ARTICLE TYPE

Size-selective Electrocatalytic Activity of $(Pt)_n/MoS_2$ for Oxygen Reduction Reaction

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TableS1. Total Binding energies ΔE_{BE} (eV) of the Pt nanoparticles on 2H and 1T MoS_2 Surfaces

Model structure	Energy (eV)
2H-MoS_2/Pt_1	-2.78
2H-MoS_2/Pt_2	-2.90
2H-MoS_2/Pt_3	-3.19
2H-MoS_2/Pt_5	-3.45
2H-MoS_2/Pt_7	-3.68
2H-MoS_2/Pt_{10}	-3.92
2H-MoS_2/Pt_{12}	-4.09
1T-MoS_2/Pt_1	-2.92
1T-MoS_2/Pt_2	-3.15
1T-MoS_2/Pt_3	-3.42
1T-MoS_2/Pt_5	-3.80
1T-MoS_2/Pt_7	-3.98
1T-MoS_2/Pt_{10}	-4.12
1T-MoS_2/Pt_{12}	-4.25

The binding energy per Pt for $(Pt)_n$ nanoparticles on MoS_2 surfaces has been computed using the following equation:

$$E_{BE} = \frac{1}{n}[E_{MoS_2+(Pt)_n} - E_{MoS_2} - nE_{Pt}] \quad (1)$$

where $E_{MoS_2+(Pt)_n}$ is the total energy of $(Pt)_n/MoS_2$ heterostructures, E_{MoS_2} is the total energy of pristine MoS_2 surface, E_{Pt} is the energy of the Pt atom. According to this convention, the negative binding energies imply stable adsorption configuration.

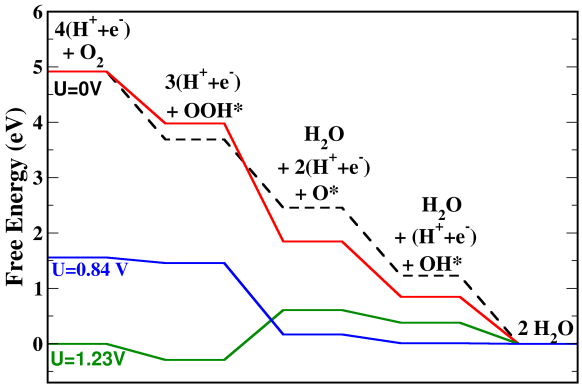

Figure S1.Free energy diagram for ORR on $(Pt)_5$/H-MoS_2 system at U = 0 V (red line), U = 1.23 V (green line) and U = 0.84 V (blue line). The optimal ORR catalyst at U = 0 V (black dashed line) is shown for comparison respectively.

Table S2. Calculated Overpotential (V) of the Pt nanoparticles on 2H and 1T-MoS_2 Surfaces

Model Structure	Overpotential (V)
H-MoS_2/Pt_1	0.52
H-MoS_2/Pt_2	1.14
H-MoS_2/Pt_3	0.61
H-MoS_2/Pt_5	0.38
H-MoS_2/Pt_7	0.33
H-MoS_2/Pt_{10}	0.37
H-MoS_2/Pt_{12}	0.47
T-MoS_2/Pt_1	0.64
T-MoS_2/Pt_2	0.81
T-MoS_2/Pt_3	0.79
T-MoS_2/Pt_5	0.71
T-MoS_2/Pt_7	0.66
T-MoS_2/Pt_{10}	0.60
T-MoS_2/Pt_{12}	0.62

We have calculate overpotential η, an experimentally measurable quantity, of the fuel cell as a difference between the equilibrium of the ORR process for each step (4.92/4 = 1.23 V) and the lowest potential (RLS) where all the elementary steps are downhill. (Table S2) However, this definition is based on the assumption that the binding energies are independent of the applied voltage which is generally an well-established approximation.

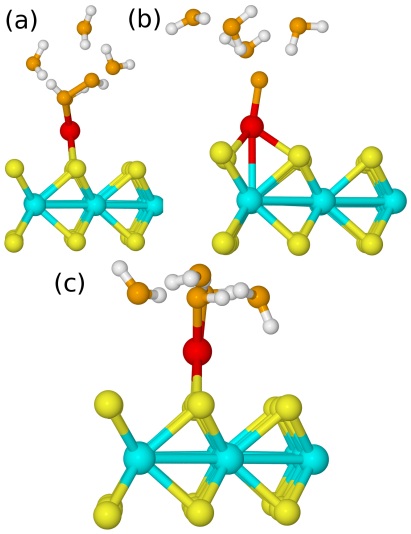

Figure S2. Side view of the adsorption configuration for (a) *OOH, (b) *O, (c) *OH in the presence of water modeled using 4 molecules respectively.