Supporting Information

$K_{1-x}Mo_3P_2O_{14}$ as Support for Single-Atom Catalysts

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Table S1. Calculated and experimental lattice parameters a, b, and c (Å) of KMPO bulk structure.

	a	b	c
calculated	8.34	6.51	10.71
experimental[1]	8.59	6.39	10.60

Table S2. Adsorption energy E_{ads} (eV) of single metal atoms on $K_{1-x}Mo_3P_2O_{14}$ monolayer.

	Ag	Au	Pd	Pt
E_{ads}	-2.29	-1.98	-2.99	-3.95

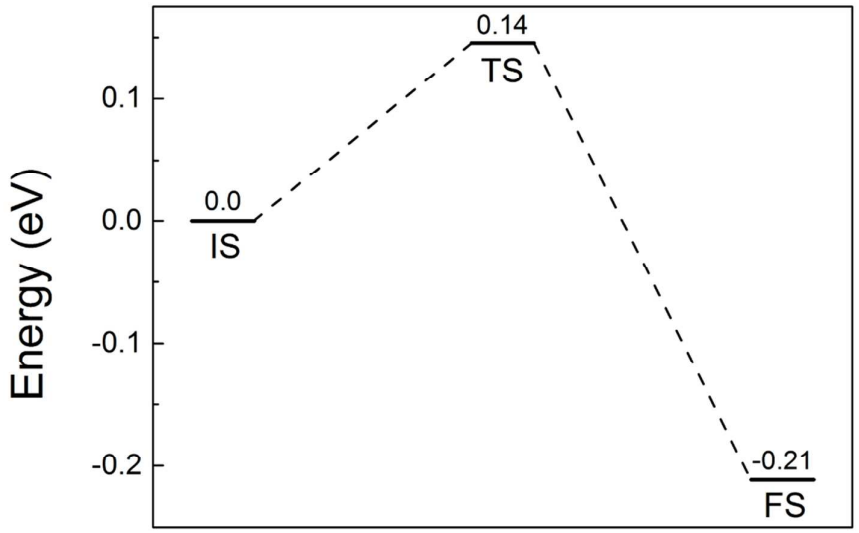

Figure S1. Energy profiles for the substitution of K by Pt atoms.

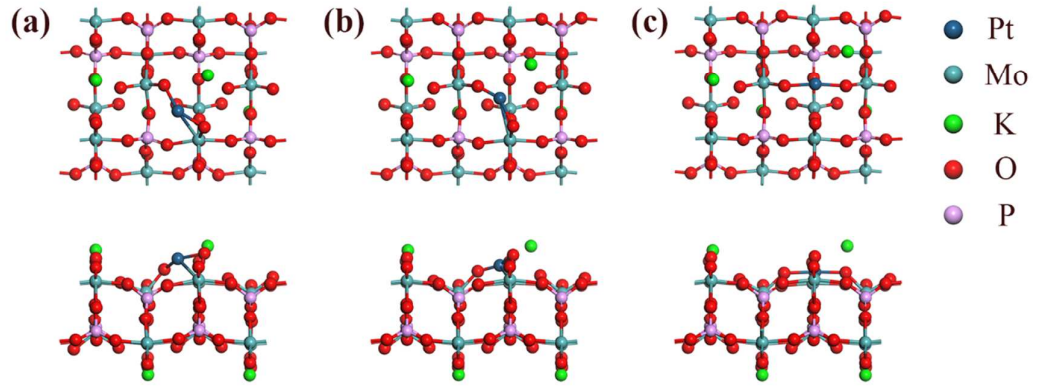

Figure S2. Top (up) and side (down) view of the atomic configurations of the initial state (IS), transition state (TS) and final state (FS) for the substitution of K by Pt atoms.

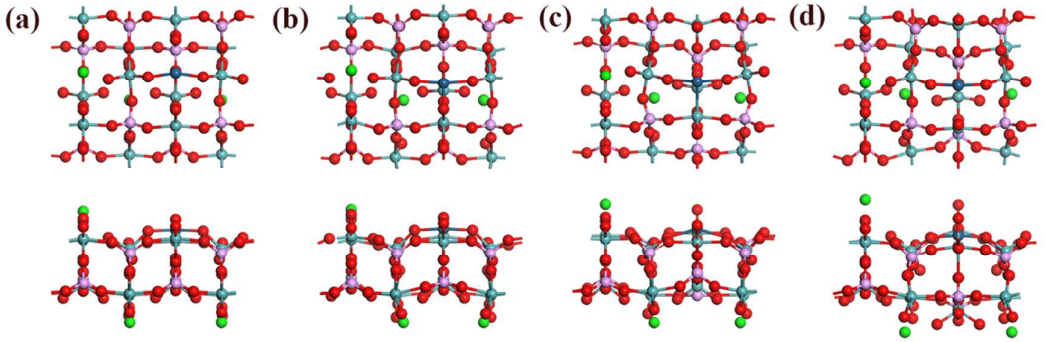

Figure S3. Top (up) and side (down) view of the optimized geometries of (a) $Pt/K_{1-x}Mo_3P_2O_{14}$ catalyst and the snapshots at (b) 800 K, (c) 1000 K, and (c) 1200 K after 10 ps AIMD simulations.

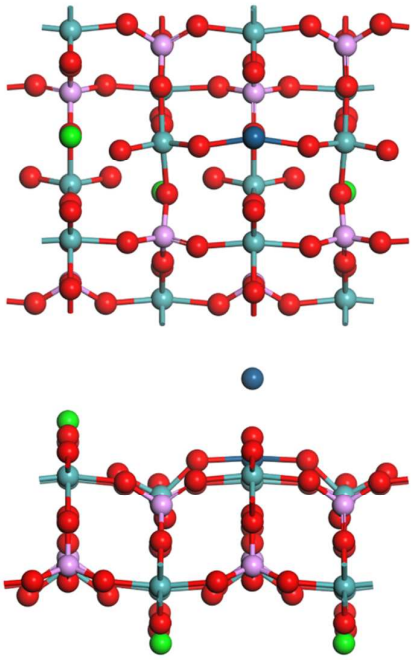

Figure S4. Top (up) and side (down) view of the second Pt atom adsorbed on $Pt/K_{1-x}Mo_3P_2O_{14}$.

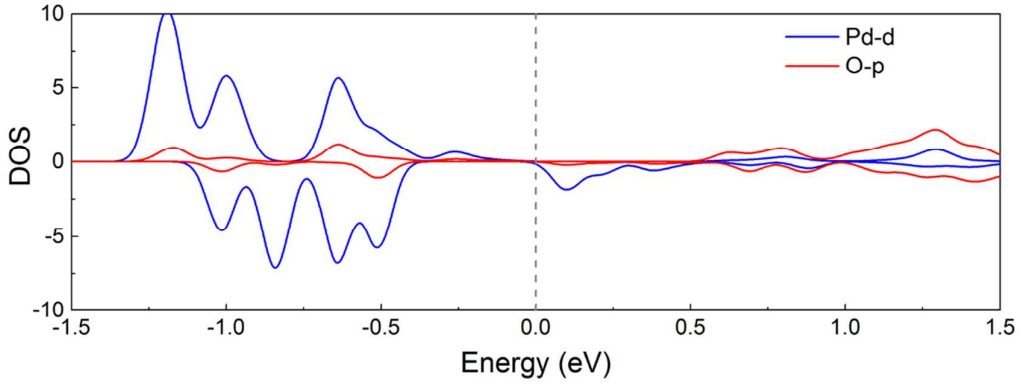

Figure S5. Computed projected density of states (PDOS) for $Pt/K_{1-x}Mo_3P_2O_{14}$ catalyst. The Fermi level is set to be zero (dash lines).

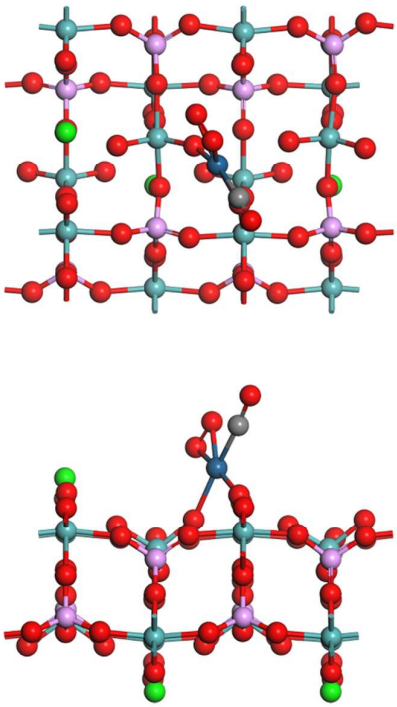

Figure S6. Top (up) and side (down) view of the coadsorption geometry before optimization.

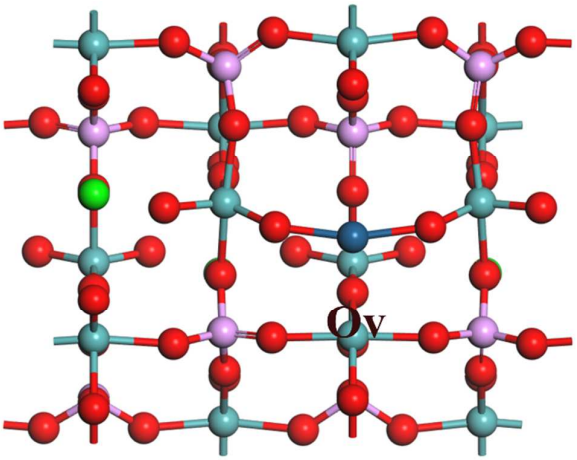

Figure S7. Top view of optimized geometries of KPtMPO catalyst containing one oxygen vacancy.

Table S3. Overall energy barriers E (eV) for CO oxidation on different single-metal atom catalysts.

Catalysts	Mechanisms	E	Ref.
KPtMPO	LH	0.49	This work
Fe/Graphene	ER	0.58	[2]
Oxygen-Defective Pt/FeO_x	LH	0.79	[3]
Gu/Graphene	LH/ER	0.54	[4]
Fe/h-BN	ER	0.61	[5]
CoPc	LH	0.65	[6]
FePc	LH	0.73	
CuPc	ER	0.90	
ZnPc	LH	0.51	
Pt/pri-Graphene	LH	1.03	[7]
Single-Vacancy Pt/Graphene	LH	0.58	
Co/BN	LH	0.55	[8]
	ER	0.59	
	TER	0.41	
$Si-GN_4$	ER	0.72	[9]
Ni_1/FeO_x	-	0.75	[10]

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