

Supporting Information for:

Trends in adsorbed hydroxyl – water interactions on late transition metal nanoparticles

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S.1. Construction of nanoparticles and micro-solvated *OH and H₂O configurations

The truncated octahedral nanoparticles for each metal were created by relaxing the converged coordinates for Pt nanoparticles found in previous works.¹ Micro-solvated *OH configurations, more specifically the hydrogen and oxygen atom positions, were taken from the equivalent configurations relaxed in previous works.¹ The OH moiety and water molecules in each configuration were adsorbed onto nanoparticle atoms, equivalent to those on the Pt nanoparticle based on the local coordination environments within the nanoparticle. From these *OH structures, corresponding H₂O configurations are generated by replacing the OH moiety by a central H₂O molecule, moving and rotating this H₂O molecule away from the surface, and adjusting the orientation and position of the remainder three water molecules to obtain a roughly tetrahedral coordination of the central H₂O oxygen atom. Ultimately, the central water molecule should form hydrogen bonds with each of the three peripheral water molecules, as shown in Figure 1 in the main text.

Additionally, corresponding *OH and H₂O configurations should agree on the features illustrated in Figure S.1: the orientation of the non-binding H atoms of the surface-bound solvent water molecules relative to the mirror plane intersecting the surface-bound solvent and solute adsorption site atom; the (downwards) orientation of one H atom on the non-surface bound water molecule; and (for non-mirror symmetric configurations) the orientation of the second H atom on the non-surface bound water molecule relative to the plane containing the solute O atom, the non-surface bound water molecule O atom and the solute adsorption site.

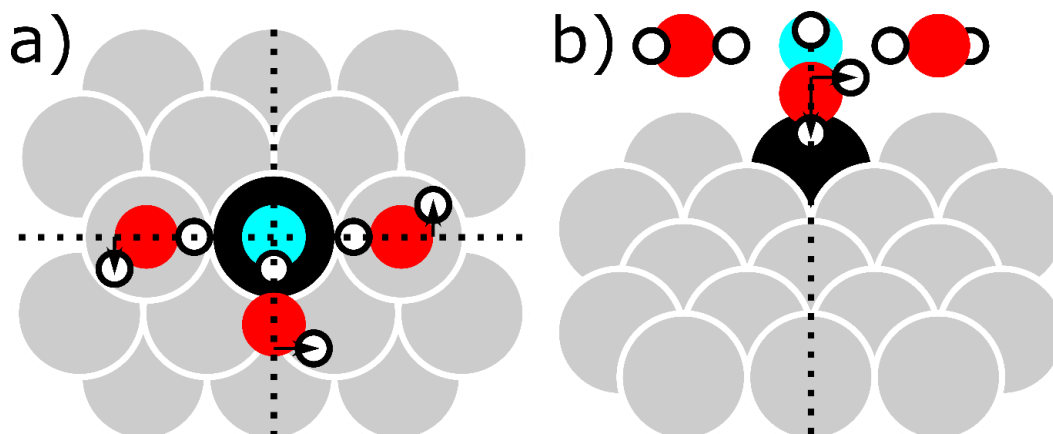


Figure S.1. Features which should be consistent between corresponding micro-solvated OH and H₂O configurations, in (a) top and (b) lateral views. Nanoparticle atoms are gray or (for the adsorption site) black; solvent oxygen atoms are red, solute oxygen atoms are cyan, and hydrogen atoms are white; relevant planes (perpendicular to the point of view) are shown as dashed lines, while defining displacements relative to these planes are shown using arrows.

S.2. Computational details

All calculations were performed using VASP 5.4.4.²⁻⁵ For each calculation, a 450-eV plane-wave cut-off and a 10^{-5} -eV electronic convergence step cutoff were used, unless specified otherwise. All calculations were performed for the Γ -point only. Free molecules were calculated in an 11 Å cubic unit cell, with $k_B T = 0.001$ eV and Gaussian smearing of the Fermi level. All systems containing or derived from nanoparticles are calculated using $k_B T = 0.2$ eV and second-order Methfessel-Paxton smearing of the Fermi level.⁶ Systems derived from 38-atom nanoparticles are calculated in 17.8 Å cubic unit cells, while systems derived from 79- and 201-atom nanoparticles are calculated in 26 Å cubic unit cells. The configurations created in Section S.1 are relaxed using a conjugate gradient algorithm until all atomic forces are smaller than 0.05 eV \AA^{-1} . After each relaxation, the relaxed configuration is re-assessed according to the rules set out in Section S.1 and, if necessary, corrected by applying mirror transformations to those solvent and solute moieties which are inconsistent.

For each relaxed configuration, vibrational energies are calculated for all O and H atoms through single-atom displacements along each Cartesian unit vector using 0.02 Å steps in both directions to assess the zero-point energies and the vibrational entropies. If no imaginary modes are found, the configuration is considered to be converged and is used for further calculations. If imaginary modes are found, relaxation of the configuration is continued up to 0.02 eV \AA^{-1} , and another vibrational energy calculation is performed. If more than one imaginary mode persists in the vibrational energy calculation after additional convergence, an additional vibrational calculation is performed using more dense electronic plane wave sampling (600 eV) and stricter electronic convergence (10^{-7} eV electronic step size) to reduce noise in the force calculations. Although the stricter criteria may increase forces on configuration ground states converged with the lesser criteria, resulting vibrational modes are sufficiently close to those obtained using the lesser criteria, as shown in Table S.1.

Table S.1. Vibrational modes for several converged configurations.

	Au ₇₉ kink, H ₂ O				Ir ₇₉ kink, OH			
	Lax (meV)	Strict (meV)	Diff (meV)	Corr ^a (-)	Lax (meV)	Strict (meV)	Diff (meV)	Corr ^a (-)
ZPE	2601	2612	11	-	2337	2337	0	-
TS_{vib}	574	696	122	-	488	528	39	-
G_{vib}^b	2027	1916	-111	-	1849	1810	-38	-
Mode 1	464.8	466.2	1.4	1.00	467.2	466.5	-0.7	1.00
Mode 2	463.7	465.8	2.1	0.91	462.7	461.9	-0.8	1.00
Mode 3	463.0	465.4	2.4	0.90	461.2	459.7	-1.6	1.00
Mode 4	462.1	465.1	3.0	0.99	433.0	431.7	-1.3	1.00
Mode 5	424.9	426.9	2.0	1.00	416.5	415.4	-1.1	1.00
Mode 6	419.4	421.7	2.4	1.00	347.4	347.0	-0.4	1.00
Mode 7	412.6	414.4	1.8	1.00	336.9	336.3	-0.5	1.00
Mode 8	384.4	385.9	1.5	1.00	199.2	198.9	-0.3	1.00
Mode 9	202.0	201.7	-0.3	1.00	195.1	194.7	-0.5	0.17
Mode 10	198.7	199.0	0.3	0.99	195.0	194.3	-0.7	0.17
Mode 11	198.2	198.4	0.2	0.99	133.1	132.0	-1.0	1.00
Mode 12	195.6	195.6	0.1	1.00	123.9	123.3	-0.7	0.99
Mode 13	114.1	114.7	0.6	1.00	121.5	121.2	-0.3	0.99
Mode 14	94.5	95.5	1.0	0.99	105.1	104.8	-0.3	1.00
Mode 15	93.1	94.0	0.9	0.99	85.4	85.0	-0.3	1.00
Mode 16	69.3	69.8	0.5	1.00	81.6	81.1	-0.6	1.00
Mode 17	60.3	59.8	-0.5	0.99	58.3	57.7	-0.6	0.98
Mode 18	58.2	58.2	0.0	0.99	56.4	55.4	-1.0	0.83
Mode 19	51.3	51.6	0.2	0.78	54.5	54.4	-0.1	0.83
Mode 20	49.3	49.8	0.5	0.78	51.5	49.9	-1.6	0.99
Mode 21	46.6	46.0	-0.6	0.99	41.1	40.7	-0.5	0.99
Mode 22	42.2	41.3	-0.9	0.99	39.9	39.4	-0.6	0.79
Mode 23	41.3	39.9	-1.3	0.99	39.2	38.7	-0.5	0.79
Mode 24	32.3	32.3	0.0	0.70	30.4	31.2	0.8	0.84
Mode 25	29.8	29.8	0.0	0.69	29.5	30.4	0.8	0.84
Mode 26	25.5	25.2	-0.3	0.98	26.1	26.5	0.3	0.90
Mode 27	24.0	23.0	-1.0	0.98	23.1	23.3	0.2	0.90
Mode 28	18.5	17.3	-1.2	0.70	15.8	16.6	0.8	0.98
Mode 29	16.9	16.7	-0.3	0.65	13.3	14.5	1.2	0.92
Mode 30	15.0	13.9	-1.1	0.94	10.4	13.1	2.7	0.94
	Au ₇₉ kink, H ₂ O				Ir ₇₉ kink, OH			
	Lax (meV)	Strict (meV)	Diff (meV)	Corr ^a (-)	Lax (meV)	Strict (meV)	Diff (meV)	Corr ^a (-)
Mode 31	11.5	10.9	-0.5	0.60	9.1	11.8	2.7	0.98
Mode 32	9.6	8.0	-1.6	0.18	7.3	9.1	1.8	0.84
Mode 33	8.0	7.0	-1.0	0.40	3.6	7.5	3.8	0.87
Mode 34	1.2	5.7	4.5	0.48	-	-	-	-
Mode 35	9.0 i	4.5	-	0.55	-	-	-	-
Mode 36	9.6 i	3.7	-	0.84	-	-	-	-

a: Inner product of lesser and stricter vibrational mode eigenvectors.

b: Vibrational free energies as defined in Section S.3.

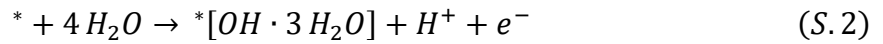
S.3. Thermodynamics

Individual formation energies were calculated for all OH and H₂O (deemed OH_{*x*}, *x* = 1,2) configurations on each nanoparticle at sites *s* in micro-solvated and vacuum environments.

Since the full formation energy for micro-solvated H₂O formation from bulk H₂O is:



and its micro-solvated *OH counterpart is:



we can use the computational hydrogen electrode to obtain the following formation energies on each metal:⁷

$$\Delta G_{MS}(OH_x; s) = E_{DFT,MS}(OH_x; s) + E_{ZPE,MS}(OH_x; s) - TS_{MS}(OH_x; s) - E_M - 4G_{H_2O} + \frac{(2-x)}{2}G_{H_2} \quad (S.3)$$

in which $\Delta E_{DFT,MS}$ is the DFT energy of the specific configuration, $E_{ZPE,MS}$ is the zero-point energy, T is the temperature (300 K), S_{MS} is the vibrational entropy, and E_M is the DFT energy of the pristine nanoparticle. G_{H_2O} and G_{H_2} are the Gibbs free energies for H₂O(*l*) and H₂(*g*) under standard conditions, respectively, defined using entropies and the heat of fusion for H₂O from literature,⁸ as:

$$G_{H_2O(l)} = E_{DFT}(H_2O) + E_{ZPE}(H_2O) + \Delta H_{fus}(H_2O) - TS_{lit}(H_2O(l))$$

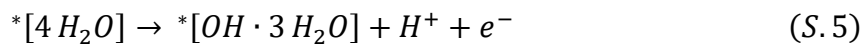
which is -2.46 eV with respect to G_{H_2} and G_{O_2} , and

$$G_{H_2(g)} = E_{DFT}(H_2) + E_{ZPE}(H_2) - TS_{lit}(H_2(g))$$

Analogously, individual formation energies were calculated for all vacuum (i.e., non-micro-solvated) OH configurations, described using: $* + H_2O(l) \rightarrow *OH + H^+ + e^-$, as:

$$\begin{aligned} \Delta G_{\text{vac}}(\text{OH}_x; s) = & E_{\text{DFT,vac}}(\text{OH}_x; s) + E_{\text{ZPE,vac}}(\text{OH}_x; s) - TS_{\text{vac}}(\text{OH}_x; s) \\ & - E_{\text{DFT,M}} - \Delta G_{\text{H}_2\text{O}} + \frac{1}{2} \Delta G_{\text{H}_2} \end{aligned} \quad (\text{S. 4})$$

From the micro-solvated formation energies, the micro-solvated formation energies for OH are calculated. This micro-solvated formation energy corresponds to the following chemical reaction:



from which we can derive the formation energy relative to analogous to electrochemical formation potentials using the computational hydrogen electrode,⁷ as:

$$\Delta G_{f,\text{MS}} = \Delta G_{\text{MS}}(\text{OH}, s) - \Delta G_{\text{MS}}(\text{H}_2\text{O}, s) + \frac{1}{2} \Delta G_{\text{H}_2} \quad (\text{S. 6})$$

Similarly, the vacuum formation energies for OH are calculated with respect to liquid water, as implied above:

$$\Delta G_{f,\text{vac}} = \Delta G_{\text{vac}}(\text{OH}, s) - \Delta G_{\text{H}_2\text{O}} + \frac{1}{2} \Delta G_{\text{H}_2} \quad (\text{S. 7})$$

Using these formation energies, we can obtain the (micro)solvation energy for OH on each specific site:

$$\Omega_{\text{OH}} = \Delta G_{f,\text{MS}} - \Delta G_{f,\text{vac}} \quad (\text{S. 8})$$

S.4. Formation energy thermodynamics and statistics

Table S.2. Binding and formation energies.

NP	site	\overline{CN}	Micro-solvated					Vacuum				$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}
				ΔE	ΔE_{ZPE}	TS	ΔG_{corr}	ΔE	ΔE_{ZPE}	TS	ΔG_{corr}			
Ag ₃₈	111	6.00	OH	-0.78	2.22	0.55	-0.06	0.34	0.32	0.11	-	0.55	0.11	-0.45
			H ₂ O	-1.10	2.58	0.68	-0.08	-	-	-	-	-	-	-
	kink	4.00	OH	-1.07	2.26	0.56	-0.06	0.23	0.34	0.11	-	0.46	-0.25	-0.71
			H ₂ O	-1.00	2.58	0.67	-0.08	-	-	-	-	-	-	-
Ag ₇₉	111E	5.00	OH	-0.87	2.26	0.52	-0.06	0.93	0.31	0.12	-0.05	1.06	0.06	-1.00
			H ₂ O	-1.17	2.59	0.60	-0.08	-	-	-	-	-	-	-
	111T	6.67	OH	-0.63	2.24	0.60	-	1.04	0.30	0.19	-0.05	1.10	0.19	-0.90
			H ₂ O	-1.11	2.60	0.60	-0.08	-	-	-	-	-	-	-
	kink	4.08	OH	-0.78	2.25	0.54	-0.06	0.93	0.31	0.10	-0.05	1.08	-0.02	-1.11
			H ₂ O	-1.01	2.59	0.70	-	-	-	-	-	-	-	-
Ag ₂₀₁	100E	5.17	OH	-0.94	2.25	0.55	-0.06	0.86	0.31	0.19	-	0.99	-0.01	-1.00
			H ₂ O	-1.17	2.59	0.72	-	-	-	-	-	-	-	-
	100T	6.33	OH	-0.79	2.23	0.52	-0.06	0.39	0.33	0.13	-	0.59	0.12	-0.47
			H ₂ O	-1.11	2.59	0.66	-0.08	-	-	-	-	-	-	-
	111E	5.00	OH	-0.99	2.25	0.54	-0.06	0.83	0.31	0.10	-0.05	0.99	-0.02	-1.01
			H ₂ O	-1.21	2.60 ^a	0.72 ^a	-	-	-	-	-	-	-	-
	111TC	7.50	OH	-0.35	2.23	0.54	-0.06	1.14	0.30	0.11	-0.05	1.28	0.35	-0.93
			H ₂ O	-0.91	2.60 ^a	0.76 ^a	-	-	-	-	-	-	-	-
	111TM	6.92	OH	-0.64	2.24 ^a	0.54 ^a	-0.06	-	-	-	-	x	0.19	x
			H ₂ O	-1.06	2.59	0.64	-0.08	-	-	-	-	-	-	-
	kink	4.25	OH	-0.85	2.24	0.60	-0.06	0.82	0.31	0.12	-0.05	0.95	-0.14	-1.10
			H ₂ O	-0.97	2.57	0.65	-0.08	-	-	-	-	-	-	-
Au ₃₈	111	6.00	OH	-0.66	2.25	0.58	-	x	x	x	-	x	0.33	x
			H ₂ O	-1.22	2.60	0.62	-0.08	-	-	-	-	-	-	-
	kink	4.00	OH	-0.98	2.29	0.53	-0.06	0.52	0.32	0.18	-	0.67	0.34	-0.32
			H ₂ O	-1.58	2.62	0.67	-	-	-	-	-	-	-	-
Au ₇₉	111E	5.00	OH	-0.43	2.29	0.55	-	1.11	0.32	0.12	-0.05	1.26	0.63	-0.63
			H ₂ O	-1.23	2.60	0.68	-	-	-	-	-	-	-	-
	111T	6.67	OH	-0.17	2.26	0.48	-0.06	1.37	0.32	0.18	-	1.51	0.71	-0.80
			H ₂ O	-1.11	2.61	0.58	-0.08	-	-	-	-	-	-	-
	kink	4.08	OH	-0.38	2.30	0.58	-	0.99	0.33	0.15	-	1.17	0.48	-0.69
			H ₂ O	-1.10	2.62	0.66	-	-	-	-	-	-	-	-
Au ₂₀₁	100E	5.17	OH	-0.47	2.30 ^a	0.55 ^a	-	1.08	0.33	0.14	-	1.27	0.51	-0.76
			H ₂ O	-1.13	2.60	0.62	-0.08	-	-	-	-	-	-	-
	100T	6.33	OH	-0.42	2.27 ^a	0.53 ^a	-0.06	x	x	x	-	x	0.43	x
			H ₂ O	-1.11	2.62 ^a	0.68 ^a	-	-	-	-	-	-	-	-
	111E	5.00	OH	-0.56	2.29	0.50	-0.06	1.07	0.32	0.10	-0.05	1.23	0.39	-0.84
			H ₂ O	-1.16	2.61	0.60	-0.08	-	-	-	-	-	-	-
	111TC	7.50	OH	0.23	2.27	0.59	-	1.41	0.31	0.11	-0.05	1.56	0.89	-0.67
			H ₂ O	-0.85	2.59	0.65	-0.08	-	-	-	-	-	-	-
	111TM	6.92	OH	-0.08	2.27	0.55	-	1.38	0.31	0.11	-0.05	1.53	0.69	-0.84
			H ₂ O	-1.00	2.62	0.67	-	-	-	-	-	-	-	-
	kink	4.25	OH	-0.41	2.28	0.66	-	0.94	0.32	0.11	-0.05	1.10	0.30	-0.80
			H ₂ O	-0.94	2.58	0.65	-0.08	-	-	-	-	-	-	-
Ir ₃₈	111	6.00	OH	-2.02	2.29	0.46	-	0.43	0.34	0.13	-	0.64	-0.18	-0.82
			H ₂ O	-2.10	2.64	0.56	-	-	-	-	-	-	-	-
	kink	4.00	OH	-2.34	2.33	0.48	-	-0.46	0.35	0.10	-	-0.21	-0.65	-0.44
			H ₂ O	-1.93	2.64	0.54	-	-	-	-	-	-	-	-
Ir ₇₉	111E	5.00	OH	-2.39	2.32	0.47	-	0.09	0.32	0.12	-0.05	0.24	-0.46	-0.69
			H ₂ O	-2.14	2.64	0.58	-	-	-	-	-	-	-	-

<i>NP</i>	site	\overline{CN}	Micro-solvated					Vacuum				$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}
				ΔE	ΔE_{ZPE}	<i>TS</i>	ΔG_{corr}	ΔE	ΔE_{ZPE}	<i>TS</i>	ΔG_{corr}			
Ir ₇₉	111T	6.67	OH	-1.52	2.27	0.42	-0.06	0.62	0.34	0.12	-	0.84	-0.01	-0.85
			H ₂ O	-1.83	2.64	0.55	-	-	-	-	-			
	kink	4.08	OH	-2.10	2.34	0.49	-	-0.32	0.33	0.14	-	-0.12	-0.60	-0.47
			H ₂ O	-1.71	2.63	0.57	-	-	-	-	-			
Ir ₂₀₁	100E	5.17	OH	-2.34	2.32	0.47	-	x	x	x	-	x	-0.48	x
			H ₂ O	-2.07	2.64	0.58	-	-	-	-	-			
	100T	6.33	OH	-1.59	2.27	0.40	-0.06	0.48	0.33	0.13	-	0.68	0.12	-0.57
			H ₂ O	-1.95	2.64	0.50	-0.08	-	-	-	-			
	111E	5.00	OH	-2.40	2.31	0.50	-	-0.10	0.35	0.11	-	0.14	-0.50	-0.63
			H ₂ O	-2.19	2.64	0.55	-	-	-	-	-			
	111TC	7.50	OH	-1.06	2.32	0.42	-	0.68	0.34	0.12	-	0.89	0.11	-0.79
		H ₂ O	-1.34	2.64	0.56	-	-	-	-	-				
	111T M	6.92	OH	-1.60	2.31	0.46	-	0.53	0.33	0.14	-	0.72	0.06	-0.66
			H ₂ O	-1.91	2.65	0.55	-	-	-	-	-			
	kink	4.25	OH	-2.12	2.33	0.44	-0.06	-0.39	0.33	0.13	-	-0.19	-0.57	-0.38
		H ₂ O	-1.72	2.61	0.54	-0.08	-	-	-	-				
Pd ₃₈	111	6.00	OH	-0.69	2.26	0.47	-0.06	1.05	0.32	0.21	-	1.15	0.59	-0.56
			H ₂ O	-1.52	2.61	0.64	-	-	-	-	-			
	kink	4.00	OH	-1.10	2.30	0.55	-	0.60	0.33	0.14	-	0.78	0.10	-0.69
			H ₂ O	-1.42	2.61	0.57	-0.08	-	-	-	-			
Pd ₇₉	111E	5.00	OH	-1.31	2.30	0.55	-	0.64	0.32	0.10	-0.05	0.81	0.11	-0.70
			H ₂ O	-1.67	2.61	0.54	-0.08	-	-	-	-			
	111T	6.67	OH	-0.93	2.28	0.52	-	0.88	0.32	0.11	-0.05	1.04	0.32	-0.72
		H ₂ O	-1.49	2.61	0.60	-	-	-	-	-				
	kink	4.08	OH	-1.07	2.29	0.53	-	0.66	0.32	0.09	-0.05	0.84	0.06	-0.77
			H ₂ O	-1.39	2.62	0.59	-	-	-	-	-			
Pd ₂₀₁	100E	5.17	OH	-1.12	2.29	0.55	-	0.73	0.33	0.16	-	0.90	0.27	-0.62
			H ₂ O	-1.59	2.61	0.68	-	-	-	-	-			
	100T	6.33	OH	-0.69	2.27	0.54	-	0.94	0.32	0.16	-	1.10	0.53	-0.57
			H ₂ O	-1.52	2.62	0.60	-	-	-	-	-			
	111E	5.00	OH	-1.29	2.29	0.48	-0.06	0.67	0.32	0.12	-0.05	0.81	0.11	-0.70
			H ₂ O	-1.66	2.62	0.62	-	-	-	-	-			
	111TC	7.50	OH	-0.67	2.29	0.48	-	0.91	0.32	0.11	-0.05	1.06	0.45	-0.62
		H ₂ O	-1.30	2.61	0.54	-0.08	-	-	-	-				
	111T M	6.92	OH	-0.92	2.28	0.50	-	0.86	0.33	0.15	-	1.04	0.39	-0.65
			H ₂ O	-1.56	2.62	0.58	-	-	-	-	-			
	kink	4.25	OH	-1.05	2.29	0.48	-0.06	0.59	0.32	0.16	-	0.75	0.13	-0.62
			H ₂ O	-1.37	2.59	0.66	-	-	-	-	-			

a: Vibrational data obtained using denser plane wave sampling, as described in S.2.

x: No stable configuration available for this adsorption site.

Table S.3. Quality of linear relations of OH formation energies between different metals and adsorption site coordination numbers.

Excluding 38–111T, 201–100T, MS^a			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b	
<i>MS</i>	Ag	<i>R</i>	0.93		0.98	0.96	0.91	0.93	–	
		<i>max</i> $\Delta G_y x$	0.11	–	0.11	0.12	0.16	0.19	0.27	
		<i>ave</i> $\Delta G_y x$	0.05		0.04	0.06	0.09	0.07	0.13	
	Au	<i>R</i>	0.90	0.95		0.94	0.90	0.94	–	
		<i>max</i> $\Delta G_y x$	0.13	0.10	–	0.14	0.19	0.16	0.31	
		<i>ave</i> $\Delta G_y x$	0.07	0.04		0.08	0.09	0.08	0.17	
	Ir	<i>R</i>	0.99	0.92	0.89		0.96	0.96	–	
		<i>max</i> $\Delta G_y x$	0.09	0.17	0.23	–	0.26	0.21	0.42	
		<i>ave</i> $\Delta G_y x$	0.04	0.10	0.12		0.09	0.08	0.28	
	Pd	<i>R</i>	0.95	0.83	0.81	0.93		0.95	–	
		<i>max</i> $\Delta G_y x$	0.08	0.12	0.15	0.12	–	0.04	0.22	
		<i>ave</i> $\Delta G_y x$	0.04	0.07	0.06	0.04		0.03	0.14	
	Pt	<i>R</i>	0.95	0.87	0.89	0.93	0.90		–	
		<i>max</i> $\Delta G_y x$	0.11	0.16	0.18	0.13	0.07	–	0.40	
		<i>ave</i> $\Delta G_y x$	0.06	0.10	0.08	0.07	0.05		0.21	
Excluding 38–111T, 201–100T, <i>vac</i>			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b	
<i>vac</i>	Ag	<i>R</i>	0.66		0.94	0.66	0.60	0.76	–	
		<i>max</i> $\Delta G_y x$	0.38	–	0.13	0.37	0.42	0.32	0.22	
		<i>ave</i> $\Delta G_y x$	0.11		0.07	0.14	0.13	0.10	0.07	
	Au	<i>R</i>	0.86	0.97		0.88	0.82	0.88	–	
		<i>max</i> $\Delta G_y x$	0.34	0.18	–	0.31	0.39	0.28	0.23	
		<i>ave</i> $\Delta G_y x$	0.09	0.07		0.10	0.09	0.10	0.15	
	Ir	<i>R</i>	0.98	0.81	0.94		0.95	0.95	–	
		<i>max</i> $\Delta G_y x$	0.14	0.48	0.32	–	0.23	0.36	0.55	
		<i>ave</i> $\Delta G_y x$	0.06	0.29	0.18		0.11	0.10	0.39	
	Pd	<i>R</i>	0.96	0.77	0.90	0.98		0.90	–	
		<i>max</i> $\Delta G_y x$	0.06	0.13	0.11	0.07	–	0.09	0.16	
		<i>ave</i> $\Delta G_y x$	0.03	0.08	0.06	0.03		0.04	0.11	
	Pt	<i>R</i>	0.97	0.87	0.94	0.97	0.95		–	
		<i>max</i> $\Delta G_y x$	0.10	0.22	0.16	0.16	0.15	–	0.34	
		<i>ave</i> $\Delta G_y x$	0.04	0.10	0.09	0.06	0.08		0.17	
	Excluding 38 sites, 201–100T, <i>MS</i>			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b
	<i>MS</i>	Ag	<i>R</i>	0.94		0.99	0.97	0.92	0.96	–
			<i>max</i> $\Delta G_y x$	0.08	–	0.06	0.09	0.13	0.09	0.27
<i>ave</i> $\Delta G_y x$			0.04		0.03	0.05	0.07	0.05	0.13	
Au		<i>R</i>	0.88	0.97		0.93	0.89	0.94	–	
		<i>max</i> $\Delta G_y x$	0.13	0.07	–	0.14	0.18	0.14	0.31	
		<i>ave</i> $\Delta G_y x$	0.07	0.03		0.08	0.10	0.07	0.15	
Ir		<i>R</i>	0.98	0.93	0.86		0.96	0.97	–	
		<i>max</i> $\Delta G_y x$	0.09	0.16	0.23	–	0.26	0.20	0.42	
		<i>ave</i> $\Delta G_y x$	0.05	0.09	0.13		0.09	0.07	0.27	
Pd		<i>R</i>	0.94	0.84	0.79	0.92		0.95	–	
		<i>max</i> $\Delta G_y x$	0.09	0.11	0.15	0.12	–	0.05	0.22	
		<i>ave</i> $\Delta G_y x$	0.04	0.06	0.07	0.04		0.03	0.13	
Pt		<i>R</i>	0.96	0.92	0.89	0.93	0.90		–	
		<i>max</i> $\Delta G_y x$	0.11	0.12	0.18	0.13	0.07	–	0.40	
		<i>ave</i> $\Delta G_y x$	0.05	0.08	0.09	0.06	0.05		0.19	

Excluding 38 sites, 201–100T, <i>vac</i>			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b
<i>vac</i>	Ag	<i>R</i>	0.78		0.80	0.77	0.79	0.87	–
		<i>max</i> $\Delta G_y x$	0.10	–	0.09	0.09	0.09	0.10	0.22
		<i>ave</i> $\Delta G_y x$	0.06		0.05	0.06	0.06	0.04	0.07
	Au	<i>R</i>	0.98	0.89		0.99	0.98	0.95	–
		<i>max</i> $\Delta G_y x$	0.05	0.18	–	0.04	0.06	0.12	0.23
		<i>ave</i> $\Delta G_y x$	0.03	0.08		0.02	0.03	0.04	0.15
	Ir	<i>R</i>	0.98	0.88	0.99		0.95	0.93	–
		<i>max</i> $\Delta G_y x$	0.14	0.49	0.10	–	0.24	0.36	0.55
		<i>ave</i> $\Delta G_y x$	0.07	0.20	0.05		0.10	0.11	0.39
	Pd	<i>R</i>	0.95	0.89	0.99	0.98		0.90	–
		<i>max</i> $\Delta G_y x$	0.06	0.12	0.05	0.06	–	0.09	0.16
		<i>ave</i> $\Delta G_y x$	0.03	0.06	0.02	0.03		0.04	0.11
	Pt	<i>R</i>	0.97	0.93	0.97	0.97	0.95		–
		<i>max</i> $\Delta G_y x$	0.10	0.21	0.13	0.16	0.13	–	0.34
		<i>ave</i> $\Delta G_y x$	0.04	0.07	0.05	0.06	0.07		0.17
Excluding 38, 79 sites, 201–100T, <i>MS</i>			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b
<i>MS</i>	Ag	<i>R</i>	0.99		0.99	0.98	0.96	0.98	–
		<i>max</i> $\Delta G_y x$	0.04	–	0.05	0.07	0.10	0.07	0.28
		<i>ave</i> $\Delta G_y x$	0.02		0.02	0.05	0.06	0.04	0.16
	Au	<i>R</i>	0.98	0.99		0.97	0.98	0.99	–
		<i>max</i> $\Delta G_y x$	0.06	0.05	–	0.10	0.08	0.04	0.33
		<i>ave</i> $\Delta G_y x$	0.04	0.02		0.07	0.06	0.03	0.19
	Ir	<i>R</i>	0.98	0.96	0.94		0.96	0.98	–
		<i>max</i> $\Delta G_y x$	0.07	0.15	0.16	–	0.21	0.15	0.39
		<i>ave</i> $\Delta G_y x$	0.05	0.08	0.08		0.09	0.07	0.29
	Pd	<i>R</i>	0.94	0.92	0.96	0.92		0.99	–
		<i>max</i> $\Delta G_y x$	0.08	0.09	0.06	0.09	–	0.03	0.18
		<i>ave</i> $\Delta G_y x$	0.03	0.04	0.03	0.04		0.02	0.12
	Pt	<i>R</i>	0.98	0.97	0.99	0.96	0.98		–
		<i>max</i> $\Delta G_y x$	0.09	0.11	0.05	0.12	0.05	–	0.35
		<i>ave</i> $\Delta G_y x$	0.04	0.05	0.03	0.07	0.03		0.22
Excluding 38, 79 sites, 201–100T, <i>MS</i>			\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b
<i>MS</i>	Ag	<i>R</i>	0.98		0.96	0.98	0.94	0.99	–
		<i>max</i> $\Delta G_y x$	0.03	–	0.05	0.04	0.08	0.03	0.23
		<i>ave</i> $\Delta G_y x$	0.02		0.04	0.03	0.04	0.02	0.11
	Au	<i>R</i>	0.99	0.98		1.00	0.99	0.99	–
		<i>max</i> $\Delta G_y x$	0.03	0.07	–	0.03	0.04	0.03	0.24
		<i>ave</i> $\Delta G_y x$	0.02	0.04		0.02	0.02	0.02	0.16
	Ir	<i>R</i>	1.00	0.99	1.00		0.99	0.99	–
		<i>max</i> $\Delta G_y x$	0.05	0.12	0.06	–	0.08	0.07	0.58
		<i>ave</i> $\Delta G_y x$	0.03	0.08	0.04		0.06	0.04	0.42
	Pd	<i>R</i>	0.98	0.97	1.00	0.99		0.98	–
		<i>max</i> $\Delta G_y x$	0.04	0.06	0.03	0.03	–	0.04	0.16
		<i>ave</i> $\Delta G_y x$	0.02	0.03	0.01	0.02		0.02	0.11
	Pt	<i>R</i>	1.00	0.99	0.99	1.00	0.99		–
		<i>max</i> $\Delta G_y x$	0.01	0.05	0.04	0.04	0.08	–	0.31
		<i>ave</i> $\Delta G_y x$	0.01	0.03	0.03	0.02	0.04		0.21

a: All ΔG_y in this table are in eV.

b: Null hypothesis, in which deviations are relative to the average as opposed to linear regression.

Table S.4. Parameters for linear regressions of OH formation energies between different metals and adsorption site coordination numbers.

	Metal y	Parameters ^a	\overline{CN}	Metal x					
				Ag	Au	Ir	Pd	Pt	
Excluding 38–111T, 201–100T	MS	Ag	a	0.13	–	0.91	0.55	1.07	0.72
			b	–0.67	–	–0.46	0.23	–0.19	–0.08
		Au	a	0.13	1.00	–	0.56	1.10	0.77
			b	–0.17	0.51	–	0.74	0.31	0.43
		Ir	a	0.23	1.53	1.40	–	1.97	1.28
			b	–1.59	–0.40	–1.11	–	–0.77	–0.55
		Pd	a	0.10	0.65	0.60	0.43	–	0.62
			b	–0.35	0.19	–0.12	0.37	–	0.12
		Pt	a	0.16	1.05	1.02	0.68	1.50	–
			b	–0.72	0.12	–0.40	0.39	–0.17	–
	vac	Ag	a	0.12	–	0.80	0.36	1.22	0.85
			b	0.35	–	0.01	0.91	–0.08	0.36
		Au	a	0.18	1.09	–	0.56	1.85	1.08
			b	0.27	0.14	–	1.09	–0.39	0.42
Ir	a	0.33	1.22	1.39	–	3.42	1.85		
	b	–1.50	–0.98	–1.45	–	–2.76	–1.16		
Pd	a	0.09	0.29	0.36	0.27	–	0.49		
	b	0.41	0.58	0.44	0.81	–	0.51		
Pt	a	0.17	0.67	0.71	0.48	1.66	–		
	b	–0.12	0.07	–0.12	0.64	–0.70	–		
Excluding 38 sites, 201–100T	MS	Ag	a	0.12	–	0.81	0.49	0.91	0.63
			b	–0.58	–	–0.39	0.22	–0.14	–0.04
		Au	a	0.13	1.17	–	0.54	1.03	0.73
			b	–0.15	0.49	–	0.74	0.34	0.45
		Ir	a	0.23	1.78	1.38	–	1.91	1.22
			b	–1.60	–0.44	–1.10	–	–0.75	–0.52
		Pd	a	0.11	0.77	0.60	0.44	–	0.61
			b	–0.37	0.17	–0.12	0.37	–	0.12
		Pt	a	0.17	1.35	1.08	0.71	1.54	–
			b	–0.79	0.08	–0.44	0.39	–0.18	–
	vac	Ag	a	0.07	–	0.51	0.19	0.73	0.48
			b	0.70	–	0.40	1.02	0.42	0.69
		Au	a	0.14	1.24	–	0.41	1.41	0.80
			b	0.57	–0.02	–	1.19	0.05	0.69
Ir	a	0.32	3.14	2.37	–	3.26	1.89		
	b	–1.47	–3.08	–2.81	–	–2.60	–1.19		
Pd	a	0.09	0.86	0.68	0.28	–	0.53		
	b	0.39	–0.03	0.00	0.81	–	0.48		
Pt	a	0.16	1.59	1.12	0.46	1.54	–		
	b	–0.08	–0.92	–0.69	0.65	–0.58	–		
Excluding 38, 79 sites, 201–100T	MS	Ag	a	0.14	–	0.75	0.46	0.81	0.53
			b	–0.73	–	–0.32	0.18	–0.17	–0.07
		Au	a	0.15	1.15	–	0.44	0.75	0.51
			b	–0.37	0.44	–	0.63	0.30	0.38
		Ir	a	0.25	1.71	1.07	–	1.84	1.13
			b	–1.70	–0.35	–0.78	–	–0.79	–0.54
		Pd	a	0.11	0.76	0.46	0.47	–	0.58
			b	–0.36	0.25	0.07	0.41	–	0.14
		Pt	a	0.21	1.43	0.90	0.82	1.67	–
			b	–0.93	0.18	–0.19	0.47	–0.23	–
	vac	Ag	a	0.05	–	0.76	0.07	–0.25	0.05
			b	0.68	–	0.08	0.93	1.19	0.92
		Au	a	0.14	1.21	–	0.45	1.43	0.79
			b	0.51	0.02	–	1.18	0.03	0.67
Ir	a	0.33	0.21	2.22	–	2.74	1.78		
	b	–1.55	0.18	–2.62	–	–2.17	–1.17		
Pd	a	0.11	–0.10	0.69	0.34	–	0.59		
	b	0.32	1.02	–0.01	0.80	–	0.43		
Pt	a	0.19	0.05	1.24	0.56	1.60	–		
	b	–0.23	0.79	–0.82	0.66	–0.64	–		

a: Parameters for linear regressions $\overline{\Delta G}_y = a\overline{\Delta G}_x + b$, for which $\overline{\Delta G}_y$, $\overline{\Delta G}_x$, a , and b are in eV.

Table S.5. Quality of linear relations between formation site binding energies.

	Site (\overline{CN})	Parameters ^a	MS, 38		MS, 79			MS, 201					
			111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink
MS, 38	111 (6.00)	R		0.84	0.65	0.61	0.71	0.84	0.79	0.77	0.60	0.68	0.86
		$\max \Delta G_y x$	–	0.25	0.36	0.35	0.33	0.26	0.21	0.29	0.39	0.34	0.22
		$\text{ave } \Delta G_y x$		0.10	0.16	0.17	0.15	0.10	0.14	0.14	0.15	0.14	0.10
MS, 79	111E (5.00)	R	0.84		0.94	0.97	0.99	0.95	0.49	0.98	0.95	0.93	0.94
		$\max \Delta G_y x$	0.32	–	0.16	0.16	0.16	0.06	0.35	0.14	0.22	0.13	0.08
		$\text{ave } \Delta G_y x$	0.15		0.11	0.12	0.09	0.02	0.20	0.07	0.09	0.09	0.04
MS, 79	111T (6.67)	R	0.61	0.92	0.97		0.96	0.92	0.50	0.94	0.96	0.96	0.91
		$\max \Delta G_y x$	0.35	0.14	0.09	–	0.10	0.14	0.37	0.11	0.13	0.12	0.15
		$\text{ave } \Delta G_y x$	0.14	0.08	0.04		0.06	0.09	0.16	0.07	0.05	0.05	0.09
MS, 201	100E (5.17)	R	0.71	0.95	0.99	0.96		0.97	0.52	1.00	0.93	0.92	0.97
		$\max \Delta G_y x$	0.41	0.16	0.05	0.15	–	0.10	0.45	0.05	0.16	0.24	0.14
		$\text{ave } \Delta G_y x$	0.21	0.10	0.03	0.09		0.08	0.26	0.03	0.11	0.11	0.07
MS, 201	100T (6.33)	R	0.84	1.00	0.95	0.92	0.97		0.71	0.98	0.93	0.94	0.99
		$\max \Delta G_y x$	0.31	0.06	0.15	0.18	0.10	–	0.34	0.09	0.21	0.15	0.06
		$\text{ave } \Delta G_y x$	0.15	0.02	0.10	0.12	0.08		0.21	0.06	0.10	0.10	0.03
MS, 201	111E (5.00)	R	0.79	0.75	0.49	0.50	0.52	0.71		0.57	0.64	0.71	0.68
		$\max \Delta G_y x$	0.22	0.20	0.26	0.27	0.26	0.21	–	0.25	0.20	0.20	0.24
		$\text{ave } \Delta G_y x$	0.09	0.12	0.17	0.17	0.16	0.12		0.15	0.15	0.14	0.12
MS, 201	111TC (7.50)	R	0.77	0.97	0.98	0.94	1.00	0.98	0.57		0.92	0.92	0.99
		$\max \Delta G_y x$	0.31	0.12	0.09	0.14	0.05	0.08	0.34	–	0.14	0.18	0.07
		$\text{ave } \Delta G_y x$	0.16	0.06	0.04	0.09	0.02	0.05	0.21		0.10	0.10	0.04
MS, 201	111TM (6.92)	R	0.60	0.94	0.95	0.96	0.93	0.93	0.64	0.92		0.98	0.90
		$\max \Delta G_y x$	0.35	0.16	0.12	0.14	0.13	0.16	0.36	0.12	–	0.07	0.18
		$\text{ave } \Delta G_y x$	0.17	0.07	0.06	0.06	0.08	0.08	0.17	0.09		0.04	0.10
vac, 38	111 (6.00)	R	0.68	0.96	0.93	0.96	0.92	0.94	0.71	0.92	0.98		0.92
		$\max \Delta G_y x$	0.28	0.08	0.15	0.11	0.16	0.10	0.29	0.15	0.06	–	0.11
		$\text{ave } \Delta G_y x$	0.13	0.06	0.06	0.05	0.07	0.07	0.12	0.08	0.03		0.08
vac, 38	kink (4.00)	R	0.86	0.99	0.94	0.91	0.97	0.99	0.68	0.99	0.90	0.92	
		$\max \Delta G_y x$	0.26	0.06	0.17	0.17	0.13	0.05	0.30	0.08	0.23	0.15	–
		$\text{ave } \Delta G_y x$	0.12	0.04	0.09	0.11	0.07	0.02	0.19	0.04	0.11	0.11	
vac, 38	111 (6.00)	R		0.61	0.38	0.61	0.42	0.57	0.60	0.50	0.32	0.56	0.63
		$\max \Delta G_y x$	–	0.25	0.33	0.25	0.32	0.27	0.28	0.29	0.36	0.29	0.23
		$\text{ave } \Delta G_y x$		0.18	0.18	0.18	0.18	0.18	0.15	0.18	0.18	0.17	0.17
vac, 79	111E (5.00)	R	0.93		0.81	0.73	0.87	0.93	0.67	0.90	0.73	0.75	0.94
		$\max \Delta G_y x$	0.17	–	0.32	0.33	0.27	0.18	0.38	0.22	0.37	0.30	0.17
		$\text{ave } \Delta G_y x$	0.11		0.17	0.22	0.15	0.12	0.24	0.13	0.21	0.21	0.11
vac, 79	111T (6.67)	R	0.56	0.81		0.83	0.94	0.85	0.29	0.92	0.81	0.74	0.85
		$\max \Delta G_y x$	0.35	0.37	–	0.36	0.23	0.32	0.48	0.25	0.37	0.42	0.31
		$\text{ave } \Delta G_y x$	0.27	0.17		0.14	0.09	0.15	0.30	0.10	0.15	0.18	0.15
vac, 79	kink (4.00)	R	0.37	0.72	0.90		0.87	0.74	0.12	0.83	0.80	0.77	0.75
		$\max \Delta G_y x$	0.41	0.24	0.17	–	0.18	0.23	0.43	0.19	0.27	0.25	0.22
		$\text{ave } \Delta G_y x$	0.19	0.15	0.09		0.11	0.15	0.19	0.12	0.11	0.13	0.15
vac, 201	100E (5.17)	R	0.64	0.79	0.88	0.76		0.84	0.28	0.90	0.72	0.67	0.85
		$\max \Delta G_y x$	0.51	0.55	0.40	0.53	–	0.49	0.68	0.40	0.54	0.60	0.48
		$\text{ave } \Delta G_y x$	0.34	0.22	0.19	0.24		0.20	0.39	0.16	0.28	0.27	0.19
vac, 201	100T (6.33)	R	0.08	0.52	0.88	0.82	0.89		0.39	0.85	0.61	0.55	0.65
		$\max \Delta G_y x$	0.31	0.23	0.13	0.16	0.14	–	0.31	0.16	0.26	0.25	0.18
		$\text{ave } \Delta G_y x$	0.15	0.16	0.07	0.10	0.07		0.15	0.09	0.13	0.15	0.15
vac, 201	111E (5.00)	R	0.80	0.80	0.50	0.72	0.51	0.73		0.57	0.73	0.89	0.70
		$\max \Delta G_y x$	0.21	0.23	0.32	0.27	0.32	0.27	–	0.31	0.24	0.16	0.27
		$\text{ave } \Delta G_y x$	0.11	0.12	0.16	0.13	0.16	0.13		0.16	0.12	0.09	0.14
vac, 201	111TC (7.50)	R	0.62	0.84	0.94	0.85	0.96	0.88	0.32		0.82	0.76	0.89
		$\max \Delta G_y x$	0.37	0.36	0.24	0.35	0.22	0.32	0.50	–	0.36	0.43	0.30
		$\text{ave } \Delta G_y x$	0.28	0.17	0.10	0.14	0.09	0.15	0.31		0.17	0.18	0.15
vac, 201	111TM (6.92)	R	0.34	0.75	0.93	0.88	0.90	0.78	0.19	0.86		0.78	0.76
		$\max \Delta G_y x$	0.34	0.22	0.14	0.18	0.17	0.22	0.35	0.20	–	0.22	0.24
		$\text{ave } \Delta G_y x$	0.19	0.13	0.07	0.08	0.08	0.12	0.20	0.10		0.13	0.12
vac, 201	111TM (6.92)	R	0.54	0.91	0.99	0.99	0.97	0.91	0.49	0.95	0.98		0.89
		$\max \Delta G_y x$	0.40	0.16	0.07	0.05	0.11	0.16	0.44	0.14	0.10	–	0.19
		$\text{ave } \Delta G_y x$	0.20	0.11	0.04	0.02	0.06	0.11	0.22	0.08	0.05		0.11

	Site ($C\bar{N}$)	Parameters ^a	MS, 38		MS, 79			MS, 201					
			111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink
vac, 201	kink (4.08)	R	0.67	0.82	0.90	0.78	0.93	0.87	0.35	0.92	0.76	0.71	
		$\max \Delta G_y x$	0.45	0.49	0.34	0.47	0.32	0.44	0.64	0.34	0.48	0.56	–
		$\text{ave } \Delta G_y x$	0.32	0.20	0.17	0.22	0.13	0.17	0.36	0.14	0.25	0.25	

	Site ($C\bar{N}$)	Parameters ^a	vac, 38		vac, 79			vac, 201					
			111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink
MS, 38	111 (6.00)	R		0.93	0.56	0.37	0.64	0.08	0.80	0.62	0.34	0.54	0.67
		$\max \Delta G_y x$	–	0.13	0.38	0.39	0.33	0.27	0.20	0.35	0.42	0.37	0.33
		$\text{ave } \Delta G_y x$		0.08	0.18	0.20	0.17	0.14	0.16	0.17	0.19	0.20	0.16
	kink (4.00)	R	0.61		0.81	0.72	0.79	0.52	0.80	0.84	0.75	0.91	0.82
		$\max \Delta G_y x$	0.36	–	0.35	0.32	0.38	0.32	0.27	0.33	0.33	0.20	0.36
		$\text{ave } \Delta G_y x$	0.19		0.17	0.21	0.18	0.16	0.14	0.16	0.20	0.15	0.16
MS, 79	111E (5.00)	R	0.38	0.81		0.90	0.88	0.88	0.50	0.94	0.93	0.99	0.90
		$\max \Delta G_y x$	0.36	0.36	–	0.23	0.26	0.18	0.31	0.21	0.21	0.09	0.24
		$\text{ave } \Delta G_y x$	0.18	0.15		0.12	0.12	0.10	0.15	0.09	0.11	0.05	0.11
	111T (6.67)	R	0.61	0.73	0.83		0.76	0.82	0.72	0.85	0.88	0.99	0.78
		$\max \Delta G_y x$	0.16	0.30	0.23	–	0.24	0.19	0.13	0.22	0.19	0.04	0.23
		$\text{ave } \Delta G_y x$	0.08	0.13	0.10		0.11	0.10	0.07	0.09	0.09	0.02	0.10
kink (4.08)	R	0.42	0.87	0.94	0.87		0.89	0.51	0.96	0.90	0.97	0.93	
	$\max \Delta G_y x$	0.39	0.31	0.22	0.28	–	0.16	0.34	0.19	0.26	0.14	0.22	
	$\text{ave } \Delta G_y x$	0.19	0.12	0.09	0.14		0.08	0.18	0.08	0.12	0.08	0.09	
MS, 201	100E (5.17)	R	0.57	0.93	0.85	0.74	0.84		0.73	0.88	0.78	0.91	0.87
		$\max \Delta G_y x$	0.39	0.21	0.30	0.34	0.33	–	0.31	0.28	0.33	0.21	0.31
		$\text{ave } \Delta G_y x$	0.19	0.10	0.15	0.20	0.16	0.15	0.15	0.14	0.18	0.15	0.14
	100T (6.33)	R	0.60	0.67	0.29	0.12	0.28	0.39		0.32	0.19	0.49	0.35
		$\max \Delta G_y x$	0.31	0.25	0.28	0.27	0.29	0.30	–	0.28	0.26	0.20	0.29
		$\text{ave } \Delta G_y x$	0.15	0.11	0.17	0.19	0.17	0.15		0.17	0.18	0.16	0.16
	111E (5.00)	R	0.50	0.90	0.92	0.83	0.90	0.85	0.57		0.86	0.95	0.92
		$\max \Delta G_y x$	0.33	0.23	0.20	0.25	0.22	0.15	0.29	–	0.25	0.16	0.20
		$\text{ave } \Delta G_y x$	0.17	0.09	0.09	0.13	0.10	0.08	0.16		0.12	0.09	0.08
	111TC (7.50)	R	0.32	0.73	0.81	0.80	0.72	0.61	0.73	0.82		0.98	0.76
		$\max \Delta G_y x$	0.23	0.29	0.27	0.27	0.27	0.23	0.16	0.25	–	0.10	0.26
		$\text{ave } \Delta G_y x$	0.13	0.15	0.11	0.12	0.15	0.15	0.11	0.12		0.05	0.14
111TM (6.92)	R	0.56	0.75	0.74	0.77	0.67	0.55	0.89	0.76	0.78		0.71	
	$\max \Delta G_y x$	0.19	0.23	0.27	0.21	0.28	0.24	0.10	0.26	0.23	–	0.27	
	$\text{ave } \Delta G_y x$	0.09	0.12	0.11	0.12	0.12	0.12	0.06	0.11	0.12		0.11	
kink (4.08)	R	0.63	0.94	0.85	0.75	0.85	0.65	0.70	0.89	0.76	0.89		
	$\max \Delta G_y x$	0.33	0.18	0.25	0.29	0.28	0.22	0.27	0.23	0.32	0.22	–	
	$\text{ave } \Delta G_y x$	0.17	0.08	0.13	0.17	0.13	0.11	0.16	0.11	0.15	0.13		

	Site ($C\bar{N}$)	Parameters ^a	vac, 38		vac, 79			vac, 201					
			111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink
vac, 38	111 (6.00)	R		0.60	0.06	0.22	0.19	0.06	0.75	0.18	0.23	0.54	0.21
		$\max \Delta G_y x$	–	0.24	0.39	0.36	0.37	0.36	0.23	0.38	0.38	0.25	0.37
		$\text{ave } \Delta G_y x$		0.17	0.20	0.18	0.19	0.24	0.11	0.19	0.19	0.16	0.19
	kink (4.00)	R	0.60		0.81	0.56	0.86	0.29	0.62	0.84	0.60	0.72	0.88
		$\max \Delta G_y x$	0.48	–	0.36	0.46	0.27	0.20	0.42	0.31	0.46	0.40	0.27
		$\text{ave } \Delta G_y x$	0.24		0.18	0.24	0.16	0.10	0.26	0.17	0.22	0.24	0.14
vac, 79	111E (5.00)	R	0.06	0.81		0.83	0.98	0.95	0.11	1.00	0.93	0.98	0.98
		$\max \Delta G_y x$	0.48	0.30	–	0.31	0.11	0.10	0.46	0.06	0.17	0.09	0.11
		$\text{ave } \Delta G_y x$	0.24	0.18		0.16	0.06	0.05	0.23	0.03	0.11	0.07	0.04
	111T (6.67)	R	0.22	0.56	0.83		0.77	0.99	0.11	0.85	0.92	0.93	0.76
		$\max \Delta G_y x$	0.15	0.35	0.19	–	0.26	0.06	0.14	0.19	0.15	0.16	0.25
		$\text{ave } \Delta G_y x$	0.09	0.15	0.12		0.14	0.03	0.10	0.12	0.09	0.08	0.14
kink (4.08)	R	0.19	0.86	0.98	0.77		0.93	0.13	0.99	0.85	0.93	1.00	
	$\max \Delta G_y x$	0.67	0.34	0.15	0.48	–	0.14	0.67	0.11	0.35	0.29	0.07	
	$\text{ave } \Delta G_y x$	0.34	0.22	0.09	0.27		0.07	0.37	0.07	0.22	0.15	0.03	
vac, 201	100E (5.17)	R	0.06	0.29	0.95	0.99	0.93		0.73	0.99	0.89	0.94	0.96
		$\max \Delta G_y x$	0.15	0.28	0.09	0.05	0.11	–	0.10	0.04	0.14	0.10	0.09
		$\text{ave } \Delta G_y x$	0.10	0.18	0.06	0.03	0.06	0.15	0.06	0.02	0.07	0.06	0.04
	100T (6.33)	R	0.75	0.62	0.11	0.11	0.13	0.73		0.17	0.08	0.99	0.20
		$\max \Delta G_y x$	0.25	0.30	0.29	0.26	0.30	0.22	–	0.31	0.24	0.03	0.31
		$\text{ave } \Delta G_y x$	0.13	0.15	0.22	0.22	0.21	0.15		0.21	0.22	0.02	0.21
	111E (5.00)	R	0.18	0.84	1.00	0.85	0.99	0.99	0.17		0.92	0.97	0.99
		$\max \Delta G_y x$	0.49	0.27	0.06	0.33	0.10	0.04	0.48	–	0.23	0.15	0.09
$\text{ave } \Delta G_y x$		0.25	0.18	0.03	0.18	0.05	0.02	0.25		0.12	0.08	0.05	
111TC (7.50)	R	0.23	0.60	0.93	0.92	0.85	0.89	0.08	0.92		0.99	0.86	
	$\max \Delta G_y x$	0.22	0.28	0.11	0.12	0.18	0.13	0.21	0.15	–	0.06	0.17	
	$\text{ave } \Delta G_y x$	0.11	0.14	0.07	0.08	0.09	0.07	0.12	0.07		0.03	0.09	

Site (\bar{CN})	Parameters ^a	vac, 38		vac, 79			vac, 201						
		111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink	
111TM (6.92)	<i>R</i>	0.54	0.72	0.98	0.93	0.93	0.94	0.99	0.97	0.99			0.93
	<i>max</i> $\Delta G_y x$	0.17	0.32	0.07	0.16	0.17	0.10	0.03	0.11	0.07	–		0.15
	<i>ave</i> $\Delta G_y x$	0.11	0.16	0.06	0.08	0.09	0.07	0.02	0.06	0.04			0.10
kink (4.08)	<i>R</i>	0.21	0.88	0.98	0.76	1.00	0.96	0.20	0.99	0.86	0.93		
	<i>max</i> $\Delta G_y x$	0.67	0.29	0.14	0.49	0.06	0.09	0.65	0.10	0.33	0.26	–	
	<i>ave</i> $\Delta G_y x$	0.33	0.19	0.06	0.27	0.03	0.04	0.33	0.06	0.20	0.16		

a: All ΔG_y are in eV.

S.5. Statistical treatment of bivariate normal distributions

We modeled formation energies for each metal M using bivariate normal distributions. These distributions are described in Equation S.10, with $x = \Delta G_{f,MS}$ and $y = \Delta G_{f,vac}$:

$$P_M(x, y) = \frac{1}{2\pi\sigma_{M1}\sigma_{M2}} \exp\left(-\frac{(\cos\theta_M(x - \bar{x}_M) + \sin\theta_M(y - \bar{y}_M))^2}{2\sigma_{M1}^2} - \frac{(-\sin\theta_M(x - \bar{x}_M) + \cos\theta_M(y - \bar{y}_M))^2}{2\sigma_{M2}^2}\right) \quad (\text{S.10})$$

where \bar{x}_M and \bar{y}_M are the mean formation energies for each metal, θ_M is the angle along which the first component of the distribution is oriented (perpendicular to the second component), and σ_{M1} and σ_{M2} are the standard deviations of respective components. θ_M is defined using the net orientation of the known x , y of the metal, so:

$$\theta_M = \tan^{-1}\left(2\frac{\overline{(x - \bar{x})(y - \bar{y})}}{\overline{(x - \bar{x})^2} - \overline{(y - \bar{y})^2}}\right) \quad (\text{S.11})$$

Subsequently, σ_{M1} and σ_{M2} are optimized such that $\prod_{site_M} P_M(x_{site_M}, y_{site_M})$ is maximized, ensuring the bivariate normal distribution is closest to the observed distribution of formation energies.

The equally weighted average of resulting P_M for Au, Ir, Pd, and Pt is compared to the linear fit of all \bar{y}_M with respect to \bar{x}_M . We obtained the correlation coefficient of the distributions with respect to this linear fit by analytically evaluating the expression in Equation S.12:

$$R_{dist} = \sqrt{1 - \frac{\sum_M \iint_{-\infty}^{\infty} (y - ax - b)^2 P_M(x, y) dx dy}{\sum_M \iint_{-\infty}^{\infty} (y - \bar{y})^2 P_M(x, y) dx dy}} \quad (\text{S.12})$$

Here, $ax + b$ is the equation corresponding to the linear fit of \bar{y}_M with respect to \bar{x}_M , and \bar{y} is the average across all \bar{y}_M . The corresponding MAE is calculated by numerically evaluating the integral in Equation S.13 using the 2D adaptive quadrature method implemented in GNU Octave 7.2.0:⁹

$$MAE = \sum_M \iint_{-\infty}^{\infty} |y - ax - b| P_M(x, y) dx dy \quad (S.13)$$

The P_M parameters and corresponding MAE are listed below in Table S.6.

Table S.6. Bivariate normal distribution parameters.

Metal	\bar{x}_M (eV)	\bar{y}_M (eV)	θ_M (°)	σ_1 (eV)	σ_2 (eV)	MAE (eV)
Ag	0.074	1.063	31.1	0.194	0.052	0.32
Au	0.574	1.328	42.8	0.254	0.123	0.15
Ir	-0.305	0.356	-35.3	0.065	0.303	0.09
Pd	0.231	0.904	39.8	0.188	0.076	0.09
Pt	0.178	0.806	42.2	0.311	0.128	0.16

S.6. Solvation energy statistics

Table S.7. Parameters for linear regressions of solvation energies between different metals and adsorption site coordination numbers.

	Metal y	Parameters ^a	\overline{CN}	Ag	Au	Ir	Pd	Pt
Excluding 38–111, 201–100T	Ag	a	0.02					
		b	-1.05	–	0.54	-0.14	0.02	0.90
	Au	a	-0.05	0.98				
		b	-0.44	0.26	–	0.33	-0.58	1.17
	Ir	a	-0.10	-0.25	0.30			
		b	-0.07	-0.85	-0.40	–	-0.25	0.64
	Pd	a	0.02	0.00	-0.06	-0.02		
		b	-0.76	-0.67	-0.72	-0.70	–	0.21
	Pt	a	0.00	0.40	0.26	0.14	0.44	
		b	-0.59	-0.23	-0.43	-0.54	-0.32	–
Excluding 38 sites, 201–100T	Ag	a	0.06					
		b	-1.30	–	-0.02	-0.45	0.23	0.18
	Au	a	-0.01	-0.02				
		b	-0.72	-0.76	–	-0.05	-0.22	-0.08
	Ir	a	-0.10	-2.09	-0.19			
		b	-0.09	-2.74	-0.78	–	-0.13	-0.04
	Pd	a	0.02	0.14	-0.10	-0.02		
		b	-0.76	-0.53	-0.75	-0.69	–	0.46
	Pt	a	0.01	0.14	-0.05	0.00	0.57	
		b	-0.71	-0.49	-0.67	-0.64	-0.25	–
Excluding 38, 79 sites, 201–100T	Ag	a	0.04					
		b	-1.25	–	0.66	-0.40	0.07	0.34
	Au	a	0.02	0.77				
		b	-0.93	0.01	–	-0.23	1.26	0.84
	Ir	a	-0.10	-2.45	-0.99			
		b	-0.05	-3.08	-1.39	–	0.56	-0.73
	Pd	a	0.00	0.03	0.36	0.03		
		b	-0.66	-0.61	-0.36	-0.63	–	0.55
	Pt	a	0.01	0.36	0.63	-0.07	1.45	
		b	-0.68	-0.24	-0.12	-0.68	0.32	–

a: Parameters for linear regressions $\overline{\Delta G}_y = a\Delta G_x + b$, for which $\overline{\Delta G}_y$, ΔG_x , a , and b are in eV.

Table S.8. Quality of linear relations between solvation energies on different metals.

	Metal y	Statistics ^a	\overline{CN}	Ag	Au	Ir	Pd	Pt	no trend ^b
Excluding 38–111T, 201–100T	Ag	R	0.16						–
		max $\Delta G_y x$	0.27	–	0.73	-0.19	0.01	0.60	0.49
		ave $\Delta G_y x$	0.08		0.14	0.29	0.25	0.13	0.13
	Au	R	-0.39	0.73					–
		max $\Delta G_y x$	0.31	0.19	–	0.31	-0.19	0.60	0.13
		ave $\Delta G_y x$	0.11	0.09	0.13	0.11	0.10	0.07	0.07
	Ir	R	-0.84	-0.19	0.31				–
		max $\Delta G_y x$	0.12	0.22	0.26	–	0.25	0.23	0.28
		ave $\Delta G_y x$	0.07	0.15	0.12	0.13	0.12	0.12	0.12
	Pd	R	0.38	0.01	-0.19	-0.08			–
		max $\Delta G_y x$	0.07	0.09	0.09	0.08	–	0.09	0.11
		ave $\Delta G_y x$	0.04	0.05	0.04	0.04	0.04	0.04	0.05
	Pt	R	-0.06	0.60	0.55	0.29	0.00		–
		max $\Delta G_y x$	0.13	0.11	0.14	0.12	0.14	–	0.19
		ave $\Delta G_y x$	0.06	0.05	0.05	0.06	0.05	0.05	0.06
	Metal y	Statistics ^a	\overline{CN}	Ag	Au	Ir	Pd	Pt	no trend ^b
	Ag	R	0.91	–	-0.02	-0.97	0.18	0.16	–

Excluding 38 sites, 201–100T		$max \Delta G_y x$	0.04		0.11	0.17	0.12	0.11	0.11
		$ave \Delta G_y x$	0.03		0.06	0.07	0.05	0.06	0.05
	Au	R	-0.09	-0.02		-0.10	-0.15	0.16	–
		$max \Delta G_y x$	0.12	0.11	–	0.12	0.12	0.12	0.13
		$ave \Delta G_y x$	0.07	0.07		0.08	0.07	0.07	0.07
		R	-0.81	-0.97	-0.10		-0.04	-0.01	–
	Ir	$max \Delta G_y x$	0.12	0.06	0.25	–	0.26	0.26	0.26
		$ave \Delta G_y x$	0.08	0.03	0.12		0.12	0.12	0.12
		R	0.36	0.18	-0.15	-0.04		0.00	–
		$max \Delta G_y x$	0.07	0.08	0.09	0.09	–	0.10	0.10
	Pd	$ave \Delta G_y x$	0.04	0.05	0.05	0.04		0.03	0.05
		R	0.29	0.16	-0.06	-0.01	0.00		–
Pt	$max \Delta G_y x$	0.09	0.10	0.10	0.09	0.09	–	0.10	
	$ave \Delta G_y x$	0.05	0.05	0.04	0.04	0.04		0.04	
			CN	Ag	Au	Ir	Pd	Pt	<i>no trend^b</i>
Excluding 38, 79 sites, 201–100T	Ag	R	0.91		0.71	-0.99	0.05	0.35	–
		$max \Delta G_y x$	0.03	–	0.07	0.16	0.09	0.08	0.49
		$ave \Delta G_y x$	0.02		0.03	0.09	0.04	0.05	0.13
		R	0.48	0.71		-0.47	0.67	0.35	–
	Au	$max \Delta G_y x$	0.09	0.07	–	0.08	0.08	0.08	0.13
		$ave \Delta G_y x$	0.05	0.04		0.06	0.04	0.04	0.07
		R	-0.87	-0.99	-0.47		0.14	-0.23	–
		$max \Delta G_y x$	0.11	0.03	0.22	–	0.22	0.24	0.28
	Ir	$ave \Delta G_y x$	0.06	0.02	0.11		0.11	0.12	0.12
		R	0.15	0.05	0.67	0.14		0.00	–
	Pd	$max \Delta G_y x$	0.06	0.06	0.04	0.06	–	0.03	0.11
		$ave \Delta G_y x$	0.03	0.03	0.02	0.03		0.01	0.05
	R	0.26	0.35	0.72	-0.23	0.00		–	
	$max \Delta G_y x$	0.08	0.09	0.06	0.07	0.04	–	0.19	
Pt	$ave \Delta G_y x$	0.04	0.05	0.03	0.04	0.02		0.06	

a: All ΔG_y are in eV.

b: Null hypothesis, in which deviations are relative to the average as opposed to linear regression.

Table S.9. Quality of linear relations between micro-solvated OH_x ($x = 1,2$) energies on different metals.

$\text{H}_2\text{O},$ MS	Ag	R	0.22	Ag	Au	Ir	Pd	<i>no trend^b</i>
		$max \Delta G_y x$	0.12	–	0.78	0.92	0.96	–
		$ave \Delta G_y x$	0.07		0.08	0.05	0.04	0.13
		R	0.48	0.78		0.86	0.95	–
	Au	$max \Delta G_y x$	0.14	0.11	–	0.10	0.11	0.17
		$ave \Delta G_y x$	0.08	0.05		0.04	0.04	0.08
		R	0.48	0.92	0.86		0.95	–
		$max \Delta G_y x$	0.33	0.19	0.23	–	0.15	0.53
	Ir	$ave \Delta G_y x$	0.21	0.09	0.12		0.06	0.20
		R	0.26	0.96	0.83	0.95		–
	Pd	$max \Delta G_y x$	0.19	0.08	0.14	0.08	–	0.20
		$ave \Delta G_y x$	0.11	0.03	0.06	0.03		0.11
$\text{OH},$ MS	Ag	R	0.82	Ag	Au	Ir	Pd	<i>no trend^b</i>
		$max \Delta G_y x$	0.18	–	0.98	0.96	0.90	–
		$ave \Delta G_y x$	0.09		0.08	0.10	0.16	0.40
		R	0.89	0.98		0.96	0.91	–
	Au	$max \Delta G_y x$	0.18	0.08	–	0.12	0.19	0.47
		$ave \Delta G_y x$	0.08	0.04		0.05	0.07	0.20
		R	0.85	0.96	0.96		0.97	–
		$max \Delta G_y x$	0.35	0.27	0.22	–	0.19	0.95
	Ir	$ave \Delta G_y x$	0.23	0.10	0.10		0.11	0.42
		R	0.75	0.90	0.91	0.97		–
	Pd	$max \Delta G_y x$	0.20	0.21	0.19	0.09	–	0.42
		$ave \Delta G_y x$	0.12	0.07	0.07	0.05		0.17

a: All ΔG_y are in eV.

b: Null hypothesis, in which deviations are relative to the average as opposed to linear regression.

Table S.10. Parameters for linear regressions between micro-solvated OH_x ($x = 1,2$) energies on different metals.

	Metal y	Parameters ^a	$\overline{\text{CN}}$	Ag	Au	Ir	Pd	
<i>H₂O, MS</i>	Ag	<i>a</i>	0.02	–	0.67	0.30	0.63	
		<i>b</i>	0.72		0.23	0.75	0.50	
	Au	<i>a</i>	0.04	0.91	–	0.33	0.63	
		<i>b</i>	0.61	0.12		0.79	0.55	
	Ir	<i>a</i>	0.10	2.75	2.23	–	1.87	
		<i>b</i>	–0.38	–2.02	–1.71		–0.71	
	Pd	<i>a</i>	0.03	1.47	1.09	0.49	–	
		<i>b</i>	0.33	–0.70	–0.45	0.39		
				$\overline{\text{CN}}$	Ag	Au	Ir	Pd
	<i>OH, MS</i>	Ag	<i>a</i>	0.13	–	0.81	0.40	0.81
<i>b</i>			0.14		–0.28	0.92	0.30	
Au		<i>a</i>	0.17	1.17	–	0.47	0.98	
		<i>b</i>	0.46	0.40		1.48	0.73	
Ir		<i>a</i>	0.34	2.35	1.95	–	2.12	
		<i>b</i>	–1.98	–2.18	–2.90		–1.62	
Pd		<i>a</i>	0.14	1.00	0.84	0.44	–	
		<i>b</i>	–0.04	–0.17	–0.49	0.76		

a: Parameters for linear regressions $\overline{\Delta G}_y = a\Delta G_x + b$, for which $\overline{\Delta G}_y$, ΔG_x , *a*, and *b* are in eV.

S.7. Bond lengths and statistics

Table S.11. Bond length statistics.

Metal	Species	Parameters ^a					
Ag	OH, vac	mean bond length			2.119	0.976	
		max deviation	–	–	0.121	0.001	–
		average deviation			0.037	0.001	
	OH, MS	mean bond length	1.053	1.506	2.253	0.983	2.016
		max deviation	0.008	0.035	0.042	0.003	0.092
		average deviation	0.005	0.020	0.023	0.002	0.048
	H ₂ O, MS	mean bond length	0.992	1.870		1.008	1.693
		max deviation	0.003	0.050	–	0.004	0.034
		average deviation	0.001	0.017		0.002	0.020
Au	OH, vac	mean bond length			2.094	0.977	
		max deviation	–	–	0.207	0.001	–
		average deviation			0.046	0.000	
	OH, MS	mean bond length	1.044	1.542	2.223	0.988	1.913
		max deviation	0.022	0.066	0.063	0.003	0.047
		average deviation	0.008	0.037	0.036	0.001	0.022
	H ₂ O, MS	mean bond length	0.994	1.863		1.009	1.686
		max deviation	0.006	0.081	–	0.005	0.038
		average deviation	0.001	0.021		0.003	0.019
Ir	OH, vac	mean bond length			1.966	0.979	
		max deviation	–	–	0.046	0.001	–
		average deviation			0.030	0.000	
	OH, MS	mean bond length	1.050	1.547	2.103	1.002	1.775
		max deviation	0.087	0.213	0.055	0.008	0.105
		average deviation	0.021	0.085	0.029	0.003	0.045
	H ₂ O, MS	mean bond length	1.002	1.845		1.019	1.640
		max deviation	0.008	0.161	–	0.010	0.066
		average deviation	0.004	0.059		0.004	0.030
Pd	OH, vac	mean bond length			1.974	0.979	
		max deviation	–	–	0.017	0.001	–
		average deviation			0.009	0.000	
	OH, MS	mean bond length	1.044	1.538	2.105	0.992	1.858
		max deviation	0.013	0.071	0.018	0.007	0.108
		average deviation	0.006	0.030	0.009	0.003	0.045
	H ₂ O, MS	mean bond length	0.997	1.836		1.015	1.657
		max deviation	0.004	0.101	–	0.007	0.048
		average deviation	0.002	0.031		0.003	0.023
Pt	OH, vac	mean bond length			2.119	0.976	
		max deviation	–	–	0.121	0.001	–
		average deviation			0.037	0.001	
	OH, MS	mean bond length	1.053	1.506	2.253	0.983	2.016
		max deviation	0.008	0.035	0.042	0.003	0.092
		average deviation	0.005	0.020	0.023	0.002	0.048
	H ₂ O, MS	mean bond length	0.992	1.870		1.008	1.693
		max deviation	0.003	0.050	–	0.004	0.034
		average deviation	0.001	0.017		0.002	0.020

^a: All values are in Å. Values for all metals and bond lengths are included to enable analysis of the effects different metal nanoparticles have on surface-bound water and (solvated) adsorbates. Specifically, all statistics are listed using 3 decimal places to enable a comparison of maximum and average deviations between metals and bonds.

S.8. Excerpts from selected tables

The tables S.ME below contain the data from respective tables S.N which are discussed in the main body.

Table S.2E. Binding and formation energies.

NP	site	Ag			Au			Ir			Pd		
		$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}	$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}	$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}	$\Delta G_{f,vac}$	$\Delta G_{f,MS}$	Ω_{OH}
M ₃₈	111	0.55	0.11	-0.45	x	0.33	x	0.64	-0.18	-0.82	1.15	0.59	-0.56
	kink	0.46	-0.25	-0.71	0.67	0.34	-0.32	-0.21	-0.65	-0.44	0.78	0.10	-0.69
M ₇₉	111E	1.06	0.06	-1.00	1.26	0.63	-0.63	0.24	-0.46	-0.69	0.81	0.11	-0.70
	111T	1.10	0.19	-0.90	1.51	0.71	-0.80	0.84	-0.01	-0.85	1.04	0.32	-0.72
	kink	1.08	-0.02	-1.11	1.17	0.48	-0.69	-0.12	-0.60	-0.47	0.84	0.06	-0.77
M ₂₀₁	100E	0.99	-0.01	-1.00	1.27	0.51	-0.76	x	-0.48	x	0.90	0.27	-0.62
	100T	0.59	0.12	-0.47	x	0.43 ^{ab}	x	0.68	0.12	-0.57	1.10	0.53	-0.57
	111E	0.99	-0.02 ^a	-1.01	1.23	0.39	-0.84	0.14	-0.50	-0.63	0.81	0.11	-0.70
	111TC	1.28	0.35 ^a	-0.93	1.56	0.89	-0.67	0.89	0.11	-0.79	1.06	0.45	-0.62
	111TM	x	0.19 ^b	x	1.53	0.69	-0.84	0.72	0.06	-0.66	1.04	0.39	-0.65
	kink	0.95	-0.14	-1.10	1.10	0.30	-0.80	-0.19	-0.57	-0.38	0.75	0.13	-0.62

a: Vibrational data for 4*H₂O obtained using denser plane wave sampling, as described in S.2.

b: Vibrational data for *OH+3*H₂O obtained using denser plane wave sampling, as described in S.2.

x: No stable configuration available for this adsorption site.

Table S.4E. Parameters for linear regressions of OH formation energies between different metals and adsorption site coordination numbers.

MS	Metal y	Parameters ^a	CN	Metal x				
				Ag	Au	Ir	Pd	Pt
MS	Ag	a	0.12	-	0.81	0.49	0.91	0.63
		b	-0.58	-	-0.39	0.22	-0.14	-0.04
	Au	a	0.13	1.17	-	0.54	1.03	0.73
		b	-0.15	0.49	-	0.74	0.34	0.45
	Ir	a	0.23	1.78	1.38	-	1.91	1.22
		b	-1.60	-0.44	-1.10	-	-0.75	-0.52
	Pd	a	0.11	0.77	0.60	0.44	-	0.61
		b	-0.37	0.17	-0.12	0.37	-	0.12
	Pt	a	0.17	1.35	1.08	0.71	1.54	-
		b	-0.79	0.08	-0.44	0.39	-0.18	-
vac	Ag	a	0.07	-	0.51	0.19	0.73	0.48
		b	0.70	-	0.40	1.02	0.42	0.69
	Au	a	0.14	1.24	-	0.41	1.41	0.80
		b	0.57	-0.02	-	1.19	0.05	0.69
	Ir	a	0.32	3.14	2.37	-	3.26	1.89
		b	-1.47	-3.08	-2.81	-	-2.60	-1.19
	Pd	a	0.09	0.86	0.68	0.28	-	0.53
		b	0.39	-0.03	0.00	0.81	-	0.48
	Pt	a	0.16	1.59	1.12	0.46	1.54	-
		b	-0.08	-0.92	-0.69	0.65	-0.58	-

a: Parameters for linear regressions $\overline{\Delta G}_y = a\Delta G_x + b$, for which $\overline{\Delta G}_y$, ΔG_x , a , and b are in eV.

Table S.5E. Pearson R values for linear relations between formation site binding energies.

	Site	<i>MS, 38</i>		<i>MS, 79</i>			<i>MS, 201</i>					
		111	kink	111E	111T	kink	100E	100T	111E	111TC	111TM	kink
<i>MS, 38</i>	111	x	0.84	0.65	0.61	0.71	0.84	0.79	0.77	0.60	0.68	0.86
	kink	0.84	x	0.94	0.92	0.95	1.00	0.75	0.97	0.94	0.96	0.99
<i>MS, 79</i>	111E	0.65	0.94	x	0.97	0.99	0.95	0.49	0.98	0.95	0.93	0.94
	111T	0.61	0.92	0.97	x	0.96	0.92	0.50	0.94	0.96	0.96	0.91
	kink	0.71	0.95	0.99	0.96	x	0.97	0.52	1.00	0.93	0.92	0.97
<i>MS, 201</i>	100E	0.84	1.00	0.95	0.92	0.97	x	0.71	0.98	0.93	0.94	0.99
	100T	0.79	0.75	0.49	0.50	0.52	0.71	x	0.57	0.64	0.71	0.68
	111E	0.77	0.97	0.98	0.94	1.00	0.98	0.57	x	0.92	0.92	0.99
	111TC	0.60	0.94	0.95	0.96	0.93	0.93	0.64	0.92	x	0.98	0.90
	111TM	0.68	0.96	0.93	0.96	0.92	0.94	0.71	0.92	0.98	x	0.92
	kink	0.86	0.99	0.94	0.91	0.97	0.99	0.68	0.99	0.90	0.92	x
<i>vac, 38</i>	111	x	0.61	0.38	0.61	0.42	0.57	0.60	0.50	0.32	0.56	0.63
	kink	0.93	x	0.81	0.73	0.87	0.93	0.67	0.90	0.73	0.75	0.94
<i>vac, 79</i>	111E	0.56	0.81	x	0.83	0.94	0.85	0.29	0.92	0.81	0.74	0.85
	111T	0.37	0.72	0.90	x	0.87	0.74	0.12	0.83	0.80	0.77	0.75
	kink	0.64	0.79	0.88	0.76	x	0.84	0.28	0.90	0.72	0.67	0.85
<i>vac, 201</i>	100E	0.08	0.52	0.88	0.82	0.89	x	0.39	0.85	0.61	0.55	0.65
	100T	0.80	0.80	0.50	0.72	0.51	0.73	x	0.57	0.73	0.89	0.70
	111E	0.62	0.84	0.94	0.85	0.96	0.88	0.32	x	0.82	0.76	0.89
	111TC	0.34	0.75	0.93	0.88	0.90	0.78	0.19	0.86	x	0.78	0.76
	111TM	0.54	0.91	0.99	0.99	0.97	0.91	0.49	0.95	0.98	x	0.89
	kink	0.67	0.82	0.90	0.78	0.93	0.87	0.35	0.92	0.76	0.71	x
	Site	<i>vac, 38</i>		<i>vac, 79</i>			<i>vac, 201</i>					
		111	kink	111E	111	kink	111E	111	kink	111E	111	kink
<i>MS, 38</i>	111	x	0.93	0.56	0.37	0.64	0.08	0.80	0.62	0.34	0.54	0.67
	kink	0.61	x	0.81	0.72	0.79	0.52	0.80	0.84	0.75	0.91	0.82
<i>MS, 79</i>	111E	0.38	0.81	x	0.90	0.88	0.88	0.50	0.94	0.93	0.99	0.90
	111T	0.61	0.73	0.83	x	0.76	0.82	0.72	0.85	0.88	0.99	0.78
	kink	0.42	0.87	0.94	0.87	x	0.89	0.51	0.96	0.90	0.97	0.93
<i>MS, 201</i>	100E	0.57	0.93	0.85	0.74	0.84	x	0.73	0.88	0.78	0.91	0.87
	100T	0.60	0.67	0.29	0.12	0.28	0.39	x	0.32	0.19	0.49	0.35
	111E	0.50	0.90	0.92	0.83	0.90	0.85	0.57	x	0.86	0.95	0.92
	111TC	0.32	0.73	0.81	0.80	0.72	0.61	0.73	0.82	x	0.98	0.76
	111TM	0.56	0.75	0.74	0.77	0.67	0.55	0.89	0.76	0.78	x	0.71
	kink	0.63	0.94	0.85	0.75	0.85	0.65	0.70	0.89	0.76	0.89	x
<i>vac, 38</i>	111	x	0.60	0.06	0.22	0.19	0.06	0.75	0.18	0.23	0.54	0.21
	kink	0.60	x	0.81	0.56	0.86	0.29	0.62	0.84	0.60	0.72	0.88
<i>vac, 79</i>	111E	0.06	0.81	x	0.83	0.98	0.95	0.11	1.00	0.93	0.98	0.98
	111T	0.22	0.56	0.83	x	0.77	0.99	0.11	0.85	0.92	0.93	0.76
	kink	0.19	0.86	0.98	0.77	x	0.93	0.13	0.99	0.85	0.93	1.00
<i>vac, 201</i>	100E	0.06	0.29	0.95	0.99	0.93	x	0.73	0.99	0.89	0.94	0.96
	100T	0.75	0.62	0.11	0.11	0.13	0.73	x	0.17	0.08	0.99	0.20
	111E	0.18	0.84	1.00	0.85	0.99	0.99	0.17	x	0.92	0.97	0.99
	111TC	0.23	0.60	0.93	0.92	0.85	0.89	0.08	0.92	x	0.99	0.86
	111TM	0.54	0.72	0.98	0.93	0.93	0.94	0.99	0.97	0.99	x	0.93
	kink	0.21	0.88	0.98	0.76	1.00	0.96	0.20	0.99	0.86	0.93	x

Table S.6E. Parameters for linear regressions of solvation energies between different metals and adsorption site coordination numbers.

Metal y	Parameters ^a	\overline{CN}	Ag	Au	Ir	Pd	Pt
Ag	a	0.06	–	-0.02	-0.45	0.23	0.18
	b	-1.30	–	-1.02	-1.29	-0.85	-0.89
Au	a	-0.01	-0.02	–	-0.05	-0.22	-0.08
	b	-0.72	-0.76	–	-0.79	-0.90	-0.81
Ir	a	-0.10	-2.09	-0.19	–	-0.13	-0.04
	b	-0.09	-2.74	-0.78	–	-0.73	-0.66
Pd	a	0.02	0.14	-0.10	-0.02	–	0.46
	b	-0.76	-0.53	-0.75	-0.69	–	-0.38
Pt	a	0.01	0.14	-0.05	0.00	0.57	–
	b	-0.71	-0.49	-0.67	-0.64	-0.25	–

a : Parameters for linear regressions $\overline{\Delta G_y} = a\overline{\Delta G_x} + b$, for which $\overline{\Delta G_y}$, ΔG_x , a , and b are in eV.

Table S.7E. Quality of linear relations between solvation energies on different metals.

	Metal y	Statistics ^a	\overline{CN}	Ag	Au	Ir	Pd	Pt	<i>no trend</i> ^b
Excluding 38 sites, 201–100T	Ag	R	0.91	–	-0.02	-0.97	0.18	0.16	–
		$\max \Delta G_y x$	0.04	–	0.11	0.17	0.12	0.11	0.11
		$\text{ave } \Delta G_y x$	0.03	–	0.06	0.07	0.05	0.06	0.05
	Au	R	-0.09	-0.02	–	-0.10	-0.15	0.16	–
		$\max \Delta G_y x$	0.12	0.11	–	0.12	0.12	0.12	0.13
		$\text{ave } \Delta G_y x$	0.07	0.07	–	0.08	0.07	0.07	0.07
	Ir	R	-0.81	-0.97	-0.10	–	-0.04	-0.01	–
		$\max \Delta G_y x$	0.12	0.06	0.25	–	0.26	0.26	0.26
		$\text{ave } \Delta G_y x$	0.08	0.03	0.12	–	0.12	0.12	0.12
	Pd	R	0.36	0.18	-0.15	-0.04	–	0.00	–
		$\max \Delta G_y x$	0.07	0.08	0.09	0.09	–	0.10	0.10
		$\text{ave } \Delta G_y x$	0.04	0.05	0.05	0.04	–	0.03	0.05
	Pt	R	0.29	0.16	-0.06	-0.01	0.00	–	–
		$\max \Delta G_y x$	0.09	0.10	0.10	0.09	0.09	–	0.10
		$\text{ave } \Delta G_y x$	0.05	0.05	0.04	0.04	0.04	–	0.04

a : All ΔG_y are in eV.

b : Null hypothesis, in which deviations are relative to the average as opposed to linear regression.

Table S.8E. Quality of linear relations between micro-solvated OH_x ($x = 1,2$) energies on different metals.

	Metal y	Statistics ^a	\overline{CN}	Ag	Au	Ir	Pd	<i>no trend</i> ^b
$\text{H}_2\text{O}, MS$	Ag	R	0.22	–	0.78	0.92	0.96	–
		$\max \Delta G_y x$	0.12	–	0.08	0.05	0.04	0.13
	Au	R	0.48	0.78	–	0.86	0.95	–
		$\max \Delta G_y x$	0.14	0.11	–	0.10	0.11	0.17
	Ir	R	0.48	0.92	0.86	–	0.95	–
		$\max \Delta G_y x$	0.33	0.19	0.23	–	0.15	0.53
	Pd	R	0.26	0.96	0.83	0.95	–	–
		$\max \Delta G_y x$	0.19	0.08	0.14	0.08	–	0.20
OH, MS	Ag	R	0.82	–	0.98	0.96	0.90	–
		$\max \Delta G_y x$	0.18	–	0.08	0.10	0.16	0.40
	Au	R	0.89	0.98	–	0.96	0.91	–
		$\max \Delta G_y x$	0.18	0.08	–	0.12	0.19	0.47
	Ir	R	0.85	0.96	0.96	–	0.97	–
		$\max \Delta G_y x$	0.35	0.27	0.22	–	0.19	0.95
	Pd	R	0.75	0.90	0.91	0.97	–	–
		$\max \Delta G_y x$	0.20	0.21	0.19	0.09	–	0.42

a : All ΔG_y are in eV.

b : Null hypothesis, in which deviations are relative to the average as opposed to linear regression.

S.9. Water bilayer on Au(111)

We computed the solvation of *OH within a water bilayer on Au(111) that appears in Table 1 in the main text using a $\sqrt{3}\times\sqrt{3}$ R30° (111) supercell. In it, the adsorption energies of atop *OH in vacuum and surrounded by water are 1.65 and 0.97 eV, such that $\Omega_{OH} = -0.68$ eV. The ZPE and TS (at T = 298.15 K) of *OH are 0.32 and 0.17 eV in vacuum, and 1.02 and 0.25 eV withing the half-dissociated water bilayer. In turn, the ZPE and TS of the full water bilayer are 1.31 and 0.32 eV.

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