Electronic Supplementary Information

Computational design of (100) alloy surfaces for the hydrogen evolution reaction

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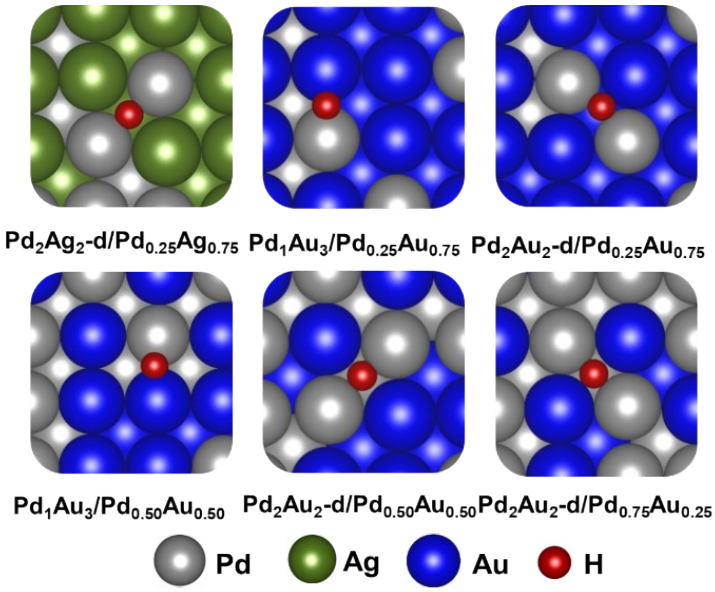

Fig. S1 Typical optimized H binding structures on the six bimetallic ensembles with highest calculated catalytic activities.

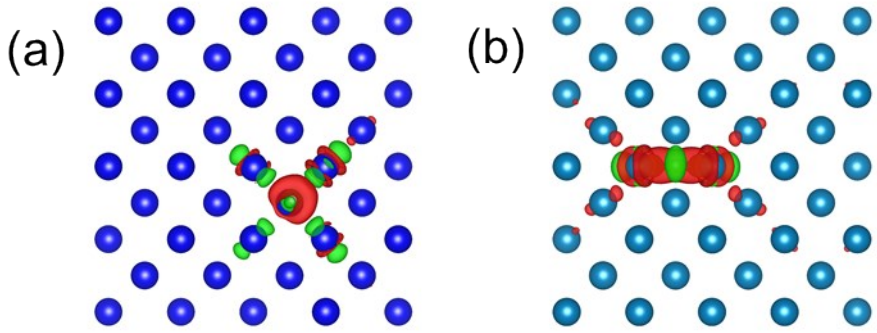

Fig. S2. Energy density differences of H binding on (a) Pd(100) and (b) Pt(100).

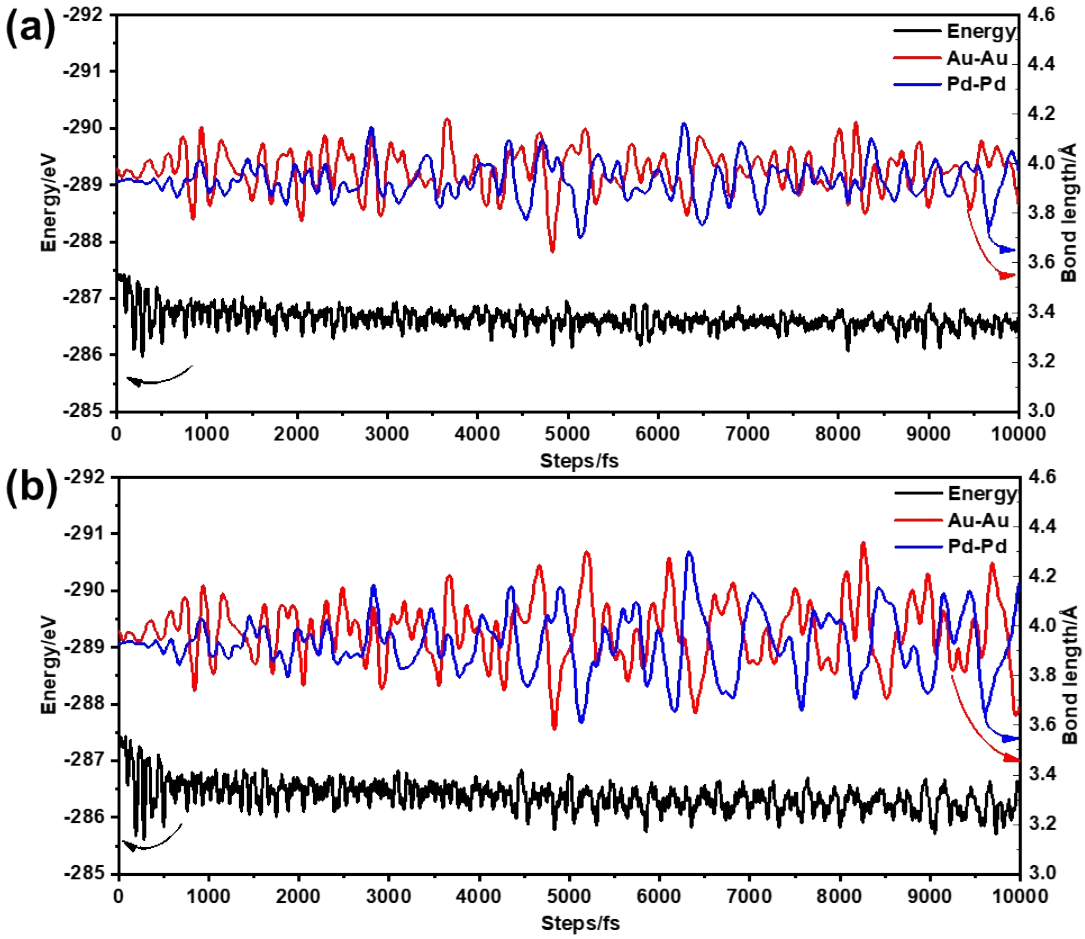

Fig. S3. Energy and bond length variations of Pd_2Au_2-d/$Pd_{0.75}Au_{0.25}$(100) under AIMD simulations, with the temperature of (a) 400 and (b) 500 K.

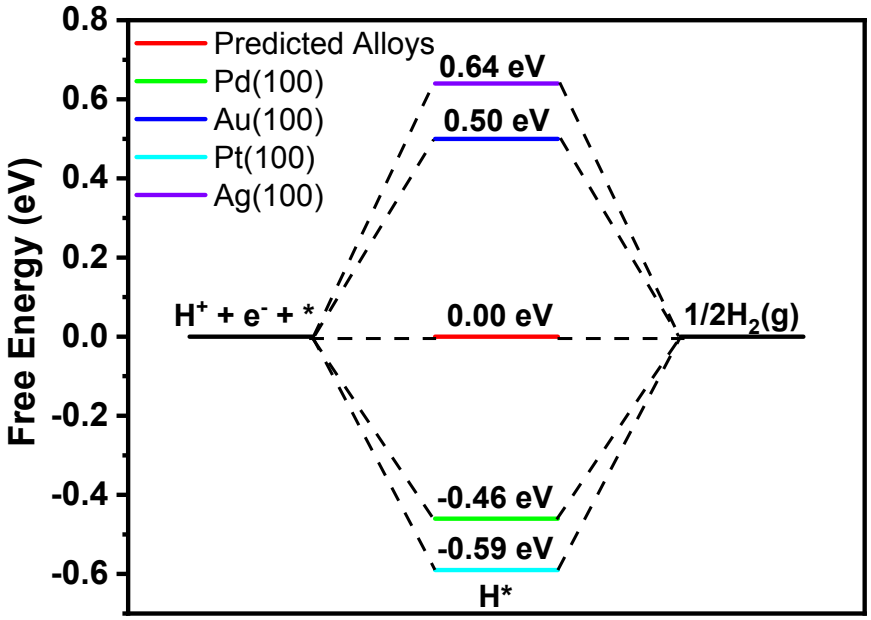

Fig. S4 Free energy diagram of acidic HER at Pd(100), Au(100), Ag(100), Pt(100), and the promising candidates predicted by machine learning (ML) model.

Table S1. Convergence tests on Pd(100) and Au(100) surfaces

H-binding	**Pd(100)**	**Au(100)**
Cutoff = 400 eV	-0.70	0.26
Cutoff = 500 eV	-0.70	0.26
Relative Error	0.00	0.00
EDIFFG = -0.05	-0.70	0.26
EDIFFG = -0.02	-0.70	0.25
Relative Error	0.00	-0.01
ISPIN = 1	-0.70	0.26
ISPIN = 2	-0.73	0.26
Relative Error	-0.03	0.00

Table S2 Input variables for ML modelling

V1	V2	V3	V4	V5	V6	V7	V8
X for atom M	*X* for atom N	*d*-n for atom M	*d*-n for atom N	*R* for atom M	*R* for atom N	*d*-o for atom M	*d*-o for atom N
V9	V10	V11	V12	V13	V14	V15	V16
d-f for atom M	*d*-f for atom N	*d*-b for atom M	*d*-b for atom N	L of M	L of N	A-M	A-N
V17	V18	V19	V20	V21	V22	V23	V24
D-r of M/N	A-r of binding sites	A-r of first layer	A-r of second layer	P-b of M-M	P-b of M-N	P-b of N-N	D-b of M-M
V25	V26						
D-b of M-N	D-b of N-N						

Note: M is the atom with smaller atomic number, N is the atom with larger atomic number, *X* is the element electronegativity, *d*-n is the *d* orbitals electron number, *R* is the atomic radius, *d*-o is the *d* orbitals occupancy, *d*-f is the idealized *d*-band filling, *d*-b is the *d*-band center, L is lattice constant, A-M is the atomic number of M atom, A-N is the atomic number of N atom, D-r is the doping ratio (containing values of 1/3, 1, 3), A-r is the atomic ratio (containing values of 0, 0.25, 0.50, 0.75, 1), P-b is the parallel bonds of binding sites, D-b is the diagnal bonds of binding sites.

Table S3 Detailed values for the calculated H binding energies on $Pd_{0.25}Ag_{0.75}$

$Pd_{0.25}Ag_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Ag_0	1	-204.56364	-208.45250	-0.51
	2	-204.47477	-208.40229	-0.55
	3	-204.66638	-208.55038	-0.51
	4	-204.55889	-208.40319	-0.47
	5	-205.35929	-209.34242	-0.61
Pd_3Ag_1	1	-204.85602	-208.70198	-0.47
	2	-204.93748	-208.80009	-0.49
	3	-204.97321	-208.79446	-0.45
	4	-204.98648	-208.75497	-0.39
	5	-205.83119	-209.69427	-0.49
Pd_2Ag_2-p	1	-204.85602	-208.68681	-0.46
	2	-205.23885	-209.05750	-0.44
	3	-204.93748	-208.79457	-0.48
	4	-206.18568	-210.05814	-0.50
	5	-204.97321	-208.77204	-0.42
Pd_2Ag_2-d	1	-204.85602	-208.49685	-0.27
	2	-204.93748	-208.55625	-0.24
	3	-205.34337	-209.01441	-0.30
	4	-205.23885	-208.84530	-0.23
	5	-205.88705	-209.56166	-0.30
Pd_1Ag_3	1	-204.85602	-208.38663	-0.16
	2	-205.23885	-208.74394	-0.13
	3	-204.93748	-208.55275	-0.24
	4	-205.54325	-209.05644	-0.14
	5	-205.34337	-208.90762	-0.19
Pd_0Ag_4	1	-205.23885	-208.35922	0.25
	2	-205.54325	-208.61779	0.30
	3	-205.34337	-208.37936	0.34
	4	-206.18568	-209.32668	0.23
	5	-206.49865	-209.61800	0.26

Table S4 Detailed values for the calculated H binding energies on $Pd_{0.50}Ag_{0.50}$

$Pd_{0.50}Ag_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Ag_0	1	-240.03706	-243.91593	-0.50
	2	-239.55943	-243.48606	-0.55
	3	-239.92508	-243.75159	-0.45
	4	-240.24814	-244.10775	-0.48
	5	-240.41104	-244.23996	-0.45
Pd_3Ag_1	1	-240.03706	-243.90293	-0.49
	2	-239.55943	-243.49425	-0.56
	3	-239.92508	-243.74801	-0.45
	4	-240.17904	-244.03283	-0.48
	5	-240.24814	-244.00979	-0.39
Pd_2Ag_2-p	1	-240.03706	-243.86971	-0.46
	2	-239.55943	-243.43416	-0.50
	3	-239.92508	-243.77959	-0.48
	4	-240.24814	-244.14051	-0.52
	5	-240.41104	-244.33551	-0.55
Pd_2Ag_2-d	1	-240.03706	-243.71404	-0.30
	2	-239.92508	-243.55254	-0.25
	3	-240.17904	-243.85449	-0.30
	4	-239.63371	-243.31780	-0.31
	5	-241.08369	-244.79436	-0.34
Pd_1Ag_3	1	-239.55943	-243.15070	-0.22
	2	-240.24814	-243.86773	-0.24
	3	-240.41104	-243.98230	-0.20
	4	-239.63371	-243.13351	-0.12
	5	-241.08369	-244.65094	-0.19
Pd_0Ag_4	1	-241.06058	-244.30707	0.13
	2	-239.55943	-242.78541	0.15
	3	-239.63371	-242.84140	0.17
	4	-241.08369	-244.27770	0.18
	5	-241.25069	-244.50441	0.12

Table S5 Detailed values for the calculated H binding energies on $Pd_{0.75}Ag_{0.25}$

$Pd_{0.75}Ag_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Ag_0	1	-273.77146	-277.65315	-0.51
	2	-274.06585	-277.94722	-0.51
	3	-274.41404	-278.31577	-0.53
	4	-274.08246	-277.99256	-0.54
	5	-274.11445	-278.01462	-0.53
Pd_3Ag_1	1	-273.77146	-277.69984	-0.55
	2	-274.06585	-277.91820	-0.48
	3	-274.41404	-278.22096	-0.43
	4	-274.08752	-277.93221	-0.47
	5	-274.08246	-277.96998	-0.51
Pd_2Ag_2-p	1	-274.41404	-278.28655	-0.50
	2	-274.60202	-278.47886	-0.50
	3	-274.09527	-278.00410	-0.53
	4	-273.78758	-277.63984	-0.48
	5	-273.77146	-277.69126	-0.54
Pd_2Ag_2-d	1	-273.78758	-277.42214	-0.26
	2	-273.77146	-277.49126	-0.34
	3	-274.06585	-277.72070	-0.28
	4	-274.41404	-278.04531	-0.26
	5	-274.08752	-277.74408	-0.28
Pd_1Ag_3	1	-274.08752	-277.59338	-0.13
	2	-273.77146	-277.40912	-0.26
	3	-274.41404	-277.60526	0.18
	4	-274.08246	-277.97664	-0.52
	5	-274.11445	-277.65934	-0.17
Pd_0Ag_4	1	-274.60202	-277.70491	0.27
	2	-273.78758	-276.90212	0.26
	3	-274.13432	-277.20270	0.31
	4	-274.72998	-277.84127	0.26
	5	-274.39759	-277.48770	0.28

Table S6 Detailed values for the calculated H binding energies on $Pd_{0.25}Cu_{0.75}$

$Pd_{0.25}Cu_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Cu_0	1	-246.68877	-249.73130	0.33
	2	-245.65149	-248.62841	0.40
	3	-245.71299	-248.69967	0.39
	4	-245.50651	-248.45408	0.43
Pd_3Cu_1	1	-246.95794	-249.77018	0.56
	2	-247.77062	-250.50397	0.64
	3	-246.74612	-249.50289	0.62
	4	-246.92218	-249.70019	0.60
Pd_2Cu_2-p	1	-246.67774	-249.37740	0.68
	2	-246.81669	-249.48848	0.70
	3	-247.15969	-249.86692	0.67
	4	-246.95794	-249.63814	0.69
	5	-247.77062	-250.54173	0.60
Pd_2Cu_2-d	1	-246.95794	-249.69589	0.64
	2	-247.15969	-249.86358	0.67
	3	-246.92218	-249.55364	0.74
	4	-246.99548	-249.76777	0.60
	5	-246.81669	-249.55788	0.63
Pd_1Cu_3	1	-246.67774	-249.22340	0.83
	2	-246.95794	-249.57954	0.75
	3	-246.91366	-249.47201	0.82
	4	-246.81669	-249.47220	0.72
	5	-247.77062	-250.53516	0.61
Pd_0Cu_4	1	-246.67774	-249.09836	0.95
	2	-246.91366	-249.37348	0.92
	3	-246.81669	-249.25239	0.94
	4	-247.77062	-250.25070	0.89
	5	-246.74612	-249.27693	0.84

Table S7 Detailed values for the calculated H binding energies on $Pd_{0.50}Cu_{0.50}$

$Pd_{0.50}Cu_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Cu_0	1	-269.46876	-271.76473	1.08
	2	-269.46876	-271.76012	1.08
	3	-269.29923	-271.65853	1.02
	4	-269.82546	-272.27218	0.93
	5	-270.47120	-272.86336	0.98
Pd_3Cu_1	1	-269.65275	-272.05697	0.97
	2	-269.87904	-272.26347	0.99
	3	-269.29923	-271.58323	1.09
	4	-269.82546	-272.17457	1.03
	5	-270.47120	-272.85602	0.99
Pd_2Cu_2-p	1	-269.46876	-271.75160	1.09
	2	-270.60665	-273.00024	0.98
	3	-269.65275	-271.92389	1.10
	4	-269.87904	-272.13085	1.12
	5	-269.29923	-271.64352	1.03
Pd_2Cu_2-d	1	-270.60665	-272.99723	0.98
	2	-269.82546	-272.26465	0.94
	3	-269.81200	-272.20436	0.98
	4	-270.47120	-272.92084	0.93
	5	-270.17921	-272.57352	0.98
Pd_1Cu_3	1	-269.46876	-271.67012	1.17
	2	-269.65275	-271.80310	1.22
	3	-269.87904	-272.13321	1.12
	4	-269.29923	-271.45071	1.22
	5	-269.82546	-272.00345	1.20
Pd_0Cu_4	1	-269.87904	-271.96029	1.29
	2	-269.29923	-271.28519	1.39
	3	-269.81200	-271.87642	1.31
	4	-269.46876	-271.43245	1.41
	5	-269.46876	-271.43324	1.41

Table S8 Detailed values for the calculated H binding energies on $Pd_{0.75}Cu_{0.25}$

$Pd_{0.75}Cu_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Cu_0	1	-290.74956	-292.81239	1.31
	2	-290.62890	-292.69929	1.30
	3	-290.78998	-292.79855	1.37
	4	-290.78204	-292.80385	1.35
	5	-290.89503	-292.88464	1.39
Pd_3Cu_1	1	-290.74956	-292.81084	1.31
	2	-290.62890	-292.66603	1.34
	3	-290.48166	-292.55072	1.31
	4	-290.78998	-292.83801	1.33
	5	-290.78204	-292.81132	1.35
Pd_2Cu_2-p	1	-290.48166	-292.49196	1.36
	2	-290.89503	-292.94841	1.32
	3	-291.21715	-293.24869	1.34
	4	-290.80049	-292.76968	1.41
	5	-290.54781	-292.49980	1.42
Pd_2Cu_2-d	1	-290.48166	-292.53529	1.32
	2	-290.78998	-292.78560	1.38
	3	-290.64727	-292.71192	1.31
	4	-290.78204	-292.73475	1.42
	5	-291.21715	-293.25826	1.33
Pd_1Cu_3	1	-290.48166	-292.27143	1.59
	2	-290.80049	-292.55634	1.62
	3	-290.54781	-292.32071	1.60
	4	-290.63398	-292.48498	1.52
	5	-290.59602	-292.40997	1.56
Pd_0Cu_4	1	-290.33876	-291.98972	1.72
	2	-290.65227	-292.25071	1.78
	3	-290.38769	-292.02065	1.74
	4	-290.47720	-292.13085	1.72
	5	-290.45543	-292.08894	1.74

Table S9 Detailed values for the calculated H binding energies on $Pd_{0.25}Au_{0.75}$

$Pd_{0.25}Au_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Au_0	1	-227.08532	-230.88360	-0.42
	2	-225.47784	-229.27284	-0.42
	3	-226.35462	-230.14230	-0.41
Pd_3Au_1	1	-225.68223	-229.48418	-0.43
	2	-227.67932	-231.46505	-0.41
	3	-225.97901	-229.79024	-0.44
Pd_2Au_2-p	1	-227.67932	-231.51692	-0.46
	2	-226.29558	-230.12686	-0.46
	3	-225.81945	-229.68723	-0.49
Pd_2Au_2-d	1	-226.29558	-229.90269	-0.23
	2	-226.06213	-229.67078	-0.23
	3	-226.54096	-230.14141	-0.23
Pd_1Au_3	1	-227.47893	-231.04810	-0.19
	2	-227.67932	-231.29363	-0.24
	3	-226.29558	-229.89776	-0.23
Pd_0Au_4	1	-227.47893	-230.82943	0.02
	2	-227.67932	-231.00466	0.05
	3	-226.29558	-229.66439	0.01

Table S10 Detailed values for the calculated H binding energies on $Pd_{0.50}Au_{0.50}$

$Pd_{0.50}Au_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Au_0	1	-255.62621	-259.54935	-0.55
	2	-255.11140	-258.97642	-0.49
	3	-254.12328	-258.09828	-0.60
Pd_3Au_1	1	-255.62621	-259.48662	-0.49
	2	-254.72013	-258.58887	-0.49
	3	-256.38939	-260.29797	-0.53
Pd_2Au_2-p	1	-255.00843	-258.89473	-0.51
	2	-255.62621	-259.51422	-0.51
	3	-254.72013	-258.59440	-0.50
Pd_2Au_2-d	1	-254.72013	-258.29218	-0.20
	2	-256.38939	-259.92906	-0.16
	3	-255.76031	-259.40470	-0.27
Pd_1Au_3	1	-255.00843	-258.62455	-0.24
	2	-254.72013	-258.37427	-0.28
	3	-256.38939	-260.02793	-0.26
Pd_0Au_4	1	-255.00843	-258.41160	-0.03
	2	-256.38939	-259.76150	0.00
	3	-255.11140	-258.52212	-0.04

Table S11 Detailed values for the calculated H binding energies on $Pd_{0.75}Au_{0.25}$

$Pd_{0.75}Au_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pd_4Au_0	1	-281.18091	-285.08723	-0.53
	2	-282.02964	-285.95494	-0.55
	3	-281.62017	-285.50268	-0.51
Pd_3Au_1	1	-281.18091	-285.05759	-0.50
	2	-282.02964	-285.85267	-0.45
	3	-282.47840	-286.37006	-0.52
Pd_2Au_2-p	1	-282.02964	-285.89907	-0.49
	2	-282.47840	-286.33961	-0.49
	3	-281.80622	-285.68930	-0.51
Pd_2Au_2-d	1	-282.47840	-286.09101	-0.24
	2	-281.31027	-284.96333	-0.28
	3	-282.11604	-285.74771	-0.26
Pd_1Au_3	1	-282.09649	-285.78765	-0.32
	2	-282.02964	-285.67773	-0.27
	3	-281.80622	-285.47784	-0.30
Pd_0Au_4	1	-282.38971	-285.72508	0.04
	2	-282.06778	-285.51024	-0.07
	3	-282.42535	-285.78923	0.01

Table S12 Detailed values for the calculated H binding energies on $Pd_{0.25}Ag_{0.75}$

$Pt_{0.25}Ag_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Ag_0	1	-214.66865	-218.84247	-0.80
	2	-215.05208	-219.09753	-0.67
	3	-215.59530	-219.53367	-0.56
	4	-213.83074	-217.89368	-0.69
	5	-214.06270	-218.14758	-0.71
Pt_3Ag_1	1	-215.62948	-219.55279	-0.55
	2	-216.15072	-220.13905	-0.61
	3	-214.34815	-218.42675	-0.70
	4	-214.74424	-218.72262	-0.60
	5	-214.66865	-218.81643	-0.77
Pt_2Ag_2-p	1	-215.62948	-219.64367	-0.64
	2	-216.15072	-220.12021	-0.59
	3	-214.66865	-218.66759	-0.62
	4	-214.74424	-218.89023	-0.77
	5	-214.34815	-218.42483	-0.70
Pt_2Ag_2-d	1	-214.34815	-218.10734	-0.38
	2	-213.80664	-217.55856	-0.38
	3	-214.74424	-218.43844	-0.32
	4	-215.42649	-219.03116	-0.23
	5	-216.15072	-219.96775	-0.44
Pt_1Ag_3	1	-216.84747	-220.66416	-0.44
	2	-215.62948	-219.56027	-0.56
	3	-216.15072	-219.96368	-0.44
	4	-214.34815	-218.26831	-0.55
	5	-214.38762	-218.22309	-0.46
Pt_0Ag_4	1	-216.84747	-220.03598	0.19
	2	-214.38762	-217.45572	0.31
	3	-214.66865	-217.88739	0.16
	4	-213.80664	-216.84327	0.34
	5	-215.03286	-217.85511	0.55

Table S13 Detailed values for the calculated H binding energies on $Pd_{0.50}Ag_{0.50}$

$Pt_{0.50}Ag_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Ag_0	1	-259.66476	-263.70267	-0.66
	2	-259.60882	-263.69706	-0.71
	3	-260.76948	-264.81018	-0.67
	4	-260.49235	-264.52396	-0.66
	5	-260.13391	-264.06720	-0.56
Pt_3Ag_1	1	-260.76948	-264.83796	-0.69
	2	-259.60882	-263.67546	-0.69
	3	-260.49235	-264.60235	-0.74
	4	-260.59947	-264.69490	-0.72
	5	-261.55599	-265.59251	-0.66
Pt_2Ag_2-p	1	-260.76948	-264.73784	-0.59
	2	-259.66476	-263.70139	-0.66
	3	-261.22477	-265.39145	-0.79
	4	-259.60882	-263.71931	-0.74
	5	-260.49235	-264.54124	-0.67
Pt_2Ag_2-d	1	-261.55599	-265.20227	-0.27
	2	-260.32112	-264.03216	-0.34
	3	-259.66476	-263.35705	-0.32
Pt_1Ag_3	1	-260.32112	-264.17461	-0.48
	2	-259.66476	-263.61071	-0.57
	3	-261.22477	-265.20527	-0.61
	4	-259.60882	-263.47716	-0.49
	5	-262.03117	-265.90566	-0.50
Pt_0Ag_4	1	-261.22477	-264.33304	0.27
	2	-262.03117	-265.26567	0.14
	3	-261.55599	-264.76590	0.17
	4	-260.87021	-263.95401	0.29
	5	-259.91716	-262.96531	0.33

Table S14 Detailed values for the calculated H binding energies on $Pd_{0.75}Ag_{0.25}$

$Pt_{0.75}Ag_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Ag_0	1	-310.34329	-314.37603	-0.66
	2	-309.11965	-313.04170	-0.55
	3	-308.28311	-312.37947	-0.72
	4	-309.82332	-313.77972	-0.58
	5	-309.45316	-313.51852	-0.69
Pt_3Ag_1	1	-310.34329	-314.35882	-0.64
	2	-309.11965	-313.12985	-0.64
	3	-308.28311	-312.38490	-0.73
	4	-309.82332	-313.85599	-0.66
	5	-309.45316	-313.37538	-0.55
Pt_2Ag_2-p	1	-309.45316	-313.53321	-0.71
	2	-310.28412	-314.32223	-0.66
	3	-309.23902	-313.24300	-0.63
	4	-308.28157	-312.34553	-0.69
	5	-309.17694	-313.20371	-0.65
Pt_2Ag_2-d	1	-310.28412	-314.14691	-0.49
	2	-309.17694	-313.04757	-0.50
	3	-309.48209	-313.49280	-0.64
	4	-309.11965	-313.07968	-0.59
	5	-309.82332	-313.65238	-0.45
Pt_1Ag_3	1	-310.28412	-314.17815	-0.52
	2	-309.23902	-313.03891	-0.42
	3	-309.48209	-313.38392	-0.53
	4	-310.44635	-314.29766	-0.48
	5	-309.54168	-313.45500	-0.54
Pt_0Ag_4	1	-310.70109	-313.73151	0.34
	2	-309.53922	-312.59931	0.31
	3	-310.18653	-313.24630	0.32
	4	-310.09844	-313.32220	0.15

Table S15 Detailed values for the calculated H binding energies on $Pt_{0.25}Cu_{0.75}$

$Pt_{0.25}Cu_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Cu_0	1	-259.77290	-263.64746	-0.50
	2	-259.97292	-264.20722	-0.86
	3	-259.39171	-263.51671	-0.75
Pt_3Cu_1	1	-260.01496	-263.83384	-0.44
	2	-259.77290	-263.64963	-0.50
	3	-260.35586	-264.07399	-0.34
Pt_2Cu_2-p	1	-259.77290	-263.69471	-0.55
	2	-248.95959	-252.95218	-0.62
	3	-260.01496	-263.93910	-0.55
Pt_2Cu_2-d	1	-248.95959	-252.82497	-0.49
	2	-260.01496	-263.91642	-0.53
	3	-259.31440	-263.27729	-0.59
Pt_1Cu_3	1	-259.77290	-263.63233	-0.48
	2	-248.95959	-252.76603	-0.43
	3	-249.72188	-253.65988	-0.56
Pt_0Cu_4	1	-259.77290	-263.28985	-0.14
	2	-248.95959	-252.42788	-0.09
	3	-249.72188	-253.19467	-0.10

Table S16 Detailed values for the calculated H binding energies on $Pt_{0.50}Cu_{0.50}$

$Pt_{0.50}Cu_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Cu_0	1	-295.01899	-299.27284	-0.88
	2	-294.59851	-298.60097	-0.63
	3	-295.31080	-299.41580	-0.73
Pt_3Cu_1	1	-293.83266	-297.80881	-0.60
	2	-295.32741	-299.18675	-0.48
	3	-294.59851	-298.49456	-0.52
Pt_2Cu_2-p	1	-293.83266	-297.73802	-0.53
	2	-295.01899	-299.10641	-0.71
	3	-295.07628	-299.14660	-0.70
Pt_2Cu_2-d	1	-293.83266	-297.89390	-0.69
	2	-295.35263	-299.33763	-0.61
	3	-295.32741	-299.22141	-0.52
Pt_1Cu_3	1	-293.83266	-297.65996	-0.45
	2	-295.56459	-299.47182	-0.53
	3	-295.01899	-298.88088	-0.49
Pt_0Cu_4	1	-295.07628	-298.55664	-0.11
	2	-294.59851	-298.11447	-0.14
	3	-295.45085	-299.02321	-0.20

Table S17 Detailed values for the calculated H binding energies on $Pt_{0.75}Cu_{0.25}$

$Pt_{0.75}Cu_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Cu_0	1	-328.52894	-332.55358	-0.65
	2	-328.35945	-332.38580	-0.65
	3	-328.82032	-332.77662	-0.58
Pt_3Cu_1	1	-328.52894	-332.55754	-0.65
	2	-328.35945	-332.46836	-0.73
	3	-328.82032	-332.74337	-0.55
Pt_2Cu_2-p	1	-328.35945	-332.38893	-0.65
	2	-328.82032	-332.80232	-0.61
	3	-328.54573	-332.52150	-0.60
Pt_2Cu_2-d	1	-328.82032	-332.62846	-0.43
	2	-328.54573	-332.40337	-0.48
	3	-328.95336	-332.85544	-0.53
Pt_1Cu_3	1	-328.54573	-332.48954	-0.57
	2	-328.33659	-332.25965	-0.55
	3	-327.90488	-331.87144	-0.59
Pt_0Cu_4	1	-328.61068	-332.03849	-0.05
	2	-328.30393	-331.70781	-0.03
	3	-327.88372	-331.33941	-0.08

Table S18 Detailed values for the calculated H binding energies on $Pt_{0.25}Au_{0.75}$

$Pt_{0.25}Au_{0.75}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Au_0	1	-231.91688	-235.94514	-0.65
	2	-232.30081	-236.33949	-0.66
	3	-231.21261	-235.24823	-0.66
	4	-232.47265	-236.42245	-0.57
	5	-233.25980	-237.22556	-0.59
Pt_3Au_1	1	-232.68635	-236.71134	-0.65
	2	-231.51299	-235.48255	-0.59
	3	-232.91860	-236.84622	-0.55
	4	-232.29432	-236.34730	-0.68
	5	-233.46071	-237.43633	-0.60
Pt_2Au_2-p	1	-232.29432	-236.33064	-0.66
	2	-232.68635	-236.69181	-0.63
	3	-231.51299	-235.49312	-0.61
	4	-232.30712	-236.37201	-0.69
	5	-233.85222	-237.93155	-0.70
Pt_2Au_2-d	1	-233.40670	-236.99714	-0.22
	2	-232.91860	-236.73522	-0.44
	3	-232.29432	-236.09837	-0.43
	4	-233.85222	-237.73512	-0.51
	5	-232.55100	-236.36316	-0.44
Pt_1Au_3	1	-234.53723	-238.26789	-0.36
	2	-232.29432	-236.14327	-0.47
	3	-232.68635	-236.56261	-0.50
	4	-234.12333	-237.91628	-0.42
	5	-233.46071	-237.33941	-0.50
Pt_0Au_4	1	-234.53723	-237.75867	0.15
	2	-232.29432	-235.77172	-0.10
	3	-234.12333	-237.40304	0.10
	4	-233.46071	-236.73819	0.10
	5	-233.40670	-236.67927	0.10

Table S19 Detailed values for the calculated H binding energies on $Pt_{0.50}Au_{0.50}$

$Pt_{0.50}Au_{0.50}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Au_0	1	-274.42353	-278.36121	-0.56
	2	-274.00034	-277.97786	-0.60
	3	-272.47139	-276.50284	-0.66
	4	-271.89338	-275.94864	-0.68
	5	-271.80062	-275.82042	-0.64
Pt_3Au_1	1	-271.80062	-275.88620	-0.71
	2	-274.24540	-278.25441	-0.63
	3	-274.42353	-278.39694	-0.60
	4	-274.51752	-278.57962	-0.69
	5	-274.63279	-278.60310	-0.60
Pt_2Au_2-p	1	-271.80062	-275.69509	-0.52
	2	-274.24540	-278.28620	-0.67
	3	-274.42353	-278.37982	-0.58
	4	-274.51752	-278.56165	-0.67
	5	-274.63279	-278.68270	-0.67
Pt_2Au_2-d	1	-271.80062	-275.46546	-0.29
	2	-274.42353	-278.24284	-0.44
	3	-274.51752	-278.28692	-0.39
	4	-273.11853	-276.95199	-0.46
	5	-274.24540	-278.05970	-0.44
Pt_1Au_3	1	-271.80062	-275.58399	-0.41
	2	-274.42353	-278.24033	-0.44
	3	-274.51752	-278.38110	-0.49
	4	-274.63279	-278.52326	-0.52
	5	-273.63660	-277.47584	-0.46
Pt_0Au_4	1	-271.80062	-275.16469	0.01
	2	-274.51752	-277.94005	-0.05
	3	-274.72205	-278.06881	0.03
	4	-274.93360	-278.35087	-0.04
	5	-274.11196	-277.45068	0.04

Table S20 Detailed values for the calculated H binding energies on $Pt_{0.75}Au_{0.25}$

$Pt_{0.75}Au_{0.25}$	Number	Catalysts (eV)	Adsorption system (eV)	E_{ads} (eV)
Pt_4Au_0	1	-315.71591	-319.69406	-0.60
	2	-313.61799	-317.64697	-0.65
	3	-313.26651	-317.06655	-0.43
	4	-314.16335	-318.24886	-0.71
	5	-315.33928	-319.34703	-0.63
Pt_3Au_1	1	-315.67247	-319.68309	-0.64
	2	-315.71591	-319.72133	-0.63
	3	-314.77016	-318.82819	-0.68
	4	-313.61799	-317.72526	-0.73
	5	-314.90394	-318.94456	-0.67
Pt_2Au_2-p	1	-315.67247	-319.74525	-0.70
	2	-315.71591	-319.69334	-0.60
	3	-314.77016	-318.76195	-0.62
	4	-314.03109	-318.15152	-0.75
	5	-314.16335	-318.01244	-0.47
Pt_2Au_2-d	1	-315.67247	-319.58698	-0.54
	2	-314.77016	-318.61547	-0.47
	3	-315.33928	-319.17577	-0.46
	4	-314.16335	-318.07085	-0.53
	5	-315.71591	-319.57928	-0.49
Pt_1Au_3	1	-314.03109	-317.86359	-0.46
	2	-315.67247	-319.58703	-0.54
	3	-315.71591	-319.50808	-0.42
	4	-314.77016	-318.58519	-0.44
	5	-314.74370	-318.65313	-0.53
Pt_0Au_4	1	-316.26124	-319.69486	-0.06
	2	-316.10337	-319.49781	-0.02
	3	-315.43756	-318.85815	-0.05
	4	-314.73747	-318.12533	-0.01
	5	-315.36108	-318.80119	-0.07