Supplementary Materials

PdAg/Ag(111) Surface Alloys: A Highly Efficient Catalyst of Oxygen Reduction Reaction

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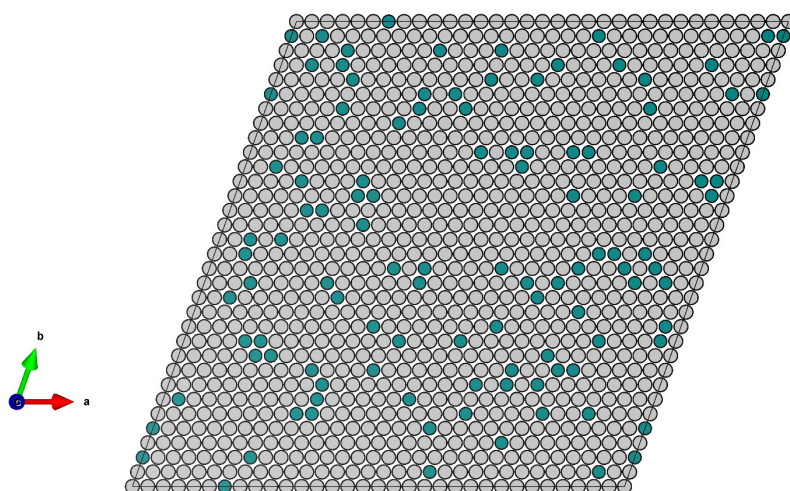* Correspondence: lxh12345@sdu.edu.cn (X.L.); tianxuelei@sdu.edu.cn (X.T.)

Table S1 The free energies of the adsorption of ORR intermediates in eV on all active site models.
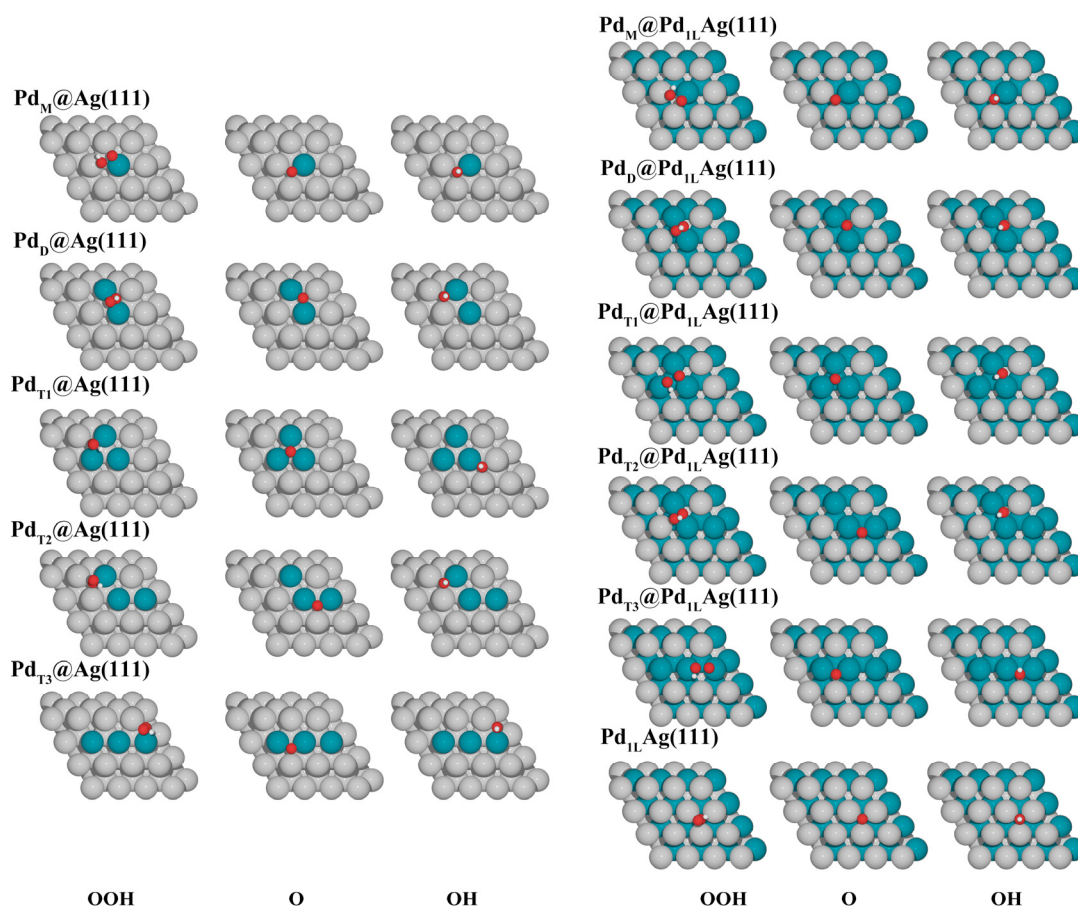
Active site model	ΔG_{*OOH}	ΔG_{*O}	ΔG_{OH*}
$Pd_M@Ag(111)$	4.15194	1.76072	0.78351
$Pd_D@Ag(111)$	4.08826	1.64927	0.80979
$Pd_{T1}@Ag(111)$	4.14847	1.62848	0.87055
$Pd_{T2}@Ag(111)$	4.16367	1.69931	0.85212
$Pd_{T3}@Ag(111)$	4.16664	1.67661	0.84570
$Pd_M@Pd_{1L}Ag(111)$	4.10261	1.84045	0.81721
$Pd_D@Pd_{1L}Ag(111)$	4.02477	1.63452	0.75966
$Pd_{T1}@Pd_{1L}Ag(111)$	3.98324	1.40795	0.78896
$Pd_{T2}@Pd_{1L}Ag(111)$	4.05015	1.54504	0.79159
$Pd_{T3}@Pd_{1L}Ag(111)$	4.04202	1.62215	0.75420
$Pd_{1L}Ag(111)$	4.23506	2.10597	0.82387

Table S2 *d*-band center and Bader charge of Pd atom at the first layer of all

models

Active site model	*d*-band center	Bader charge
Pd_M@Ag(111)	−1.545	−0.229
Pd_D@Ag(111)	−1.496	−0.207
Pd_{T1}@Ag(111)	−1.617	−0.188
Pd_{T2}@Ag(111)	−1.609	−0.198
Pd_{T3}@Ag(111)	−1.593	−0.198
Pd_M@Pd_{1L}Ag(111)	−1.329	−0.154
Pd_D@Pd_{1L}Ag(111)	−1.292	−0.129
Pd_{T1}@Pd_{1L}Ag(111)	−1.426	−0.116
Pd_{T2}@Pd_{1L}Ag(111)	−1.410	−0.121
Pd_{T3}@Pd_{1L}Ag(111)	−1.394	−0.122

Figure S1 MC snapshots of the atomic structure of PdAg(111) with 50% Pd at

1200 K.[1] The silver and green balls represent Ag and Pd atoms, respectively.

Figure S2 Top view of the atomistic structures of all ORR intermediates on all considered surfaces. The silver, cyan, red, and white balls represent Ag, Pd, O, and H atoms, respectively.

References

[1] Hua, M.; Tian, X.; Li, S.; Zhang, X.; Shao, A.; Song, L.; Lin, X. A casting combined quenching strategy to prepare PdAg single atom alloy designed by Cluster Expansion combined Monte Carlo method. *Phys. Chem. Chem. Phys.* 2021, doi:10.1039/d1cp05046j.