (n = 9-18, 28) on $Mo_2C(101)$ (Ni/gray blue, C/gray, Mo/cyan).

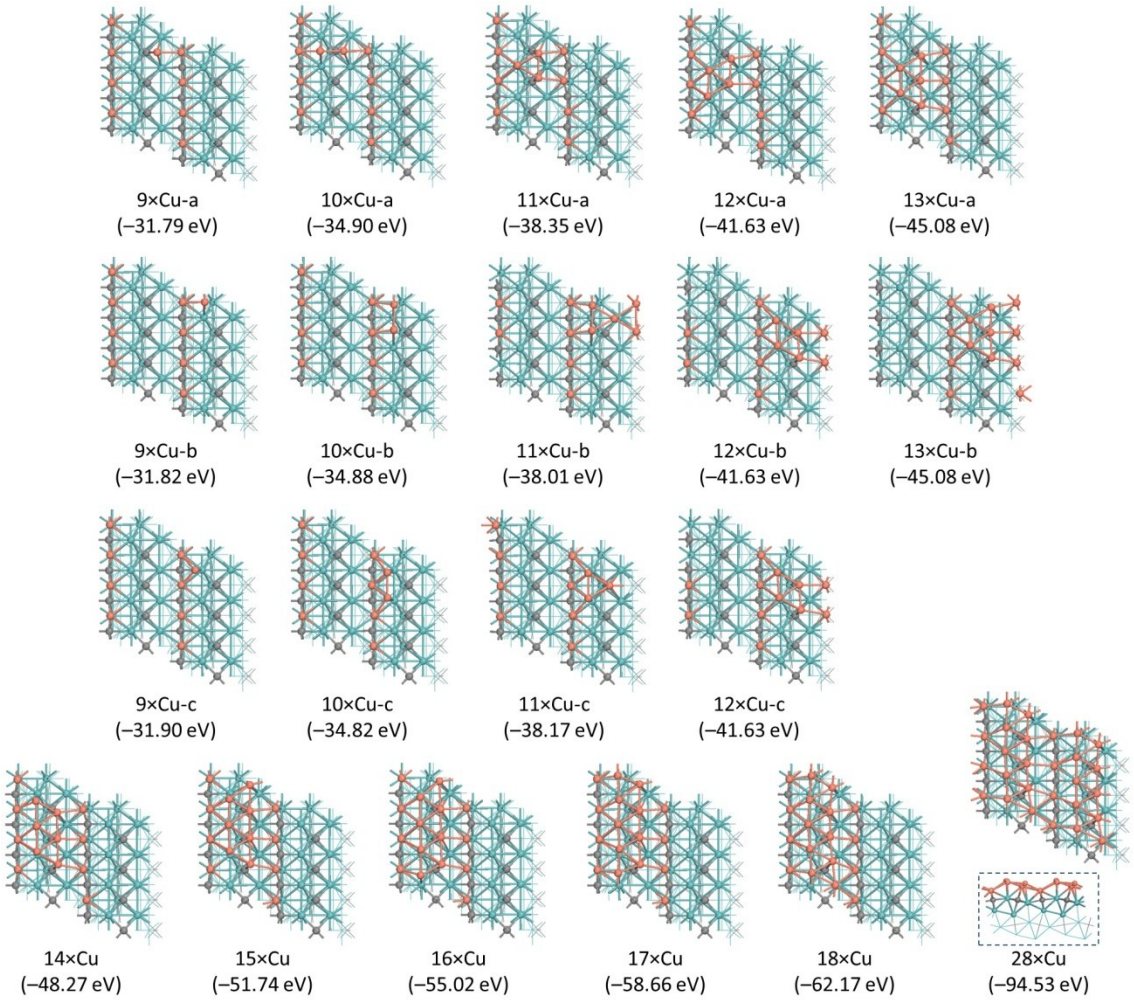

Figure S13. Structures (side views in square) and adsorption energies of aggregation mode for Cu_n (n = 9-18, 28) on Mo_2C(101) (Cu/orange, C/gray, Mo/cyan).

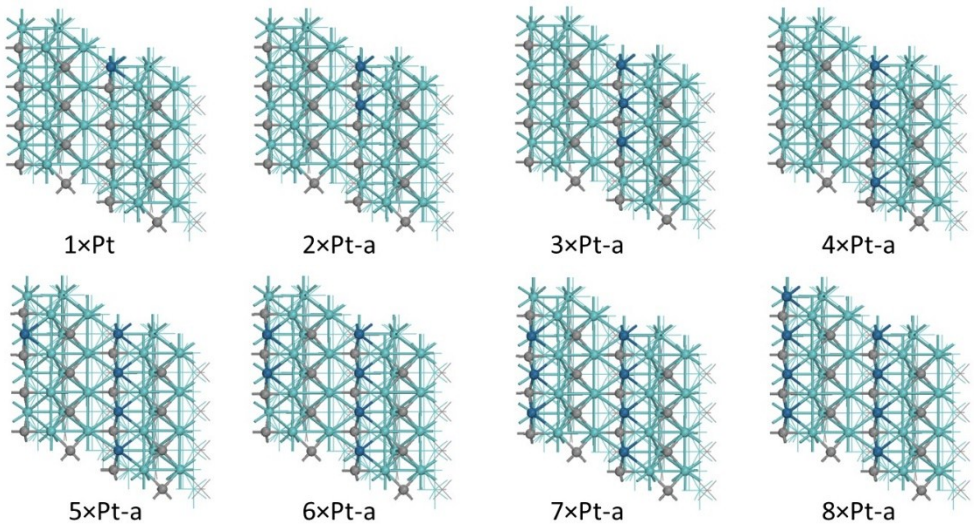

Figure S14. Structures of dispersion mode for Pt_n (n = 1-8) on Mo_2C(101) (Pt/blue, C/gray, Mo/cyan).

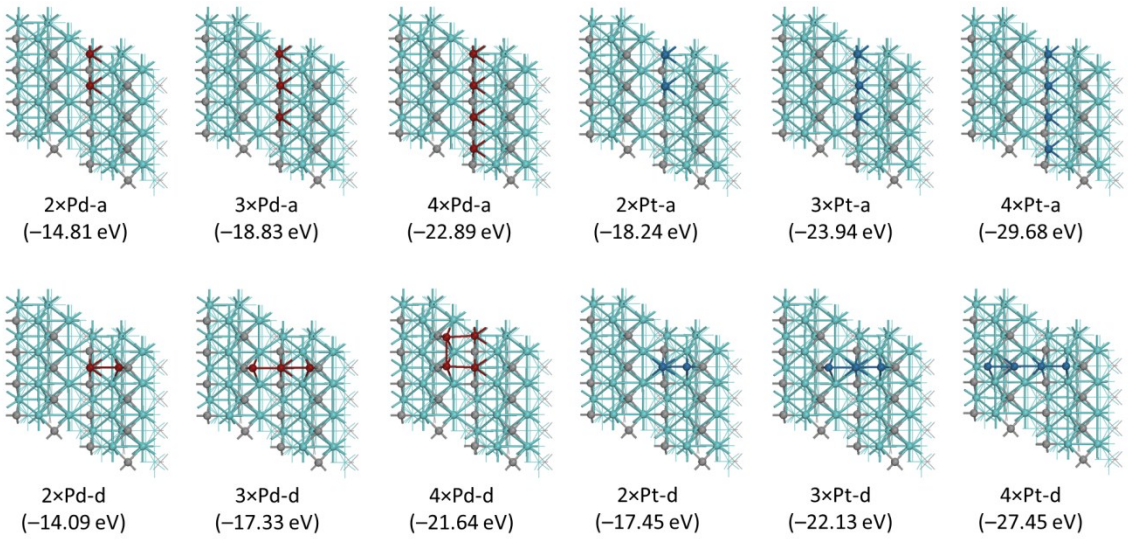

Figure S15. Structures and adsorption energies of dispersion and aggregation mode for Pd_n and Pt_n, (n = 2-4) on Mo_2C(101) (Pd/brown, Pt/blue, C/gray, Mo/cyan).

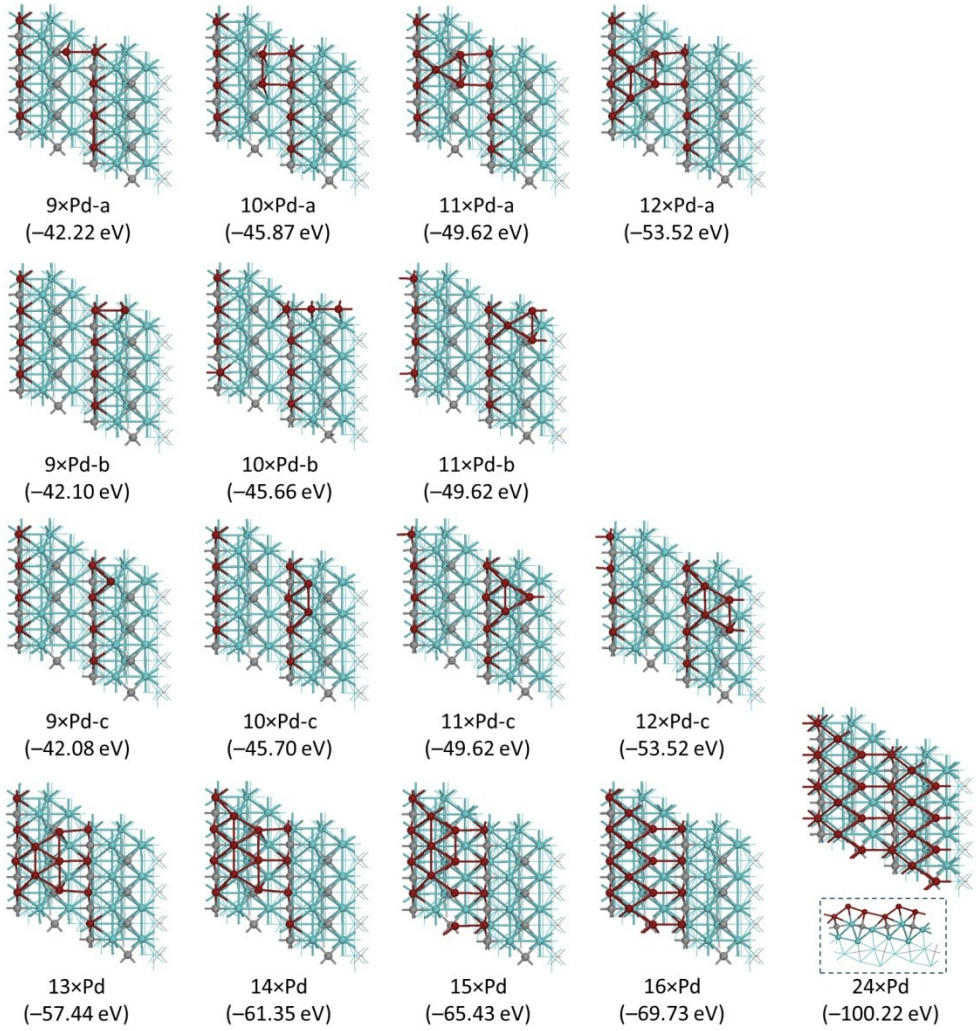

Figure S16. Structures (side views in square) and adsorption energies of aggregation mode for Pd_n (n = 9-16, 24) on Mo_2C(101) (Pd/brown, C/gray, Mo/cyan).

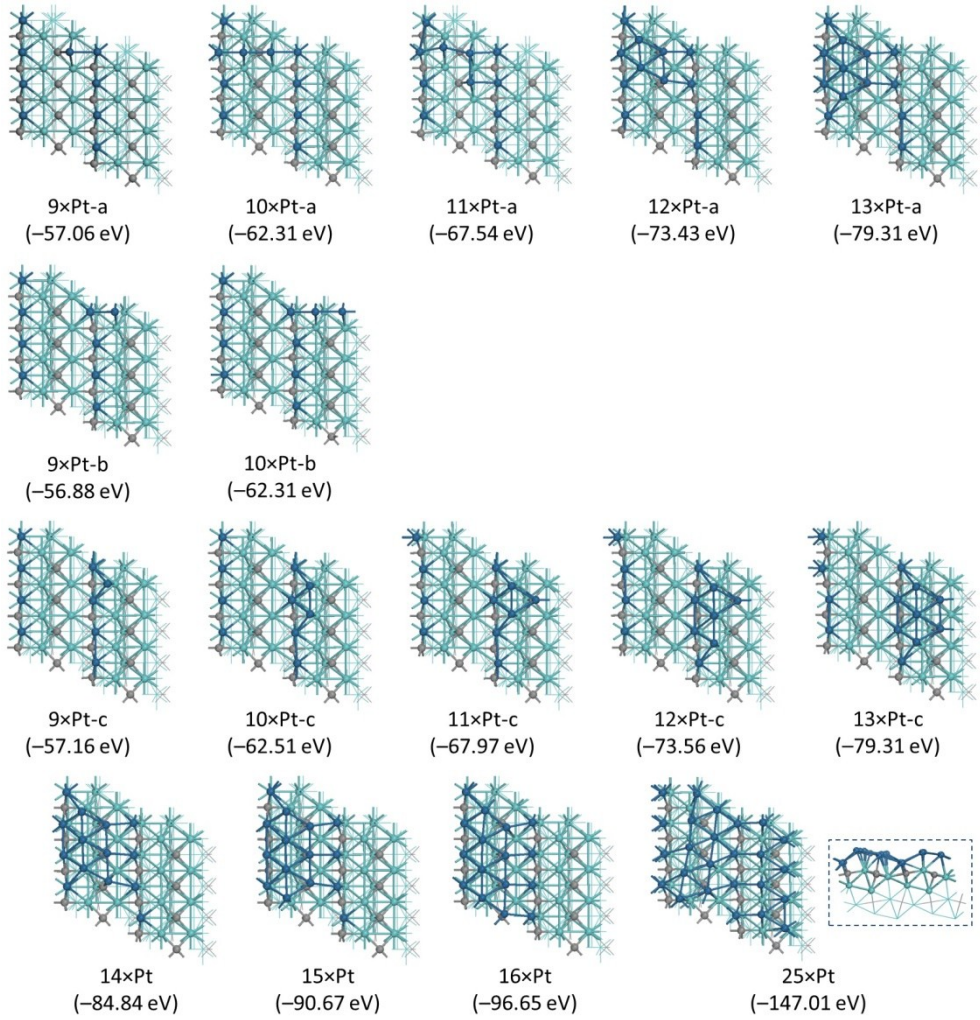

Figure S17. Structures (side views in square) and adsorption energies of aggregation mode for Pt_n (n = 9-16, 25) on Mo_2C(101) (Pt/blue, C/gray, Mo/cyan).

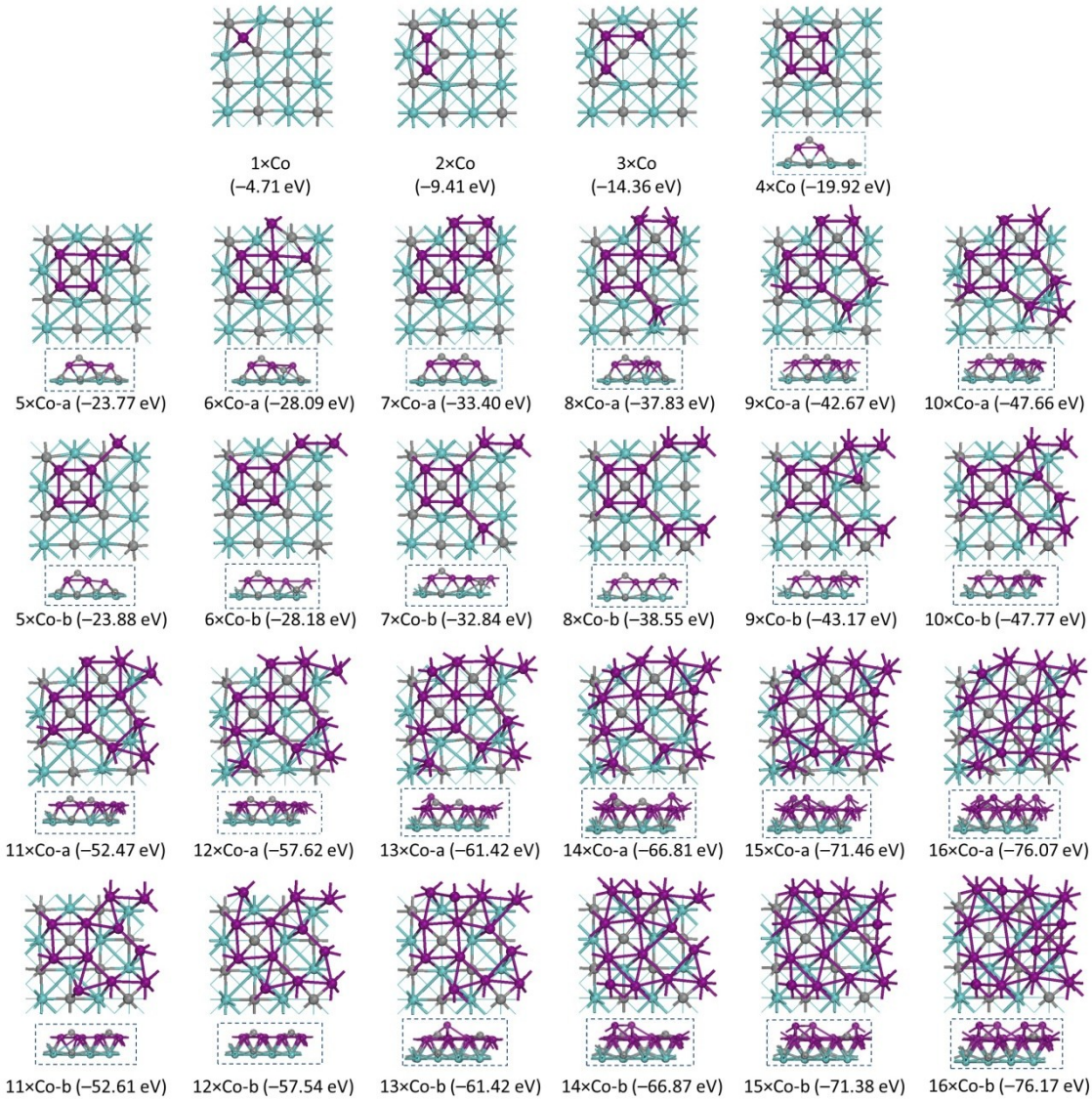

Figure S18. Top and side (in square) views of the structures and adsorption energies of aggregation mode for Co_n (n = 1-16) on MoC(001) (Co/purple, C/gray, Mo/cyan).

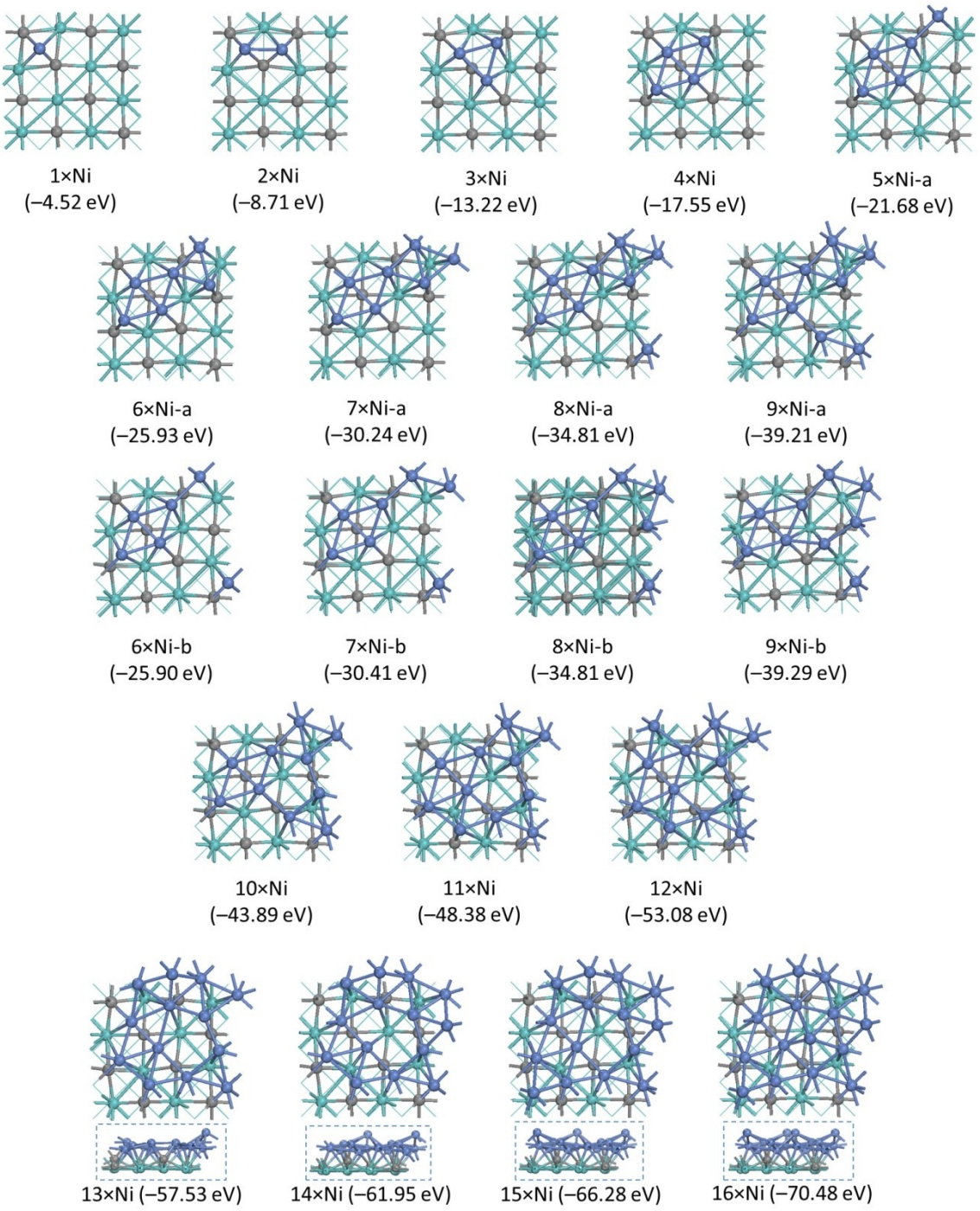

Figure S19. Top and side (in square) views of the structures and adsorption energies of aggregation mode for Ni_n (n = 1-16) on MoC(001) (Ni/gray blue, C/gray, Mo/cyan).

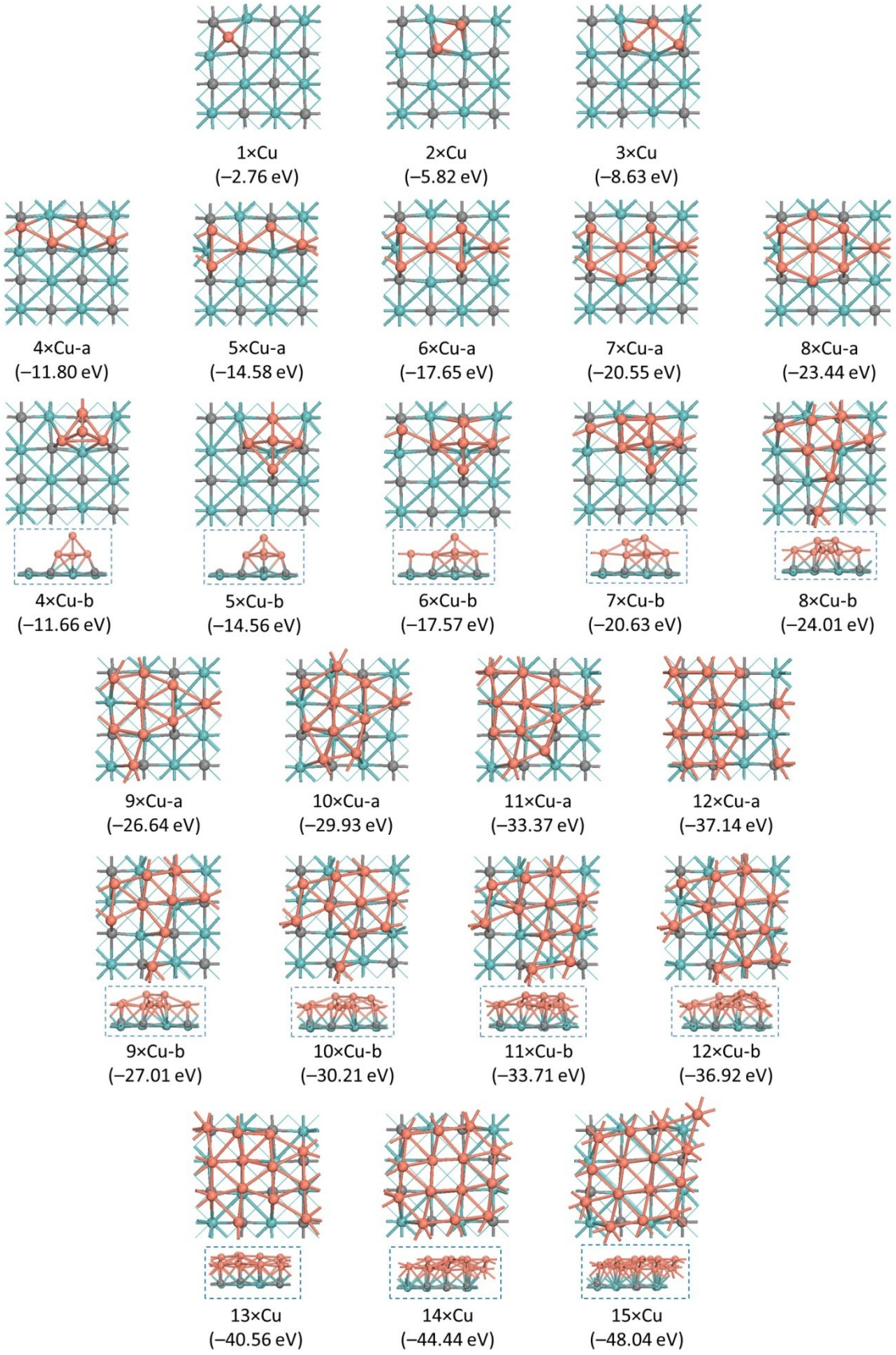

Figure S20. Top and side (in square) views of the structures and adsorption energies of aggregation mode for Cu_n (n = 1-15) on MoC(001) (Cu/orange, C/gray, Mo/cyan).

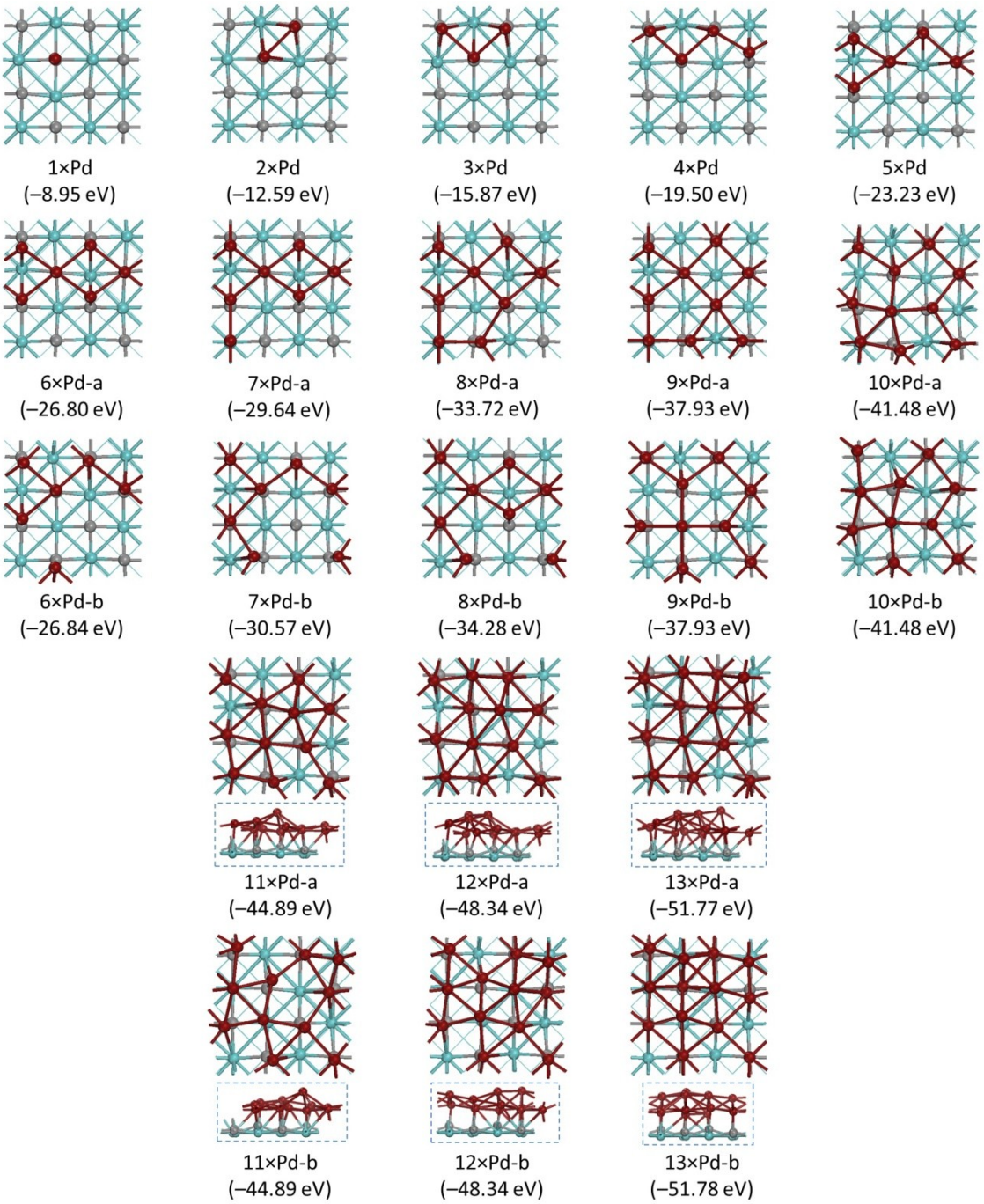

Figure S21. Top and side (in square) views of structures and adsorption energies of aggregation mode for Pd_n (n = 1-13) on MoC(001) (Pd/brown, C/gray, Mo/cyan).

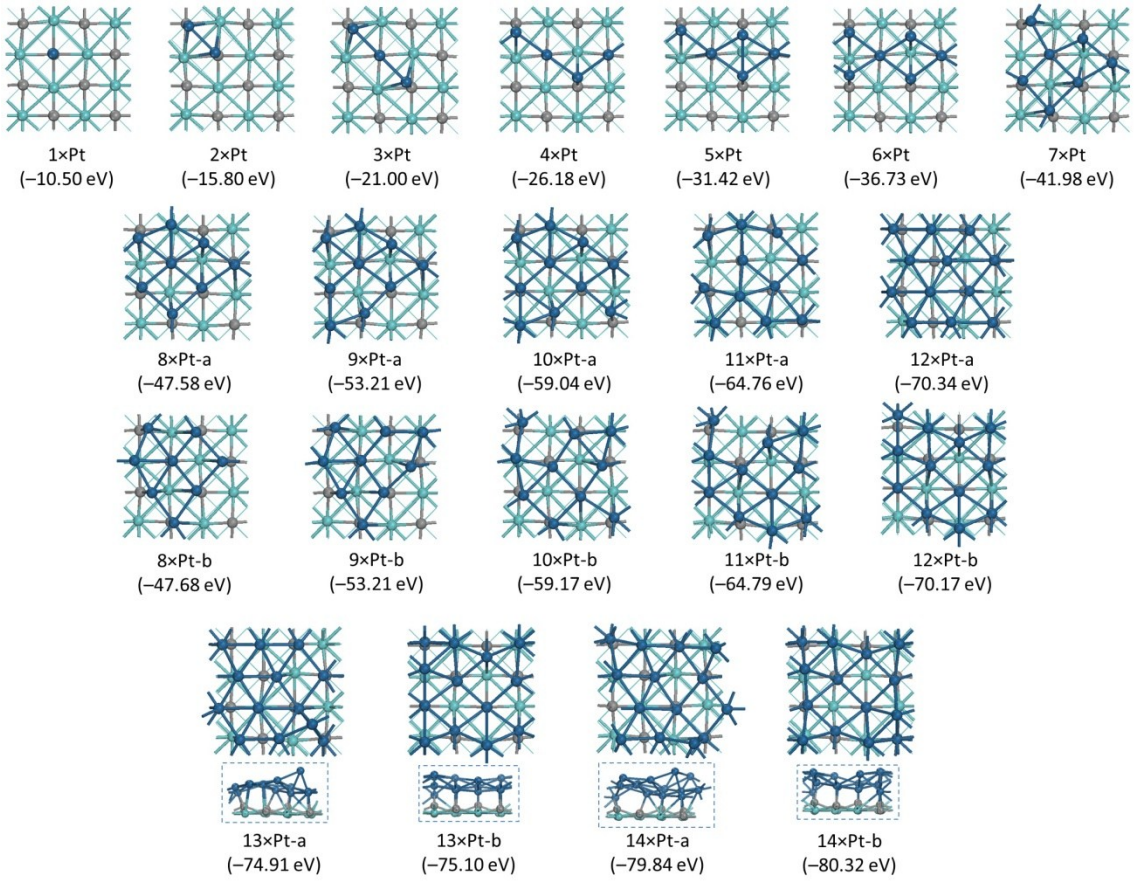

Figure S22. Top and side (in square) views of structures and adsorption energies of aggregation mode for Pt_n (n = 1-14) on MoC(001) (Pt/blue, C/gray, Mo/cyan).

Table S1. Average aggregation energy [$E_{agg}(M_{gas})$, eV], stepwise growth energy [$E_{growth}(M_{gas})$, eV], average M-M distance (R, Å) and average magnetic moment (M, μ_B) of M_n clusters in gas phase (M = Co, Ni, Cu, Pd, Pt).

M_n	$-E_{agg}(M_{gas})$	$E_{growth}(M_{gas})$	R	M
Co_2	1.54 (2.51,[1] 1.45[2])	–3.07	1.992 (1.95,[1] 1.95,[3] 1.96[2])	2.05 (2.00,[1] 2.08,[4] 2.00[2])
Co_3	1.91 (2.92,[1] 1.78[2])	–2.65	2.201 (2.16,[1] 2.19[2])	2.07 (1.63,[1] 2.33[2])
Co_4	2.39 (3.24[a], 2.44[b], 3.36,[1] 2.27[2])	–3.84	2.338 (2.34,[1] 2.34[2])	2.20 (2.50,[1] 2.50[2])
Co_5	2.68 (3.52,[1] 2.55[2])	–3.84	2.346 (2.40,[1] 2.34[2])	2.25 (2.00,[1] 2.60[2])
Co_6	3.05(4.06[a], 3.13[b], 2.93[2])	–4.92	2.279(2.27[2])	2.10 (2.33[2])
Ni_2	1.45 (1.74,[1]1.67[5])	–2.90	2.090 (2.10,[1] 2.07,[5] 2.20,[6] 2.155±0.01[7])	1.00 (1.00,[1] 1.00[5])
Ni_3	1.87 (2.00,[1] 2.20[5])	–2.72	2.207 (2.23,[1] 2.18[5])	0.76 (0.70,[1] 0.67[5])
Ni_4	2.18 (2.91[a], 2.32[b], 2.33,[1] 2.51[5])	–3.10	2.262 (2.31,[1] 2.25[5])	0.85 (1.00,[1] 1.00[5])
Ni_5	2.44 (2.50,[1] 2.85[8])	–3.48	2. 309 (2.34,[1] 2.36[8])	0.78 (0.78,[1] 0.80[8])
Ni_6	2.67 (3.61[a], 2.83[b], 3.06[8])	–3.79	2.320 (2.33[8])	1.07 (1.33[8])
Cu_2	1.13 (1.02±0.09,[9] 0.96,[10] 1.04,[11]1.16,[12] 1.06,[13] 1.13[14])	–2.26 (–2.26[14])	2.216 (2.25,[10] 2.21,[11] 2.25,[12] 2.52,[13] 2.22[14])	–
Cu_3	1.22 (1.07±0.12,[9] 1.00,[10] 0.98,[11] 1.20,[12] 1.19,[13] 1.23,[14] 1.43[15])	–1.39 (–1.43[14])	2.335 (2.45,[10] 2.34,[11] 2.40,[12] 2.54,[13] 2.35,[14] 2.25[15])	0.07
Cu_4	1.59 (1.88[a], 1.63[b], 1.48±0.14,[9] 1.31,[10] 1.40,[11] 1.58,[13] 1.59,[14] 2.00[15])	–2.72 (–2.67[14])	2.351 (2.42,[10] 2.34,[11] 2.52,[13] 2.36,[14] 2.23[15])	–
Cu_5	1.73 (1.56±0.15,[9] 1.43,[10] 1.49,[11] 1.72,[13] 1.72,[14] 2.24[15])	–2.26 (–2.26[14])	2.360 (2.43,[10] 2.34,[11] 2.50,[13] 2.36,[14] 2.23[15])	0.06
Cu_6	1.92 (1.73±0.18,[9] 1.61,[10] 1.67,[11] 1.92,[12] 1.91,[13] 1.92[14])	–2.88 (–2.88[14])	2.361 (2.43,[10] 2.35,[11] 2.39,[12] 2.47,[13] 2.36[14])	–
Cu_7	2.04 (1.86±0.22,[9] 1.69,[10] 1.72,[11] 2.03,[13] 2.63[14])	–2.79 (–2.80[14])	2.422 (2.50,[10] 2.39,[11] 2.48,[13] 2.41[14])	0.04
Pd_2	0.67 (0.95[a], 0.67[b])	–1.34	2.472 (2.627-2.668,[16] 2.548-2.612[16])	0.60
Pd_3	1.28	–2.50	2.482	0.63
Pd_4	1.69 (2.22[a], 1.72[b], 1.45[17])	–2.92	2.595 (2.591,[18] 2.661[18])	0.48
Pd_5	1.82	–2.32	2.632	0.39
Pd_6	1.96	–2.67	2.651	0.33
Pd_7	1.99	–2.18	2.689	0.27
Pt_2	1.98 (1.95,[19] 1.86,[20] 1.63,[21] 1.76[22])	–3.96	2.322 (2.32,[19] 2.33,[20] 2.45,[21] 2.34,[22] 2.462-2.605,[16] 2.427-2.574[16])	0.89
Pt_3	2.53 (2.50,[19] 2.41,[20] 2.22,[21] 2.33[22])	–3.64	2.467 (2.48,[19] 2.49,[20] 2.57,[21] 2.47[22])	0.59
Pt_4	2.77 (3.43[a], 2.88[b], 2.06,[19] 2.70,[20] 2.62[22])	–3.49	2.489 (2.51,[19] 2.51,[20] 2.51-2.58[22])	–0.82
Pt_5	3.08 (3.80,[19] 2.96,[20] 2.89[22])	–4.33	2.463 (2.46,[19] 2.43-2.51[22])	0.37
Pt_6	3.33 (4.04[a], 3.41[b], 3.31,[19] 3.21,[20] 2.94,[21] 3.08[22])	–4.54	2.511 (2.54,[19] 2.49-2.61[22])	–0.12
Pt_7	3.40 (3.37[19], 3.26[20])	–3.87	2.523 (2.53[19])	0.38

[a]Calculated results from RPBE functional. [b]Calculated results under van der Waals dispersion correction (D3 method).

Table S2. Possible adsorption site, optimized adsorption site and adsorption energy (eV) for single Co, Ni, Cu, Pd, Pt atoms on Mo_2C(001), Mo_2C(101) and MoC(001).

Site	Optimized adsorption site					Adsorption energy (eV)				
	Co	Ni	Cu	Pd	Pt	Co	Ni	Cu	Pd	Pt
Mo_2C(001) surface										
t1	3h1	3h1	3h1	3h1	3h1	–4.78	–4.78	–3.46	–9.49	–11.70
t2	3h1	3h1	3h1	3h1	3h1	–4.78	–4.78	–3.46	–9.49	–11.70
b1	3h2	3h2	3h2	3h2	3h2	–4.53	–4.59	–3.41	–9.39	–11.52
b2	3h1	3h4	3h1	3h1	3h1	–4.78	–4.64	–3.46	–9.49	–11.70
b3	3h1	3h1	3h1	3h1	3h1	–4.78	–4.78	–3.46	–9.49	–11.70
b4	3h1	3h1	3h2	3h1	3h2	–4.78	–4.78	–3.41	–9.49	–11.52
3h1	3h1	3h1	3h1	3h1	3h1	–4.78	–4.78	–3.46	–9.49	–11.70
3h2	3h2	3h2	3h2	3h2	3h2	–4.53	–4.59	–3.41	–9.39	–11.52
3h3	3h3	3h3	3h3	3h3	3h3	–4.19	–4.30	–3.27	–9.03	–11.06
3h4	3h1	3h4	3h1	3h1	3h1	–4.78	–4.64	–3.46	–9.49	–11.70
Mo_2C(101) surface										
t1	b2	b2	b2	t1	t1	–5.70	–5.48	–3.72	–9.81	–11.50
t2	b2	b2	b2	b2	b2	–5.70	–5.48	–3.72	–10.78	–12.48
t3	3h1	3h1	3h1	3h1	3h1	–3.44	–3.55	–2.30	–9.77	–11.42
t4	b8	b8	b2	3h3	3h1	–3.31	–3.30	–3.72	–9.78	–11.42
b1	3h1	3h1	b2	3h1	3h1	–3.41	–3.55	–3.72	–9.78	–11.42
b2	b2	b2	b2	b2	b2	–5.70	–5.47	–3.71	–10.78	–12.48
b3	b3	b3	b2	3h3	3h3	–3.29	–3.53	–3.72	–9.78	–11.16
b4	b2	b2	b2	b2	t1	–5.70	–5.48	–3.72	–10.78	–11.50
b5	b2	b2	tilted b4	tilted t1	tilted t1	–5.70	–5.48	–2.58	–9.81	–11.50
b6	3h1	3h1	3h1	3h1	3h1	–3.41	–3.55	–2.30	–9.77	–11.42
b7	b2	b3	b7	3h3	3h3	–5.70	–3.49	–2.10	–9.78	–11.17
b8	b8	b8	b8	b8	3h1	–3.28	–3.26	–1.94	–9.41	–11.42
3h1	3h1	3h1	3h1	3h1	3h1	–3.41	–3.51	–2.26	–9.75	–11.42
3h2	b8	3h2	3h1	3h3	3h1	–3.31	–3.35	–2.30	–9.78	–11.42
3h3	b2	b3	b2	3h3	3h3	–5.70	–3.49	–3.72	–9.75	–11.17
4h1	b2	b2	b2	b2	b2	–5.70	–5.48	–3.72	–10.78	–12.48
4h2	b2	b2	b2	b2	b2	–5.70	–5.48	–3.72	–10.78	–12.48
MoC(001) surface										
t1	4h	4h	t1	t1	t1	–4.71	–4.52	–2.67	–8.95	–10.50
t2	4h	4h	t2	4h	4h	–4.71	–4.52	–1.82	–8.76	–10.31
b	4h	4h	tilted t1	t1	t1	–4.71	–4.52	–2.68	–8.95	–10.50
4h	4h	4h	4h	t1	4h	–4.71	–4.52	–2.76	–8.95	–10.24

Table S3. Adsorption energy [$E(Co_{n/ads})$; eV], average adsorption energy [$E(Co_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Co_{ads})$; eV], average aggregation energy [$E_{agg}(Co_{ads})$; eV], stepwise growth energy [$E_{growth}(Co_{ads})$; eV], coordination number (CN) with surface Mo atoms, number of Co-Co bond (NB-Co), average Co-Co distance (d, Å), and average Bader charge (q, e) for Co_n on Mo_2C(001).

Species	$E(Co_{n/ads})$	$E(Co_{ads/av})$	$\Delta E(Co_{ads})$	$E_{agg}(Co_{ads})$	$E_{growth}(Co_{ads})$	CN	NB-Co	d	q
1×Co	–4.78	–4.78	–4.78	–	–	3	0	–	–0.44
Aggregation mode									
2×Co	–9.53	–4.77	–4.75	0.02	0.03	6	1	2.336	–0.37
3×Co	–14.47	–4.82	–4.94	–0.04	–0.16	9	3	2.483	–0.32
4×Co	–19.62	–4.91	–5.15	–0.13	–0.37	12	4	2.459	–0.32
5×Co	–24.33	–4.87	–4.71	–0.09	0.07	15	7	2.515	–0.29
Mode a									
6×Co-a	–29.42	–4.90	–5.09	–0.12	–0.31	18	9	2.483	–0.27
7×Co-a	–34.79	–4.97	–5.37	–0.19	–0.59	21	10	2.486	–0.28
8×Co-a	–39.94	–4.99	–5.15	–0.21	–0.37	24	11	2.460	–0.29
9×Co-a	–45.02	–5.00	–5.08	–0.22	–0.30	26	13	2.456	–0.29
10×Co-a	–50.15	–5.02	–5.13	–0.23	–0.35	28	16	2.478	–0.27
11×Co-a	–55.69	–5.06	–5.54	–0.28	–0.76	29	20	2.486	–0.26
12×Co-a	–60.67	–5.06	–4.98	–0.28	–0.20	32	23	2.491	–0.25
Mode b									
6×Co-b	–29.41	–4.90	–5.08	–0.12	–0.30	18	8	2.481	–0.29
7×Co-b	–34.54	–4.93	–5.13	–0.15	–0.35	21	10	2.451	–0.29
8×Co-b	–39.77	–4.97	–5.23	–0.19	–0.45	22	13	2.481	–0.27
9×Co-b	–44.84	–4.98	–5.07	–0.20	–0.29	25	16	2.486	–0.26
10×Co-b	–50.33	–5.03	–5.49	–0.25	–0.71	28	20	2.485	–0.24
11×Co-b	–55.43	–5.04	–5.10	–0.26	–0.32	31	22	2.492	–0.24
12×Co-b	–60.67	–5.06	–5.24	–0.28	–0.46	34	24	2.485	–0.23
Aggregation mode									
13×Co	–66.07	–5.08	–5.40	–0.30	–0.62	35	27	2.491	–0.24
14×Co	–71.22	–5.09	–5.15	–0.31	–0.37	40	30	2.482	–0.23
15×Co	–76.55	–5.10	–5.33	–0.32	–0.55	43	34	2.470	–0.22
25×Co	–131.64	–5.27		–0.49		55	75	2.468	–0.17
Dispersion mode c									
2×Co-c	–9.70	–4.85	–4.92	–0.07	–0.14	6	0	–	–0.42
3×Co-c	–14.47	–4.82	–4.77	–0.04	0.01	9	0	–	–0.42
4×Co-c	–19.31	–4.83	–4.84	–0.05	–0.06	12	0	–	–0.43
5×Co-c	–24.01	–4.80	–4.70	–0.02	0.08	15	0	–	–0.39
Three-dimensional mode d									
4×Co-d	–17.72	–4.43	–3.25	0.35	1.53	9	6	2.357	–0.24
5×Co-d	–23.06	–4.61	–5.34	0.17	–0.56	12	8	2.382	–0.24
6×Co-d	–28.20	–4.70	–5.14	0.08	–0.36	15	10	2.395	–0.24
7×Co-d	–32.97	–4.71	–4.77	0.07	0.01	18	11	2.416	–0.25

Table S4. Adsorption energy [$E(Ni_{n/ads})$; eV], average adsorption energy [$E(Ni_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Ni_{ads})$; eV], average aggregation energy [$E_{agg}(Ni_{ads})$; eV], stepwise growth energy [$E_{growth}(Ni_{ads})$; eV], coordination number (CN) with surface Mo atoms, number of Ni-Ni bond (NB-Ni), average Ni-Ni distance (d, Å), and average Bader charge (q, e) for Ni_n on $Mo_2C(001)$.

Species	$E(Ni_{n/ads})$	$E(Ni_{ads/av})$	$\Delta E(Ni_{ads})$	$E_{agg}(Ni_{ads})$	$E_{growth}(Ni_{ads})$	CN	NB-Ni	d	q
1×Ni	–4.78	–4.78	–4.78	–	–	3	0	–	–0.44
Aggregation mode									
2×Ni	–9.48	–4.74	–4.70	0.04	0.08	6	1	2.396	–0.36
3×Ni	–14.58	–4.86	–5.10	–0.08	–0.32	9	3	2.550	–0.33
Mode a									
4×Ni-a	–19.33	–4.83	–4.75	–0.05	0.03	12	4	2.488	–0.31
5×Ni-a	–24.41	–4.88	–5.08	–0.10	–0.30	14	5	2.501	–0.32
6×Ni-a	–29.49	–4.92	–5.08	–0.14	–0.30	17	7	2.486	–0.31
7×Ni-a	–34.43	–4.92	–4.94	–0.14	–0.16	21	10	2.498	–0.29
8×Ni-a	–39.45	–4.93	–5.02	–0.15	–0.24	23	13	2.499	–0.27
9×Ni-a	–44.60	–4.96	–5.15	–0.18	–0.37	26	16	2.518	–0.26
10×Ni-a	–49.89	–4.99	–5.29	–0.21	–0.51	30	20	2.522	–0.25
11×Ni-a	–54.84	–4.99	–4.95	–0.21	–0.17	32	22	2.520	–0.24
12×Ni-a	–59.99	–5.00	–5.15	–0.22	–0.37	34	25	2.526	–0.23
13×Ni-a	–65.24	–5.02	–5.25	–0.24	–0.47	37	27	2.526	–0.24
14×Ni-a	–70.46	–5.03	–5.22	–0.25	–0.44	39	30	2.519	–0.23
Mode b									
4×Ni-b	–19.32	–4.83	–4.74	–0.05	0.04	12	5	2.640	–0.30
5×Ni-b	–24.32	–4.86	–5.00	–0.08	–0.22	15	7	2.613	–0.29
6×Ni-b	–29.26	–4.88	–4.94	–0.10	–0.16	18	9	2.562	–0.27
7×Ni-b	–34.43	–4.92	–5.17	–0.14	–0.39	20	10	2.545	–0.28
8×Ni-b	–39.58	–4.95	–5.15	–0.17	–0.37	22	11	2.503	–0.29
9×Ni-b	–44.64	–4.96	–5.06	–0.18	–0.28	25	13	2.485	–0.28
10×Ni-b	–49.68	–4.97	–5.04	–0.19	–0.26	28	15	2.481	–0.27
11×Ni-b	–54.84	–4.99	–5.16	–0.21	–0.38	31	20	2.518	–0.25
12×Ni-b	–59.99	–5.00	–5.15	–0.22	–0.37	34	23	2.516	–0.24
13×Ni-b	–65.06	–5.00	–5.07	–0.22	–0.29	36	25	2.514	–0.23
14×Ni-b	–70.46	–5.03	–5.40	–0.25	–0.62	38	30	2.520	–0.22
Aggregation mode									
15×Ni	–75.87	–5.06	–5.41	–0.28	–0.63	42	34	2.515	–0.22
25×Ni	–127.64	–5.11		–0.33		54	75	2.438	–0.17
Dispersion mode c									
2×Ni-c	–9.64	–4.82	–4.86	–0.04	–0.08	6	0	–	–0.44
3×Ni-c	–14.50	–4.83	–4.86	–0.05	–0.08	9	0	–	–0.43
4×Ni-c	–19.34	–4.84	–4.84	–0.05	–0.06	12	0	–	–0.43
5×Ni-c	–24.14	–4.83	–4.80	–0.05	–0.02	15	1	2.893	–0.38
Three-dimensional mode d									
4×Ni-d	–17.76	–4.44	–3.26	0.34	1.52	9	6	2.437	–0.25
5×Ni-d	–22.80	–4.56	–5.04	0.22	–0.26	12	8	2.512	–0.25
6×Ni-d	–27.84	–4.64	–5.04	0.14	–0.26	15	10	2.457	–0.24
7×Ni-d	–32.89	–4.70	–5.05	0.08	–0.27	18	12	2.454	–0.23

Table S5. Adsorption energy [$E(Cu_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Cu_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Cu_{ads})$; eV], average aggregation energy [$E_{agg}(Cu_{ads})$; eV], stepwise growth energy [$E_{growth}(Cu_{ads})$; eV], coordination number (CN) with surface Mo atoms, number of Cu-Cu bond (NB-Cu), average Cu-Cu distance (*d*, Å), and average Bader charge (q, e) for Cu_n on $Mo_2C(001)$.

Species	$E(Cu_{n/ads})$	$E(Cu_{ads/av})$	$\Delta E(Cu_{ads})$	$E_{agg}(Cu_{ads})$	$E_{growth}(Cu_{ads})$	CN	NB-Cu	*d*	q
1×Cu	–3.46	–3.46	–3.46	–	–	3	0	–	–0.32
Aggregation mode									
2×Cu	–7.07	–3.54	–3.61	–0.08	–0.15	6	1	2.551	–0.26
3×Cu	–10.83	–3.61	–3.76	–0.15	–0.30	9	3	2.569	–0.21
Mode a									
4×Cu-a	–14.49 (–10.91)	–3.62	–3.66	–0.16	–0.20	12	5	2.644	–0.20
5×Cu-a	–18.29	–3.66	–3.8	–0.20	–0.34	15	7	2.696	–0.21
6×Cu-a	–22.02 (–19.74)	–3.67	–3.73	–0.21	–0.27	18	9	2.705	–0.20
7×Cu-a	–25.88	–3.70	–3.86	–0.24	–0.40	21	11	2.754	–0.19
8×Cu-a	–29.80 (–28.25)	–3.73	–3.92	–0.27	–0.46	24	14	2.707	–0.18
Mode b									
4×Cu-b	–14.45	–3.61	–3.62	–0.15	–0.16	12	5	2.679	–0.20
5×Cu-b	–18.12	–3.62	–3.67	–0.16	–0.21	15	7	2.709	–0.20
6×Cu-b	–21.94	–3.66	–3.82	–0.20	–0.36	18	9	2.727	–0.20
7×Cu-b	–25.67	–3.67	–3.73	–0.21	–0.27	21	11	2.733	–0.20
8×Cu-b	–29.52	–3.69	–3.85	–0.23	–0.39	24	13	2.773	–0.19
Aggregation mode									
9×Cu	–33.44	–3.72	–3.64	–0.26	–0.18	27	16	2.777	–0.18
10×Cu	–37.19	–3.72	–3.75	–0.26	–0.29	30	19	2.769	–0.17
11×Cu	–40.96	–3.72	–3.77	–0.26	–0.31	33	21	2.776	–0.17
12×Cu	–44.67	–3.72	–3.71	–0.26	–0.25	36	25	2.795	–0.16
25×Cu	–93.07	–3.72		–0.26		53	76	2.446	–0.12
Dispersion mode c									
2×Cu-c	–6.98 (–2.52)	–3.49	–3.52	–0.03	–0.06	6	0	–	–0.33
3×Cu-c	–10.67	–3.56	–3.69	–0.10	–0.23	9	0	–	–0.32
4×Cu-c	–13.95 (–10.38)	–3.49	–3.28	–0.03	0.18	12	0	–	–0.30
5×Cu-c	–17.69	–3.54	–3.74	–0.08	–0.28	15	2	2.895	–0.29
Three-dimensional mode d									
4×Cu-d	–13.21 (–9.42)	–3.30	–2.54	0.16	0.92	9	6	2.555	–0.16
5×Cu-d	–17.05	–3.41	–3.84	0.05	–0.38	12	9	2.589	–0.16
6×Cu-d	–20.85 (–18.01)	–3.48	–3.80	–0.02	–0.34	15	10	2.569	–0.16
7×Cu-d	–24.63	–3.52	–3.78	–0.06	–0.32	18	12	2.600	–0.17

Table S6. Adsorption energy [$E(Pd_{n/ads})$; eV], average adsorption energy [$E(Pd_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pd_{ads})$; eV], average aggregation energy [$E_{agg}(Pd_{ads})$; eV], stepwise growth energy [$E_{growth}(Pd_{ads})$; eV], coordination number (CN) with surface Mo atoms, number of Pd-Pd bond (NB-Pd), average Pd-Pd distance (d, Å), and average Bader charge (q, e) for Pd_n on $Mo_2C(001)$.

Species	$E(Pd_{n/ads})$	$E(Pd_{ads/av})$	$\Delta E(Pd_{ads})$	$E_{agg}(Pd_{ads})$	$E_{growth}(Pd_{ads})$	CN	NB-Pd	d	q
1×Pd	–9.49	–9.49	–9.49	–	–	3	0	–	–0.57
Aggregation mode									
2×Pd	–13.73	–6.87	–4.24	2.63	5.25	6	1	2.987	–0.50
3×Pd	–18.35	–6.12	–4.62	3.37	4.87	9	2	2.992	–0.50
4×Pd	–22.76	–5.69	–4.41	3.80	5.08	12	4	3.057	–0.46
5×Pd	–26.94	–5.39	–4.18	4.10	5.31	15	6	3.027	–0.44
Mode a									
6×Pd-a	–31.38	–5.23	–4.44	4.26	5.05	18	9	2.998	–0.42
7×Pd-a	–35.75	–5.11	–4.37	4.38	5.12	21	12	2.979	–0.40
8×Pd-a	–40.37	–5.05	–4.62	4.44	4.87	24	16	2.991	–0.38
9×Pd-a	–44.90	–4.99	–4.53	4.50	4.96	27	18	3.004	–0.39
10×Pd-a	–49.44	–4.94	–4.54	4.55	4.95	30	20	3.009	–0.40
11×Pd-a	–54.11	–4.92	–4.67	4.57	4.82	33	24	2.983	–0.37
12×Pd-a	–58.83	–4.90	–4.72	4.59	4.77	36	28	2.975	–0.36
13×Pd-a	–63.40	–4.88	–4.57	4.61	4.92	39	32	3.005	–0.35
14×Pd-a	–68.10	–4.86	–4.70	4.63	4.79	42	37	3.018	–0.34
15×Pd-a	–72.77	–4.85	–4.67	4.64	4.82	45	42	3.032	–0.33
Mode b									
6×Pd-b	–31.29	–5.22	–4.35	4.28	5.14	18	9	2.991	–0.42
7×Pd-b	–35.83	–5.12	–4.54	4.37	4.95	21	10	3.013	–0.43
8×Pd-b	–40.44	–5.06	–4.61	4.44	4.88	24	11	3.023	–0.43
9×Pd-b	–45.03	–5.00	–4.59	4.49	4.90	27	16	2.964	–0.41
10×Pd-b	–49.57	–4.96	–4.54	4.53	4.95	30	20	2.988	–0.39
11×Pd-b	–54.25	–4.93	–4.68	4.56	4.81	33	25	2.958	–0.37
12×Pd-b	–58.78	–4.90	–4.53	4.59	4.96	36	28	3.006	–0.36
13×Pd-b	–63.49	–4.88	–4.71	4.61	4.78	39	33	3.017	–0.35
14×Pd-b	–68.10	–4.86	–4.61	4.63	4.88	42	37	3.018	–0.34
15×Pd-b	–72.77	–4.85	–4.67	4.64	4.82	45	42	3.032	–0.33
Aggregation mode									
16×Pd	–77.61	–4.85	–4.84	4.64	4.65	48	48	3.021	–0.32
Dispersion mode c									
2×Pd-c	–13.96	–6.98	–4.47	2.51	5.02	6	0	–	–0.56
3×Pd-c	–18.36	–6.12	–4.40	3.37	5.09	9	0	–	–0.58
4×Pd-c	–22.86	–5.72	–4.50	3.78	4.99	12	0	–	–0.57
5×Pd-c	–27.26	–5.45	–4.40	4.04	5.09	15	2	3.009	–0.53
6×Pd-c	–31.69	–5.28	–4.43	4.21	5.06	18	4	3.060	–0.49
7×Pd-c	–36.00	–5.14	–4.31	4.35	5.18	21	6	3.043	–0.47
8×Pd-c	–40.35	–5.04	–4.35	4.45	5.14	24	8	3.061	–0.45
Three-dimensional mode d									
4×Pd-d	–20.63	–5.16	–2.28	4.33	7.21	9	6	2.754	–0.34
5×Pd-d	–24.83	–4.97	–4.20	4.52	5.29	12	8	2.838	–0.33
6×Pd-d	–29.33	–4.89	–4.50	4.60	4.99	15	10	2.824	–0.33
7×Pd-d	–33.76	–4.82	–4.43	4.67	5.06	18	12	2.836	–0.34

Table S7. Adsorption energy [$E(Pt_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Pt_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pt_{ads})$; eV], average aggregation energy [$E_{agg}(Pt_{ads})$; eV], stepwise growth energy [$E_{growth}(Pt_{ads})$; eV], coordination number (CN) with surface Mo atoms, number of Pt-Pt bond (NB-Pt), average Pt-Pt distance (d, Å), and average Bader charge (q, e) for Pt_n on $Mo_2C(001)$.

Species	$E(Pt_{n/ads})$	$E(Pt_{ads/av})$	$\Delta E(Pt_{ads})$	$E_{agg}(Pt_{ads})$	$E_{growth}(Pt_{ads})$	CN	NB-Pt	d	q
1×Pt	–11.70	–11.70	–11.70	–	–	3	0	–	–0.79
Aggregation mode									
2×Pt	–17.95 (–13.94)	–8.98	–6.25	2.73	5.45	6	1	3.006	–0.70
3×Pt	–24.71	–8.24	–6.76	3.46	4.94	9	2	2.973	–0.67
4×Pt	–31.07 (–28.27)	–7.77	–6.36	3.93	5.34	12	4	3.050	–0.62
5×Pt	–37.17	–7.43	–6.10	4.27	5.60	15	6	3.027	–0.59
6×Pt	–43.51 (–42.00)	–7.25	–6.34	4.45	5.36	18	9	2.999	–0.57
7×Pt	–49.77	–7.11	–6.26	4.59	5.44	21	12	2.640	–0.53
Mode a									
8×Pt-a	–56.30	–7.04	–6.53	4.66	5.17	24	14	2.974	–0.54
9×Pt-a	–62.80	–6.98	–6.50	4.72	5.20	27	16	2.961	–0.52
10×Pt-a	–69.35	–6.94	–6.55	4.77	5.15	30	19	3.000	–0.53
11×Pt-a	–75.96	–6.91	–6.61	4.79	5.09	33	22	2.943	–0.50
12×Pt-a	–82.64	–6.89	–6.68	4.81	5.02	36	26	2.927	–0.47
13×Pt-a	–89.15	–6.86	–6.51	4.84	5.19	39	28	2.911	–0.46
14×Pt-a	–95.78	–6.84	–6.63	4.86	5.07	42	33	2.887	–0.43
15×Pt-a	–102.10	–6.81	–6.32	4.89	5.38	45	41	3.006	–0.43
16×Pt-a	–108.75	–6.80	–6.65	4.90	5.05	48	45	2.998	–0.41
Mode b									
8×Pt-b	–56.24	–7.03	–6.47	4.67	5.23	24	16	2.990	–0.52
9×Pt-b	–62.31	–6.92	–6.07	4.78	5.63	27	18	2.920	–0.49
10×Pt-b	–69.23	–6.92	–6.92	4.78	4.78	30	18	2.915	–0.50
11×Pt-b	–75.81	–6.89	–6.58	4.81	5.12	33	20	2.926	–0.50
12×Pt-b	–82.47	–6.87	–6.66	4.83	5.04	36	25	2.944	–0.47
13×Pt-b	–89.13	–6.86	–6.66	4.84	5.04	39	29	2.949	–0.45
14×Pt-b	–95.78	–6.84	–6.65	4.86	5.05	42	32	2.921	–0.43
15×Pt-b	–102.16	–6.81	–6.38	4.89	5.32	45	36	2.947	–0.42
16×Pt-b	–108.72	–6.80	–6.56	4.91	5.14	48	45	2.996	–0.41
Dispersion mode c									
2×Pt-c	–18.35	–9.18	–6.65	2.53	5.05	6	0	–	–0.75
3×Pt-c	–25.00	–8.33	–6.65	3.37	5.05	9	0	–	–0.76
4×Pt-c	–31.69 (–28.87)	–7.92	–6.69	3.78	5.01	12	0	–	–0.76
5×Pt-c	–38.01	–7.60	–6.32	4.10	5.38	15	2	2.997	–0.71
6×Pt-c	–44.38	–7.40	–6.37	4.30	5.33	18	4	3.054	–0.66
7×Pt-c	–50.59	–7.23	–6.21	4.47	5.49	21	6	3.033	–0.63
8×Pt-c	–56.84	–7.11	–6.25	4.60	5.45	24	8	3.055	–0.60
Three-dimensional mode d									
4×Pt-d	–28.91 (–25.76)	–7.23	–4.38	4.47	7.32	9	6	2.728	–0.49
5×Pt-d	–34.89	–6.98	–5.98	4.72	5.72	12	8	2.889	–0.49
6×Pt-d	–41.41 (–39.61)	–6.90	–6.52	4.80	5.18	15	10	2.848	–0.50
7×Pt-d	–47.94	–6.85	–6.53	4.85	5.17	18	12	2.804	–0.48

Table S8. Adsorption energy [$E(Co_{n/ads})$; eV], average adsorption energy [$E(Co_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Co_{ads})$; eV], average aggregation energy [$E_{agg}(Co_{ads})$; eV], stepwise growth energy [$E_{growth}(Co_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Co-Co bond (NB-Co), average Co-Co distance (d, Å), and average Bader charge (q, e) for Co_n on $Mo_2C(101)$.

Species	$E(Co_{n/ads})$	$E(Co_{ads/av})$	$\Delta E(Co_{ads})$	$E_{agg}(Co_{ads})$	$E_{growth}(Co_{ads})$	CN-Mo	CN-C	NB-Co	d	q
1×Co	–5.70	–5.70	–5.70	–	–	2	2	0	–	0.20
Dispersion mode a										
2×Co-a	–11.44	–5.72	–5.74	–0.02	–0.04	4	4	0	–	0.20
3×Co-a	–17.13	–5.71	–5.69	–0.01	0.01	6	6	0	–	0.20
4×Co-a	–22.88	–5.72	–5.75	–0.02	–0.05	8	8	0	–	0.21
5×Co-a	–28.33	–5.67	–5.45	0.03	0.25	10	10	0	–	0.21
6×Co-a	–33.79	–5.63	–5.46	0.07	0.24	12	12	0	–	0.21
7×Co-a	–39.24	–5.61	–5.45	0.09	0.25	14	14	0	–	0.21
8×Co-a	–44.69	–5.59	–5.45	0.11	0.25	16	16	0	–	0.21
Aggregation mode										
9×Co	–48.66	–5.41	–3.97	0.29	1.73	20	17	1	2.375	0.19
10×Co	–53.30	–5.33	–4.64	0.37	1.06	20	18	3	2.388	0.18
Mode a										
11×Co-a	–57.70	–5.25	–4.40	0.45	1.30	20	19	5	2.431	0.18
12×Co-a	–61.76	–5.15	–4.06	0.55	1.64	24	19	8	2.466	0.15
13×Co-a	–67.18	–5.17	–5.42	0.53	0.28	24	19	12	2.500	0.14
14×Co-a	–71.57	–5.11	–4.39	0.59	1.31	28	19	15	2.483	0.12
15×Co-a	–76.09	–5.07	–4.52	0.63	1.18	27	20	17	2.496	0.12
16×Co-a	–81.27	–5.08	–5.18	0.62	0.52	31	20	21	2.498	0.10
17×Co-a	–86.50	–5.09	–5.23	0.61	0.47	31	20	25	2.491	0.09
18×Co-a	–91.75	–5.10	–5.25	0.60	0.45	35	21	29	2.464	0.09
28×Co-a	–136.97	–4.89		0.81		60	26	49	2.471	0.06
Mode b										
11×Co-b	–57.19	–5.20	–3.89	0.50	1.81	14	17	5	2.387	0.16
12×Co-b	–62.21	–5.18	–5.02	0.52	0.68	15	18	8	2.366	0.14
13×Co-b	–66.46	–5.11	–4.25	0.59	1.45	17	19	10	2.411	0.13
14×Co-b	–70.88	–5.06	–4.42	0.64	1.28	20	19	13	2.414	0.11
15×Co-b	–76.38	–5.09	–5.50	0.61	0.20	25	20	14	2.418	0.11
16×Co-b	–81.37	–5.09	–4.99	0.61	0.71	24	21	21	2.483	0.11
17×Co-b	–86.40	–5.08	–5.03	0.62	0.67	25	21	25	2.472	0.10
18×Co-b	–91.75	–5.10	–5.35	0.60	0.35	25	21	28	2.467	0.09
28×Co-b	–136.89	–4.89		0.81		63	25	50	2.491	0.06
Aggregation mode c										
2×Co-c	–9.60	–4.80	–3.90	0.90	1.80	6	3	1	2.365	0.11
3×Co-c	–13.66	–4.55	–4.06	1.15	1.64	6	4	2	2.350	0.09
4×Co-c	–19.79	–4.95	–6.13	0.75	–0.43	8	6	3	2.594	0.13

Table S9. Adsorption energy [$E(Ni_{n/ads})$; eV], average adsorption energy [$E(Ni_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Ni_{ads})$; eV], average aggregation energy [$E_{agg}(Ni_{ads})$; eV], stepwise growth energy [$E_{growth}(Ni_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Ni-Ni bond (NB-Ni), average Ni-Ni distance (d, Å), and average Bader charge (q, e) for Ni_n on $Mo_2C(101)$.

Species	$E(Ni_{n/ads})$	$E(Ni_{ads/av})$	$\Delta E(Ni_{ads})$	$E_{agg}(Ni_{ads})$	$E_{growth}(Ni_{ads})$	CN-Mo	CN-C	NB-Ni	d	q
1×Ni	–5.48	–5.48	–5.48	–	–	2	2	0	–	0.18
Dispersion mode a										
2×Ni-a	–10.95	–5.48	–5.47	0.01	0.01	4	4	0	–	0.18
3×Ni-a	–16.38	–5.46	–5.43	–0.02	0.05	6	6	0	–	0.18
4×Ni-a	–21.83	–5.46	–5.45	0.02	0.03	8	8	0	–	0.18
5×Ni-a	–27.17	–5.43	–5.34	0.05	0.14	10	10	0	–	0.18
6×Ni-a	–32.55	–5.43	–5.38	0.06	0.10	12	12	0	–	0.18
7×Ni-a	–37.89	–5.41	–5.34	0.07	0.14	14	14	0	–	0.18
8×Ni-a	–43.26	–5.41	–5.37	0.07	0.11	16	16	0	–	0.17
Aggregation mode										
9×Ni	–47.22	–5.25	–3.96	0.23	1.52	18	17	1	2.438	0.16
10×Ni	–51.54	–5.15	–4.32	0.33	1.16	20	18	3	2.490	0.16
Mode a										
11×Ni-a	–55.80	–5.07	–4.26	0.41	1.22	24	19	5	2.533	0.15
12×Ni-a	–60.06	–5.01	–4.26	0.48	1.22	25	19	8	2.556	0.13
Mode b										
11×Ni-b	–55.75	–5.07	–4.21	0.41	1.27	22	18	6	2.563	0.13
12×Ni-b	–60.13	–5.01	–4.38	0.47	1.10	23	18	8	2.582	0.12
Aggregation mode										
13×Ni	–64.76	–4.98	–4.70	0.50	0.78	26	19	12	2.580	0.11
14×Ni	–69.15	–4.94	–4.39	0.54	1.09	30	19	15	2.555	0.10
15×Ni	–73.71	–4.91	–4.56	0.57	0.92	27	20	19	2.606	0.10
16×Ni	–78.18	–4.89	–4.47	0.59	1.01	31	20	22	2.587	0.08
17×Ni	–83.19	–4.89	–5.01	0.59	0.47	33	20	26	2.554	0.07
18×Ni	–87.94	–4.89	–4.75	0.59	0.73	35	21	30	2.518	0.07
28×Ni	–131.74	–4.71		0.78		60	36	47	2.497	0.03
Aggregation mode c										
2×Ni-c	–9.38	–4.69	–3.90	0.79	1.58	4	3	1	2.446	0.10
3×Ni-c	–13.27	–4.42	–3.89	1.06	1.59	6	4	2	2.449	0.08
4×Ni-c	–19.14	–4.79	–5.87	0.70	–0.39	9	6	3	2.474	0.11

Table S10. Adsorption energy [$E(Cu_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Cu_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Cu_{ads})$; eV], average aggregation energy [$E_{agg}(Cu_{ads})$; eV], stepwise growth energy [$E_{growth}(Cu_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Cu-Cu bond (NB-Cu), average Cu-Cu distance (d, Å), and average Bader charge (q, e) for Cu_n on Mo_2C(101).

Species	$E(Cu_{n/ads})$	$E(Cu_{ads/av})$	$\Delta E(Cu_{ads})$	$E_{agg}(Cu_{ads})$	$E_{growth}(Cu_{ads})$	CN-Mo	CN-C	NB-Cu	d	q
1×Cu	–3.72	–3.72	–3.72	–	–	2	2	0	–	0.26
Dispersion mode a										
2×Cu-a	–7.43	–3.72	–3.71	0.01	0.01	4	4	0	–	0.26
3×Cu-a	–11.09	–3.70	–3.66	0.02	0.06	6	6	0	–	0.27
4×Cu-a	–14.76 (–9.82)	–3.69	–3.67	0.03	0.05	8	8	0	–	0.27
5×Cu-a	–18.38	–3.68	–3.62	0.04	0.10	10	10	0	–	0.26
6×Cu-a	–22.01	–3.67	–3.63	0.05	0.09	12	12	0	–	0.26
7×Cu-a	–25.59	–3.66	–3.58	0.06	0.14	14	14	0	–	0.25
8×Cu-a	–29.18(–26.34)	–3.65	–3.59	0.07	0.13	16	16	0	–	0.25
Aggregation mode a										
9×Cu-a	–31.79	–3.53	–2.61	0.19	1.11	18	17	1	2.444	0.24
10×Cu-a	–34.90	–3.49	–3.11	0.23	0.61	20	17	3	2.471	0.20
11×Cu-a	–38.35	–3.49	–3.45	0.23	0.27	21	18	7	2.618	0.20
12×Cu-a	–41.63	–3.47	–3.28	0.25	0.44	22	18	10	2.615	0.17
13×Cu-a	–45.08	–3.47	–3.45	0.25	0.27	24	19	12	2.597	0.17
Mode b										
9×Cu-b	–31.82	–3.54	–2.64	0.18	1.08	18	16	1	2.443	0.22
10×Cu-b	–34.88	–3.49	–3.06	0.23	0.66	20	16	3	2.496	0.19
11×Cu-b	–38.01	–3.46	–3.13	0.26	0.59	21	16	9	2.611	0.18
12×Cu-b	–41.63	–3.47	–3.62	0.25	0.10	23	18	8	2.501	0.17
13×Cu-b	–45.08	–3.47	–3.45	0.25	0.14	23	19	14	2.625	0.17
Mode c										
9×Cu-c	–31.90(–29.44)	–3.54	–2.72	0.18	1.00	17	16	2	2.456	0.22
10×Cu-c	–34.82	–3.48	–2.92	0.24	0.80	18	16	5	2.507	0.20
11×Cu-c	–38.17	–3.47	–3.35	0.25	0.37	20	17	7	2.582	0.18
12×Cu-c	–41.63	–3.47	–3.46	0.25	0.26	22	18	11	2.630	0.17
Aggregation mode										
14×Cu	–48.27	–3.45	–3.19	0.27	0.53	24	19	15	2.588	0.15
15×Cu	–51.74	–3.45	–3.47	0.27	0.25	26	20	19	2.628	0.14
16×Cu	–55.02	–3.44	–3.28	0.28	0.44	27	20	22	2.621	0.13
17×Cu	–58.66	–3.45	–3.64	0.27	0.08	27	20	26	2.650	0.12
18×Cu	–62.17	–3.45	–3.51	0.27	0.21	28	18	32	2.553	0.10
28×Cu	–94.53	–3.38		0.34		35	22	54	2.584	0.05
Aggregated mode d										
2×Cu-d	–6.40 (–0.34)	–3.20	–2.68	0.52	1.04	4	2	1	2.459	0.12
3×Cu-d	–9.12	–3.04	–2.72	0.68	1.00	6	3	2	2.542	0.09
4×Cu-d	–13.15 (–8.04)	–3.29	–4.03	0.43	-0.31	8	5	3	2.408	0.11

Table S11. Adsorption energy [$E(Pd_{n/ads})$; eV], average adsorption energy [$E(Pd_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pd_{ads})$; eV], average aggregation energy [$E_{agg}(Pd_{ads})$; eV], stepwise growth energy [$E_{growth}(Pd_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Pd-Pd bond (NB-Pd), average Pd-Pd distance (d, Å), and average Bader charge (q, e) for Pd_n on $Mo_2C(101)$.

Species	$E(Pd_{n/ads})$	$E(Pd_{ads/av})$	$\Delta E(Pd_{ads})$	$E_{agg}(Pd_{ads})$	$E_{growth}(Pd_{ads})$	CN-Mo	CN-C	NB-Pd	d	q
1×Pd	–10.78	–10.78	–10.78	–	–	2	2	0	–	–0.01
Dispersion mode a										
2×Pd-a	–14.81	–7.41	–4.03	3.38	6.75	4	4	0	–	–0.03
3×Pd-a	–18.83	–6.28	–4.02	4.50	6.76	6	6	0	–	–0.04
4×Pd-a	–22.89	–5.72	–4.06	5.06	6.72	8	8	0	–	–0.05
5×Pd-a	–26.82	–5.36	–3.93	5.42	6.85	10	10	0	–	–0.04
6×Pd-a	–30.81	–5.14	–3.99	5.65	6.79	12	12	0	–	–0.04
7×Pd-a	–34.80	–4.97	–3.99	5.81	6.79	14	14	0	–	–0.04
8×Pd-a	–38.81	–4.85	–4.01	5.93	6.77	16	16	0	–	–0.04
Aggregation mode a										
9×Pd-a	–42.22	–4.69	–3.41	6.09	7.37	18	17	1	2.763	–0.05
10×Pd-a	–45.87	–4.59	–3.65	6.19	7.13	20	18	3	2.815	–0.04
11×Pd-a	–49.62	–4.51	–3.75	6.27	7.03	15	18	7	2.824	–0.05
12×Pd-a	–53.52	–4.46	–3.90	6.32	6.88	15	18	11	2.814	–0.05
Mode b										
9×Pd-b	–42.10	–4.68	–3.29	6.10	7.49	18	16	1	2.931	–0.04
10×Pd-b	–45.66	–4.57	–3.56	6.21	7.22	20	17	3	2.700	–0.06
11×Pd-b	–49.62	–4.51	–3.96	6.27	6.82	15	18	7	2.825	–0.05
Mode c										
9×Pd-c	–42.08	–4.68	–3.27	6.10	7.51	17	16	2	2.703	–0.05
10×Pd-c	–45.70	–4.57	–3.62	6.21	7.16	18	16	5	2.751	–0.06
11×Pd-c	–49.62	–4.51	–3.92	6.27	6.86	18	17	8	2.436	–0.06
12×Pd-c	–53.52	–4.46	–3.90	6.32	6.88	15	18	11	2.813	–0.06
Aggregation mode										
13×Pd	–57.44	–4.42	–3.92	6.36	6.86	16	19	14	2.822	–0.05
14×Pd	–61.35	–4.38	–3.91	6.40	6.87	17	19	18	2.823	–0.06
15×Pd	–65.43	–4.36	–4.08	6.42	6.70	23	20	19	2.840	–0.06
16×Pd	–69.73	–4.36	–4.30	6.42	6.48	28	20	20	2.831	–0.06
24×Pd	–100.22	–4.18		6.60		40	24	40	2.866	–0.08
Aggregation mode d										
2×Pd-d	–14.09	–7.05	–4.60	3.74	6.18	4	3	1	2.889	–0.07
3×Pd-d	–17.33	–5.78	–3.24	5.00	7.54	6	4	2	2.846	–0.07
4×Pd-d	–21.64	–5.41	–4.31	5.37	6.47	8	6	3	2.788	–0.06

Table S12. Adsorption energy [$E(Pt_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Pt_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pt_{ads})$; eV], average aggregation energy [$E_{agg}(Pt_{ads})$; eV], stepwise growth energy [$E_{growth}(Pt_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Pt-Pt bond (NB-Pt), average Pt-Pt distance (*d*, Å), and average Bader charge (q, e) for Pt_n on $Mo_2C(101)$.

Species	$E(Pt_{n/ads})$	$E(Pt_{ads/av})$	$\Delta E(Pt_{ads})$	$E_{agg}(Pt_{ads})$	$E_{growth}(Pt_{ads})$	CN-Mo	CN-C	NB-Pt	*d*	q
1×Pt	–12.48	–12.48	–12.48	–	–	2	2	0	–	–0.25
Dispersion mode a										
2×Pt-a	–18.24(–12.75)	–9.12	–5.76	3.36	6.72	4	4	0	–	–0.25
3×Pt-a	–23.94	–7.98	–5.70	4.50	6.78	6	6	0	–	–0.24
4×Pt-a	–29.68(–25.57)	–7.42	–5.74	5.06	6.74	8	8	0	–	–0.23
5×Pt-a	–35.26	–7.05	–5.58	5.43	6.90	10	10	0	–	–0.23
6×Pt-a	–40.90	–6.82	–5.64	5.66	6.84	12	12	0	–	–0.22
7×Pt-a	–46.50	–6.64	–5.60	5.84	6.88	14	14	0	–	–0.21
8×Pt-a	–52.06	–6.51	–5.56	5.97	6.92	16	16	0	–	–0.21
Aggregation mode a										
9×Pt-a	–57.06	–6.34	–5.00	6.14	7.48	18	17	1	2.672	–0.21
10×Pt-a	–62.31	–6.23	–5.25	6.25	7.23	20	17	3	2.686	–0.21
11×Pt-a	–67.54	–6.14	–5.23	6.34	7.25	22	18	6	2.769	–0.20
12×Pt-a	–73.43	–6.12	–5.89	6.36	6.59	23	18	11	2.747	–0.17
13×Pt-a	–79.31	–6.10	–5.88	6.38	6.60	25	18	16	2.767	–0.16
Mode b										
9×Pt-b	–56.88	–6.32	–4.82	6.16	7.66	18	16	1	2.659	–0.21
10×Pt-b	–62.31	–6.23	–5.43	6.25	7.05	20	16	3	2.686	–0.21
Mode c										
9×Pt-c	–57.16(–56.36)	–6.35	–5.10	6.13	7.38	16	16	2	2.614	–0.20
10×Pt-c	–62.51	–6.25	–5.35	6.23	7.13	20	16	6	2.736	–0.19
11×Pt-c	–67.97	–6.18	–5.46	6.30	7.02	22	17	8	2.734	–0.18
12×Pt-c	–73.56	–6.13	–5.59	6.35	6.89	24	17	12	2.755	–0.18
13×Pt-c	–79.31	–6.10	–5.75	6.38	6.73	25	18	16	2.767	–0.16
Aggregation mode										
14×Pt	–84.84	–6.06	–5.53	6.42	6.95	24	19	20	2.768	–0.15
15×Pt	–90.67	–6.04	–5.83	6.44	6.65	30	19	22	2.762	–0.14
16×Pt	–96.65	–6.04	–5.98	6.44	6.50	27	20	26	2.783	–0.13
25×Pt	–147.01	–5.88		6.60		45	17	48	2.762	–0.17
Aggregation mode d										
2×Pt-d	–17.45(–11.78)	–8.73	–4.97	3.76	7.51	6	3	1	2.636	–0.27
3×Pt-d	–22.13	–7.38	–4.68	5.10	7.80	8	4	2	2.687	–0.24
4×Pt-d	–27.45(–22.89)	–6.86	–5.32	5.62	7.16	8	3	3	2.684	–0.28

Table S13. Adsorption energy [$E(Co_{n/ads})$; eV], average adsorption energy [$E(Co_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Co_{ads})$; eV], average aggregation energy [$E_{agg}(Co_{ads})$; eV], stepwise growth energy [$E_{growth}(Co_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Co-Co bond (NB-Co), average Co-Co distance (d, Å), and average Bader charge (q, e) for Co_n on MoC(001).

Species	$E(Co_{n/ads})$	$E(Co_{ads/av})$	$\Delta E(Co_{ads})$	$E_{agg}(Co_{ads})$	$E_{growth}(Co_{ads})$	CN-Mo	CN-C	NB-Co	d	q
1×Co	–4.71	–4.71	–4.71	–	–	2	2	–	–	0.21
Aggregation mode										
2×Co	–9.41	–4.71	–4.70	0.00	0.01	4	4	1	2.373	0.20
3×Co	–14.36	–4.79	–4.95	–0.08	–0.24	6	6	2	2.560	0.21
4×Co	–19.92	–4.98	–5.56	–0.27	–0.85	8	8	4	2.410	0.27
Mode a										
5×Co-a	–23.77	–4.75	–3.85	–0.04	0.86	10	10	5	2.381	0.22
6×Co-a	–28.09	–4.68	–4.32	0.03	0.39	12	12	6	2.389	0.22
7×Co-a	–33.40	–4.77	–5.31	–0.06	–0.60	14	14	8	2.426	0.24
8×Co-a	–37.83	–4.73	–4.43	–0.02	0.28	16	15	11	2.445	0.21
9×Co-a	–42.67	–4.74	–4.84	–0.03	–0.13	18	16	12	2.460	0.19
10×Co-a	–47.66	–4.77	–4.99	–0.06	–0.28	20	18	17	2.490	0.19
11×Co-a	–52.47	–4.77	–4.81	–0.06	–0.10	22	19	21	2.508	0.17
12×Co-a	–57.62	–4.80	–5.15	–0.09	–0.44	24	20	24	2.501	0.16
13×Co-a	–62.17	–4.78	–4.55	–0.07	0.16	24	20	29	2.471	0.15
14×Co-a	–66.81	–4.77	–4.64	–0.06	0.07	24	20	34	2.475	0.14
15×Co-a	–71.46	–4.76	–4.65	–0.05	0.06	24	20	39	2.461	0.13
16×Co-a	–76.07	–4.75	–4.61	–0.04	0.10	24	20	43	2.460	0.12
Mode b										
5×Co-b	–23.88	–4.78	–3.96	–0.07	0.75	10	10	5	2.460	0.21
6×Co-b	–28.18	–4.70	–4.30	0.01	0.41	12	12	7	2.467	0.21
7×Co-b	–32.84	–4.69	–4.66	0.02	0.05	14	14	9	2.503	0.19
8×Co-b	–38.55	–4.82	–5.71	–0.11	–1.00	16	16	12	2.532	0.25
9×Co-b	–43.17	–4.80	–4.62	–0.09	0.09	18	17	14	2.535	0.21
10×Co-b	–47.77	–4.78	–4.60	–0.07	0.11	20	18	17	2.490	0.19
11×Co-b	–52.61	–4.78	–4.84	–0.07	–0.13	22	19	21	2.540	0.18
12×Co-b	–57.54	–4.80	–4.93	–0.09	–0.22	24	20	22	2.480	0.17
13×Co-b	–61.42	–4.72	–3.88	–0.01	0.83	24	20	29	2.497	0.16
14×Co-b	–66.87	–4.78	–5.45	–0.07	–0.74	24	20	37	2.535	0.16
15×Co-b	–71.38	–4.76	–4.51	–0.05	0.20	24	20	39	2.485	0.15
16×Co-b	–76.17	–4.76	–4.79	–0.05	–0.08	24	20	48	2.513	0.15
Dispersion mode c										
2×Co-c	–9.00	–4.50	–4.29	0.21	0.42	4	4	–	–	0.22
3×Co-c	–13.03	–4.34	–4.03	0.37	0.68	6	6	–	–	0.22
4×Co-c	–16.94	–4.24	–3.91	0.48	0.80	8	8	–	–	0.22
5×Co-c	–21.22	–4.24	–4.28	0.47	0.43	10	10	2	2.362	0.20
Three-dimensional mode d										
4×Co-d	–16.83	–4.21	–3.29	0.50	1.42	6	5	6	2.370	0.09
5×Co-d	–22.18	–4.44	–5.35	0.27	–0.64	8	8	6	2.356	0.16
6×Co-d	–27.27	–4.55	–5.09	0.17	–0.38	10	11	8	2.450	0.18
7×Co-d	–31.77	–4.54	–4.50	0.17	0.21	12	11	10	2.497	0.18

Table S14. Adsorption energy [$E(Ni_{n/ads})$; eV], average adsorption energy [$E(Ni_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Ni_{ads})$; eV], average aggregation energy [$E_{agg}(Ni_{ads})$; eV], stepwise growth energy [$E_{growth}(Ni_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Ni-Ni bond (NB-Ni), average Ni-Ni distance (d, Å), and average Bader charge (q, e) for Ni_n on MoC(001).

Species	$E(Ni_{n/ads})$	$E(Ni_{ads/av})$	$\Delta E(Ni_{ads})$	$E_{agg}(Ni_{ads})$	$E_{growth}(Ni_{ads})$	CN-Mo	CN-C	NB-Ni	d	q
1×Ni	–4.52	–4.52	–4.52	–	–	2	2	–	–	0.18
Aggregation mode										
2×Ni	–8.71	–4.36	–4.19	0.16	0.33	4	4	1	2.405	0.15
3×Ni	–13.22	–4.41	–4.51	0.11	0.01	6	5	3	2.494	0.11
4×Ni	–17.55	–4.39	–4.33	0.13	0.19	8	6	5	2.503	0.09
5×Ni	–21.68	–4.34	–4.13	0.18	0.39	10	8	6	2.495	0.10
Mode a										
6×Ni-a	–25.93	–4.32	–4.25	0.20	0.27	12	9	8	2.495	0.08
7×Ni-a	–30.24	–4.32	–4.31	0.20	0.21	14	10	11	2.529	0.08
8×Ni-a	–34.81	–4.35	–4.57	0.17	–0.05	16	12	14	2.573	0.08
9×Ni-a	–39.21	–4.36	–4.40	0.16	0.12	18	13	17	2.601	0.07
Mode b										
6×Ni-b	–25.90	–4.32	–4.22	0.20	0.30	12	10	8	2.568	0.10
7×Ni-b	–30.41	–4.34	–4.51	0.18	0.01	14	11	11	2.572	0.08
8×Ni-b	–34.81	–4.35	–4.40	0.17	0.12	16	12	14	2.574	0.08
9×Ni-b	–39.29	–4.37	–4.48	0.15	0.04	18	14	17	2.586	0.09
Aggregation mode										
10×Ni	–43.89	–4.39	–4.68	0.13	–0.16	20	14	21	2.555	0.07
11×Ni	–48.38	–4.40	–4.49	0.12	0.03	22	15	24	2.566	0.06
12×Ni	–53.08	–4.42	–4.7	0.10	–0.18	24	16	28	2.553	0.06
13×Ni	–57.53	–4.43	–4.45	0.09	0.07	24	16	33	2.533	0.06
14×Ni	–61.95	–4.43	–4.42	0.09	0.10	24	16	36	2.505	0.05
15×Ni	–66.28	–4.42	–4.33	0.10	0.19	24	16	43	2.511	0.05
16×Ni	–70.48	–4.41	–4.20	0.11	0.32	24	16	48	2.509	0.05
Dispersion mode c										
2×Ni-c	–8.60	–4.30	–4.08	0.22	0.44	4	4	–	–	0.19
3×Ni-c	–12.39	–4.13	–3.79	0.39	0.73	6	6	–	–	0.20
4×Ni-c	–16.14	–4.04	–3.75	0.48	0.77	8	8	–	–	0.20
5×Ni-c	–20.41	–4.08	–4.27	0.44	0.25	10	9	2	2.490	0.15
Three-dimensional mode d										
4×Ni-d	–16.55	–4.14	–3.33	0.38	1.19	6	5	6	2.424	0.06
5×Ni-d	–21.25	–4.25	–4.70	0.27	–0.18	8	6	9	2.488	0.07
6×Ni-d	–25.29	–4.22	–4.04	0.31	0.48	10	8	10	2.456	0.08
7×Ni-d	–29.76	–4.25	–4.47	0.27	0.05	12	10	11	2.441	0.08

Table S15. Adsorption energy [$E(Cu_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Cu_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Cu_{ads})$; eV], average aggregation energy [$E_{agg}(Cu_{ads})$; eV], stepwise growth energy [$E_{growth}(Cu_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Cu-Cu bond (NB-Cu), average Cu-Cu distance (d, Å), and average Bader charge (q, e) for Cu_n on MoC(001).

Species	$E(Cu_{n/ads})$	$E(Cu_{ads/av})$	$\Delta E(Cu_{ads})$	$E_{agg}(Cu_{ads})$	$E_{growth}(Cu_{ads})$	CN-Mo	CN-C	NB-Cu	d	q
1×Cu	–2.76	–2.76	–2.76	–	–	2	2	–	–	0.25
Aggregation mode										
2×Cu	–5.82	–2.91	–3.06	–0.15	–0.30	4	2	1	2.362	0.11
3×Cu	–8.63	–2.88	–2.81	–0.12	–0.05	6	3	2	2.389	0.11
Mode a										
4×Cu-a	–11.80 (–8.10)	–2.95	–3.17	–0.19	–0.41	9	4	4	2.478	0.08
5×Cu-a	–14.58	–2.92	–2.78	–0.16	–0.02	8	5	7	2.562	0.09
6×Cu-a	–17.65	–2.94	–3.07	–0.18	–0.31	8	6	10	2.575	0.09
7×Cu-a	–20.55	–2.94	–2.90	–0.18	–0.14	8	6	13	2.554	0.07
8×Cu-a	–23.44 (–21.75)	–2.93	–2.89	–0.17	–0.13	8	6	16	2.549	0.05
9×Cu-a	–26.64	–2.96	–3.20	–0.20	–0.44	7	7	19	2.613	0.05
10×Cu-a	–29.93	–2.99	–3.29	–0.23	–0.53	15	8	21	2.555	0.06
11×Cu-a	–33.37	–3.03	–3.44	–0.27	–0.68	13	8	24	2.527	0.05
12×Cu-a	–37.14	–3.10	–3.77	–0.34	–1.01	11	8	27	2.506	0.04
Mode b (Three-dimensional)										
4×Cu-b	–11.66 (–7.79)	–2.92	–3.03	–0.16	–0.27	6	3	6	2.494	0.09
5×Cu-b	–14.56	–2.91	–2.90	–0.15	–0.14	8	4	8	2.470	0.09
6×Cu-b	–17.57	–2.93	–3.01	–0.17	–0.25	10	5	10	2.544	0.08
7×Cu-b	–20.63	–2.95	–3.06	–0.19	–0.30	11	5	14	2.552	0.05
8×Cu-b	–24.01(–22.27)	–3.00	–3.38	–0.24	–0.62	12	6	15	2.517	0.05
9×Cu-b	–27.01	–3.00	–3.00	–0.24	–0.24	12	7	19	2.559	0.05
10×Cu-b	–30.21	–3.02	–3.20	–0.26	–0.44	12	7	22	2.558	0.04
11×Cu-b	–33.71	–3.06	–3.50	–0.30	–0.74	10	8	25	2.547	0.04
12×Cu-b	–36.92	–3.08	–3.21	–0.32	–0.45	12	8	24	2.508	0.04
Aggregation mode										
13×Cu	–40.56	–3.12	–3.42	–0.36	–0.66	13	8	34	2.575	0.03
14×Cu	–44.44	–3.17	–3.88	–0.41	–1.12	15	8	37	2.557	0.02
15×Cu	–48.04	–3.20	–3.60	–0.44	–0.84	14	8	44	2.507	0.02
Dispersion mode c										
2×Cu-c	–5.03 (–0.53)	–2.52	–2.27	0.25	0.49	4	4	–	–	0.25
3×Cu-c	–6.98	–2.33	–1.95	0.43	0.81	6	6	–	–	0.25
4×Cu-c	–8.92 (–5.40)	–2.23	–1.94	0.53	0.82	8	8	–	–	0.24
5×Cu-c	–14.08	–2.82	–5.16	–0.06	–2.40	10	5	5	2.512	0.09

Table S16. Adsorption energy [$E(Pd_{n/ads})$; eV], average adsorption energy [$E(Pd_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pd_{ads})$; eV], average aggregation energy [$E_{agg}(Pd_{ads})$; eV], stepwise growth energy [$E_{growth}(Pd_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Pd-Pd bond (NB-Pd), average Pd-Pd distance (d, Å), and average Bader charge (q, e) for Pd_n on MoC(001).

Species	$E(Pd_{n/ads})$	$E(Pd_{ads/av})$	$\Delta E(Pd_{ads})$	$E_{agg}(Pd_{ads})$	$E_{growth}(Pd_{ads})$	CN-Mo	CN-C	NB-Pd	d	q
1×Pd	–8.95	–8.95	–8.95	–	–	0	1	–	–	–0.05
Aggregation mode										
2×Pd	–12.59	–6.30	–3.64	2.66	5.31	4	2	1	2.754	–0.10
3×Pd	–15.87	–5.29	–3.28	3.66	5.67	7	3	2	2.747	–0.09
4×Pd	–19.50	–4.88	–3.63	4.08	5.32	8	4	4	2.720	–0.09
5×Pd	–23.23	–4.65	–3.73	4.30	5.22	7	5	6	2.770	–0.09
Mode a										
6×Pd-a	–26.80	–4.47	–3.57	4.48	5.38	8	6	8	2.747	–0.07
7×Pd-a	–29.64	–4.23	–2.84	4.72	6.11	7	6	10	2.739	–0.08
8×Pd-a	–33.72	–4.22	–4.08	4.74	4.87	8	7	13	2.821	–0.07
9×Pd-a	–37.93	–4.21	–4.21	4.74	4.74	8	8	16	2.837	–0.06
10×Pd-a	–41.48	–4.15	–3.55	4.80	5.40	14	8	18	2.717	–0.06
11×Pd-a	–44.89	–4.08	–3.41	4.87	5.54	13	8	25	2.757	–0.06
12×Pd-a	–48.34	–4.03	–3.45	4.92	5.50	12	8	32	2.773	–0.05
13×Pd-a	–51.77	–3.98	–3.43	4.97	5.52	14	8	36	2.789	–0.05
Mode b										
6×Pd-b	–26.84	–4.47	–3.61	4.48	5.34	9	6	8	2.846	–0.07
7×Pd-b	–30.57	–4.37	–3.73	4.58	5.22	8	7	10	2.850	–0.07
8×Pd-b	–34.28	–4.29	–3.71	4.67	5.24	7	8	12	2.856	–0.05
9×Pd-b	–37.93	–4.21	–3.65	4.74	5.30	8	8	16	2.837	–0.06
10×Pd-b	–41.48	–4.15	–3.55	4.80	5.40	14	8	18	2.717	–0.06
11×Pd-b	–44.89	–4.08	–3.41	4.87	5.54	13	8	25	2.757	–0.06
12×Pd-b	–48.34	–4.03	–3.45	4.92	5.50	12	8	31	2.787	–0.05
13×Pd-b	–51.78	–3.98	–3.44	4.97	5.51	7	8	37	2.798	–0.06
Dispersion mode c										
2×Pd-c	–11.94	–5.97	–2.99	2.98	5.96	0	2	0	–	–0.06
3×Pd-c	–16.03	–5.34	–4.09	3.61	4.86	7	3	2	2.734	–0.12
Three-dimensional mode d										
4×Pd-d	–18.78	–4.70	–2.91	4.26	6.04	8	3	5	2.695	–0.09
5×Pd-d	–22.60	–4.52	–3.82	4.43	5.13	6	4	8	2.755	–0.07
6×Pd-d	–26.32	–4.39	–3.72	4.56	5.23	9	5	10	2.774	–0.07
7×Pd-d	–30.08	–4.30	–3.76	4.65	5.19	10	6	12	2.818	–0.08

Table S17. Adsorption energy [$E(Pt_{n/ads})$; eV] (values in parentheses corrected by D3 method), average adsorption energy [$E(Pt_{ads/av})$; eV], stepwise adsorption energy [$\Delta E(Pt_{ads})$; eV], average aggregation energy [$E_{agg}(Pt_{ads})$; eV], stepwise growth energy [$E_{growth}(Pt_{ads})$; eV], coordination number (CN) with surface Mo and C atoms, number of Pt-Pt bond (NB-Pt), average Pt-Pt distance (d, Å), and average Bader charge (q, e) for Pt_n on MoC(001).

Species	$E(Pt_{n/ads})$	$E(Pt_{ads/av})$	$\Delta E(Pt_{ads})$	$E_{agg}(Pt_{ads})$	$E_{growth}(Pt_{ads})$	CN-Mo	CN-C	NB-Pt	d	q
1×Pt	–10.50	–10.50	–10.50	–	–	0	1	–	–	–0.27
Aggregation mode										
2×Pt	–15.80(–11.60)	–7.90	–5.30	2.60	5.20	4	2	1	2.660	–0.26
3×Pt	–21.00	–7.00	–5.20	3.50	5.30	4	3	2	2.636	–0.23
4×Pt	–26.18(–23.38)	–6.55	–5.18	3.96	5.32	2	4	4	2.704	–0.25
5×Pt	–31.42	–6.28	–5.24	4.22	5.26	5	5	6	2.696	–0.14
6×Pt	–36.73(–35.26)	–6.12	–5.31	4.38	5.19	4	6	8	2.679	–0.14
7×Pt	–41.98	–6.00	–5.25	4.50	5.25	8	6	10	2.751	–0.12
Mode a										
8×Pt-a	–47.58	–5.95	–5.60	4.55	4.90	6	7	13	2.703	–0.11
9×Pt-a	–53.21	–5.91	–5.63	4.59	4.87	8	7	16	2.679	–0.11
10×Pt-a	–59.04	–5.90	–5.83	4.60	4.67	8	8	18	2.685	–0.11
11×Pt-a	–64.76	–5.89	–5.72	4.61	4.78	8	7	21	2.636	–0.10
12×Pt-a	–70.34	–5.86	–5.58	4.64	4.92	8	7	31	2.679	–0.09
13×Pt-a	–74.91	–5.76	–4.57	4.74	5.93	10	7	32	2.676	–0.09
14×Pt-a	–79.84	–5.70	–4.93	4.80	5.57	7	7	37	2.711	–0.08
Mode b										
8×Pt-b	–47.68(–47.78)	–5.96	–5.70	4.54	4.80	9	7	14	2.718	–0.12
9×Pt-b	–53.21	–5.91	–5.53	4.59	4.97	8	7	16	2.679	–0.12
10×Pt-b	–59.17	–5.92	–5.96	4.58	4.54	8	8	17	2.718	–0.11
11×Pt-b	–64.79	–5.89	–5.62	4.61	4.88	10	8	23	2.672	–0.10
12×Pt-b	–70.17	–5.85	–5.38	4.65	5.12	7	8	32	2.695	–0.09
13×Pt-b	–75.10	–5.78	–4.93	4.72	5.57	8	8	36	2.740	–0.07
14×Pt-b	–80.32	–5.74	–5.22	4.76	5.28	6	8	39	2.735	–0.07
Dispersed mode c										
2×Pt-c	–14.84(–10.66)	–7.42	–4.34	3.08	6.16	6	2	0	–	–0.31
3×Pt-c	–19.62	–6.54	–4.78	3.96	5.72	6	3	2	2.859	–0.21
Three-dimensional mode d										
4×Pt-d	–25.66	–6.42	–5.00	4.09	5.50	5	3	5	2.683	–0.21
5×Pt-d	–30.67(–28.35)	–6.13	–5.01	4.37	5.49	5	4	9	2.787	–0.17
6×Pt-d	–36.21	–6.04	–5.54	4.47	4.96	6	5	10	2.739	–0.15
7×Pt-d	–41.59	–5.94	–5.38	4.56	5.12	6	6	11	2.733	–0.13

Table S18. Comparison of lattice constants of Mo_xC_y and bulk metals.

Bulk	Phase	Theory	Experiment
Mo_2C	hcp	3.040	3.002[23]
δ-MoC	fcc	4.360	4.278[24]
Co	fcc	3.53[25]	3.55[26]
Co	hcp	3.521[27]	3.545[28]
Ni	fcc	3.52[29]	3.524[30]
Cu	fcc	3.637[31]	3.610[32]
Pd	fcc	3.95[33]	3.89[34]
Pt	fcc	4.00[35]	3.912[36]

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