Supplementary Information

Size-dependent catalytic activity for CO oxidation over sub-nano Au clusters

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Supplementary Tables

Table S1 Gupta potential parameters for Au clusters[1]

Parameter	*A/eV*	*ζ/eV*	*p*	*q*	r_0/ *Å*
Au-Au	0.2061	1.7900	10.229	4.0360	2.884

Table S2 Energies and structures information of Au_N(2-20) at pure DFT level. ***Size***: also denoted as N, ranging from 2 to 20.$\boldsymbol{E_{coh}}$: cohesive energy of cluster. $\boldsymbol{\Delta E}$: the cohesive energy difference between the GM identified in this work and those reported previously[2]. $\boldsymbol{D_2E}$: second finite energy difference of cluster, defined as $D_2E=E_{N+1}+E_{N-1}-2E_N$. $\boldsymbol{E_{fit}}$: by fitting $\boldsymbol{E_{coh}}$ to N arrived at $0.716-1.822N^{-1}-1.237N^{-2/3}+2.656N^{-1/3}$. $\boldsymbol{\Delta E_{coh}}$: relative choesive energies of clusters calculated by $\boldsymbol{E_{fit}-E_{coh}}$. ***Dia***: effective diameter of cluster, which is defined as the largest distance among atoms. ***BL***: average bond length of cluster, with cutoff length be 3.3 Å.

Size	$\boldsymbol{E_{coh}}$ ***(eV)***	$\boldsymbol{\Delta E}$ ***(eV)***	$\boldsymbol{D_2E}$ ***(eV)***	$\boldsymbol{\Delta E_{coh}}$ ***(eV)***	$\boldsymbol{E_{fit}}$ ***(eV)***	***Dia (nm)***	***BL (Å)***
2	0.948	-0.003	-	-0.064	0.884	0.2512	2.6992
3	0.965	0.023	-1.375	0.196	1.16	0.466	2.8311
4	1.317	-0.02	0.413	0.015	1.332	0.4683	2.8306
5	1.445	-0.033	-0.997	0.007	1.452	0.4614	2.8295
6	1.697	-0.016	1.624	-0.155	1.542	0.5275	2.8375
7	1.646	-0.016	-1.329	-0.032	1.613	0.7054	2.8215
8	1.773	-0.013	1.249	-0.102	1.671	0.7037	2.8316
9	1.733	-0.024	-1.137	-0.014	1.719	0.8093	2.8309
10	1.815	0.003	0.629	-0.054	1.76	0.9537	2.8313
11	1.825	-0.01	-0.368	-0.029	1.796	0.9311	2.8267
12	1.863	-0.002	0.44	-0.036	1.827	0.7984	2.8227
13	1.862	-0.007	-0.844	-0.007	1.855	1.2284	2.7129
14	1.922	-0.011	0.941	-0.042	1.88	0.787	2.835
15	1.91	-0.011	-0.53	-0.008	1.902	0.7254	2.8307
16	1.934	-0.03	-0.19	-0.011	1.922	0.7595	2.8346
17	1.965	-0.008	-0.17	-0.024	1.941	0.9391	2.8347
18	2.003	-0.026	1.351	-0.045	1.958	0.7161	2.833
19	1.965	0.045	-1.337	0.008	1.974	0.8368	2.8301
20	1.999	-0.098	1.828	-0.01	1.988	0.9153	2.8238

Table S3 Energies and structures information of Au_N (20-300) at Gupta combined DFT level. ***Size***: also denoted as N, ranging from 21 to 300. E_{coh}: cohesive energy of cluster. D_2E: second finite energy difference of cluster, defined as $D_2E=E_{N+1}+E_{N-1}-2E_N$. E_{fit} : by fitting E_{coh} to N arrived at $0.716-1.822N^{-1}-1.237N^{-2/3}+2.656N^{-1/3}$. ΔE_{coh}: relative cohesive energies of clusters calculated by $E_{fit} - E_{coh}$. ***Dia***: diameters of cluster, which is defined as the largest distance among atoms. ***BL***: average bond length of cluster, with cutoff length be 3.3 Å.

Size	E_{coh} *(eV)*	D_2E *(eV)*	ΔE_{coh}*(eV)*	E_{fit} *(eV)*	*Dia(nm)*	*BL(Å)*
21	1.941	-0.803	0.06	2.002	0.7087	2.824
22	1.926	-0.952	0.088	2.014	0.7421	2.8311
23	1.953	-0.142	0.073	2.026	0.8095	2.8361
24	1.984	0.377	0.053	2.037	0.8373	2.7797
25	1.998	0.307	0.05	2.048	0.8482	2.8313
26	1.998	0.514	0.059	2.058	0.8033	2.8359
27	1.98	-1.44	0.087	2.067	0.8152	2.8238
28	2.014	0.576	0.062	2.076	0.8613	2.8326
29	2.026	-0.863	0.058	2.084	0.8457	2.8351
30	2.066	1.204	0.026	2.092	0.8334	2.8256
31	2.065	-0.57	0.036	2.1	0.8168	2.8327
32	2.081	0.509	0.026	2.107	0.8282	2.8348
33	2.081	-1.515	0.033	2.114	0.8073	2.8361
34	2.126	-0.701	-0.005	2.121	0.8858	2.8321
35	2.188	7.436	-0.06	2.127	0.9484	2.7246
36	2.04	-8.277	0.094	2.134	1.0138	2.8358
37	2.124	3.172	0.016	2.139	0.9182	2.8251
38	2.12	0.204	0.026	2.145	0.8921	2.8259
39	2.11	-0.38	0.04	2.151	0.92	2.8358
40	2.111	-1.265	0.045	2.156	0.9051	2.8371
41	2.143	1.56	0.018	2.161	0.9505	2.8366
42	2.136	-0.545	0.03	2.166	1.003	2.83
43	2.142	-0.062	0.029	2.171	0.8655	2.8249
44	2.149	0.14	0.026	2.175	1.0412	2.8371
45	2.153	-0.627	0.027	2.18	1.0111	2.8296
46	2.17	0.798	0.014	2.184	1.1063	2.7715
47	2.169	-0.479	0.019	2.188	0.9647	2.8343
48	2.179	0.989	0.013	2.192	0.9703	2.8369
49	2.168	-1.993	0.028	2.196	1.075	2.8395
50	2.197	1.283	0.003	2.2	1.082	2.8343
51	2.2	0.576	0.004	2.203	0.9649	2.8427
52	2.191	-4.714	0.016	2.207	1.0738	2.8379
53	2.272	10.273	-0.062	2.21	1.0499	2.8431
54	2.16	-11.798	0.054	2.214	1.0836	2.8283
55	2.266	11.35	-0.049	2.217	1.2459	2.8261
56	2.166	-9.219	0.054	2.22	1.0171	2.8277
57	2.231	3.382	-0.008	2.224	1.1913	2.8039
58	2.236	-0.17	-0.009	2.227	1.0836	2.8427
59	2.243	0.598	-0.014	2.23	1.0888	2.839

60	2.24	-1.357	-0.008	2.233	1.162	2.8458
61	2.26	1.598	-0.024	2.235	1.0851	2.8366
62	2.253	-0.485	-0.014	2.238	1.1009	2.8297
63	2.254	-0.128	-0.013	2.241	1.1896	2.841
64	2.256	0.106	-0.013	2.243	1.0868	2.841
65	2.258	-0.129	-0.011	2.246	1.1473	2.8395
66	2.261	0.759	-0.012	2.249	1.1472	2.8293
67	2.252	-1.377	-0.001	2.251	1.1539	2.8389
68	2.264	0.39	-0.011	2.254	1.1785	2.8082
69	2.271	0.163	-0.015	2.256	1.1021	2.8393
70	2.274	-0.231	-0.016	2.258	1.1614	2.8301
71	2.281	1.416	-0.021	2.261	1.3618	2.8401
72	2.268	-7.201	-0.005	2.263	1.1239	2.8415
73	2.354	11.575	-0.089	2.265	1.1754	2.8415
74	2.281	0.153	-0.014	2.267	1.2787	2.8452
75	2.208	-11.159	0.061	2.269	1.304	2.8406
76	2.284	2.092	-0.013	2.271	1.2628	2.8444
77	2.331	9.91	-0.058	2.273	1.3334	2.838
78	2.249	-9.129	0.026	2.275	1.2144	2.841
79	2.285	2.019	-0.008	2.277	1.2472	2.7994
80	2.295	0.643	-0.016	2.279	1.3324	2.8321
81	2.297	0.199	-0.016	2.281	1.2927	2.8386
82	2.296	-0.359	-0.014	2.283	1.2036	2.8325
83	2.3	-0.079	-0.015	2.285	1.3617	2.8315
84	2.305	0.658	-0.018	2.287	1.345	2.8395
85	2.301	-0.449	-0.013	2.288	1.2435	2.8431
86	2.303	0.325	-0.013	2.29	1.3661	2.8415
87	2.301	-0.658	-0.01	2.292	1.2285	2.8408
88	2.307	0.471	-0.014	2.293	1.3226	2.8332
89	2.307	0.074	-0.012	2.295	1.3576	2.8421
90	2.307	-0.143	-0.01	2.297	1.3397	2.8088
91	2.308	-0.183	-0.01	2.298	1.3536	2.8312
92	2.311	0.036	-0.011	2.3	1.3405	2.8319
93	2.313	-0.11	-0.012	2.301	1.3942	2.8409
94	2.317	0.941	-0.014	2.303	1.2568	2.8334
95	2.311	-1.615	-0.006	2.304	1.3468	2.8385
96	2.321	1.595	-0.015	2.306	1.3981	2.8325
97	2.315	-0.698	-0.008	2.307	1.3239	2.8427
98	2.316	-1.722	-0.008	2.309	1.4898	2.8344
99	2.335	1.773	-0.025	2.31	1.3848	2.8354
100	2.335	1.428	-0.024	2.312	1.4022	2.8432
101	2.322	-1.869	-0.009	2.313	1.3816	2.8369
102	2.327	0.453	-0.012	2.314	1.2541	2.8444
103	2.327	-0.143	-0.012	2.316	1.358	2.8322
104	2.329	0.117	-0.012	2.317	1.5096	2.8353
105	2.33	-0.096	-0.011	2.318	1.3825	2.8416
106	2.331	0.156	-0.012	2.319	1.4162	2.8409
107	2.331	-0.031	-0.01	2.321	1.3739	2.8342
108	2.331	-0.817	-0.009	2.322	1.4483	2.8424

109	2.339	1.767	-0.016	2.323	1.4108	2.8346
110	2.331	-1.045	-0.006	2.324	1.4611	2.8318
111	2.332	-0.251	-0.006	2.326	1.3825	2.8435
112	2.335	0.836	-0.009	2.327	1.5499	2.5122
113	2.331	-1.626	-0.003	2.328	1.5259	2.7989
114	2.342	1.65	-0.013	2.329	1.5594	2.8456
115	2.337	-1.395	-0.007	2.33	1.4311	2.8406
116	2.345	1.066	-0.014	2.331	1.3912	2.8333
117	2.344	0.101	-0.011	2.332	1.5421	2.8387
118	2.342	-2.613	-0.008	2.334	1.3793	2.8439
119	2.361	2.175	-0.027	2.335	1.4121	2.8382
120	2.363	2.056	-0.027	2.336	1.6196	2.8334
121	2.347	-1.993	-0.01	2.337	1.5302	2.8392
122	2.348	-0.082	-0.01	2.338	1.5884	2.832
123	2.349	-0.211	-0.011	2.339	1.5198	2.8438
124	2.353	0.241	-0.013	2.34	1.5188	2.7993
125	2.354	0.33	-0.013	2.341	1.4829	2.8409
126	2.353	0.339	-0.011	2.342	1.5295	2.8457
127	2.348	-1.271	-0.006	2.343	1.5478	2.8363
128	2.354	0.555	-0.011	2.344	1.5188	2.8442
129	2.356	1.027	-0.011	2.345	1.5359	2.8424
130	2.35	-1.413	-0.004	2.346	1.5034	2.8414
131	2.354	0.448	-0.007	2.347	1.6068	2.8352
132	2.355	0.055	-0.007	2.348	1.5909	2.8412
133	2.356	-0.155	-0.007	2.349	1.6247	2.8416
134	2.357	0.276	-0.008	2.349	1.4715	2.846
135	2.357	-0.421	-0.007	2.35	1.5461	2.7729
136	2.36	0.98	-0.009	2.351	1.6907	2.8372
137	2.356	-1.764	-0.003	2.352	1.6897	2.8464
138	2.364	0.546	-0.011	2.353	1.4977	2.8468
139	2.368	-2.988	-0.015	2.354	1.6072	2.8376
140	2.394	7.957	-0.039	2.355	1.5633	2.8374
141	2.363	-4.071	-0.007	2.356	1.542	2.8367
142	2.361	-1.289	-0.005	2.356	1.5473	2.8358
143	2.368	0.711	-0.011	2.357	1.5359	2.8362
144	2.37	0.104	-0.012	2.358	1.514	2.8374
145	2.371	-0.052	-0.012	2.359	1.6257	2.847
146	2.373	0.537	-0.013	2.36	1.6328	2.7813
147	2.371	0.06	-0.01	2.361	1.7809	2.8428
148	2.368	-0.245	-0.007	2.361	1.602	2.8359
149	2.368	-1.078	-0.005	2.362	1.764	2.8475
150	2.374	1.436	-0.011	2.363	1.6344	2.8378
151	2.371	-0.617	-0.007	2.364	1.6947	2.8439
152	2.372	0.293	-0.007	2.364	1.6407	2.842
153	2.371	-0.016	-0.006	2.365	1.7178	2.8475
154	2.37	0.109	-0.004	2.366	1.6924	2.8373
155	2.368	0.612	-0.002	2.367	1.6844	2.8388
156	2.363	-2.423	0.005	2.367	1.6168	2.8385
157	2.373	1.053	-0.005	2.368	1.7493	2.7739

158	2.376	0.475	-0.007	2.369	1.6085	2.8463
159	2.376	-0.367	-0.007	2.37	1.6792	2.8446
160	2.379	0.664	-0.008	2.37	1.6691	2.8382
161	2.377	0.621	-0.006	2.371	1.7535	2.8385
162	2.372	-2.341	0	2.372	1.7046	2.8395
163	2.38	2.015	-0.008	2.372	1.6676	2.838
164	2.377	0.117	-0.004	2.373	1.6607	2.8379
165	2.373	-1.814	0.001	2.374	1.6562	2.838
166	2.38	1.441	-0.005	2.374	1.6835	2.8455
167	2.378	0.111	-0.003	2.375	1.7216	2.8404
168	2.375	-0.829	0	2.376	1.7064	2.7816
169	2.378	-0.798	-0.002	2.376	1.6944	2.8388
170	2.385	1.671	-0.008	2.377	1.7616	2.8386
171	2.382	-0.982	-0.005	2.378	1.6965	2.8382
172	2.385	1.975	-0.007	2.378	1.5772	2.8383
173	2.377	-2.402	0.002	2.379	1.7707	2.8447
174	2.382	1.069	-0.003	2.38	1.6951	2.8449
175	2.382	0.192	-0.001	2.38	1.7129	2.849
176	2.38	-1.488	0.001	2.381	1.7216	2.8457
177	2.386	1.738	-0.005	2.381	1.6775	2.8369
178	2.383	-0.513	-0.001	2.382	1.8569	2.8474
179	2.383	-0.6	0	2.383	1.7292	2.7922
180	2.386	0.569	-0.003	2.383	1.7413	2.8454
181	2.386	0.485	-0.002	2.384	1.9682	2.8376
182	2.383	-2.106	0.001	2.384	1.7549	2.8391
183	2.392	2.68	-0.007	2.385	1.9685	2.8475
184	2.386	-1.198	0	2.386	1.6703	2.8413
185	2.386	0.691	0	2.386	1.9492	2.8486
186	2.383	-1.952	0.003	2.387	1.7771	2.8466
187	2.391	0.294	-0.003	2.387	1.8672	2.851
188	2.396	2.308	-0.008	2.388	1.8042	2.8488
189	2.39	-0.445	-0.001	2.388	1.7912	2.8378
190	2.386	-1.897	0.003	2.389	1.6996	2.8046
191	2.391	-0.352	-0.002	2.389	1.7876	2.846
192	2.399	3.787	-0.009	2.39	1.5802	2.8467
193	2.387	-2.582	0.004	2.391	1.831	2.8499
194	2.388	-0.245	0.003	2.391	1.7367	2.8474
195	2.391	0.405	0.001	2.392	1.9036	2.8396
196	2.391	-1.034	0.001	2.392	1.988	2.8478
197	2.397	2.786	-0.004	2.393	1.7508	2.8388
198	2.389	-2.419	0.005	2.393	1.816	2.8389
199	2.392	1.101	0.001	2.394	1.8945	2.8451
200	2.391	-0.781	0.003	2.394	1.9028	2.8489
201	2.393	-0.149	0.002	2.395	1.8422	2.7788
202	2.396	0.887	-0.001	2.395	1.8965	2.8396
203	2.394	-0.498	0.001	2.396	1.8992	2.8409
204	2.395	0.395	0.001	2.396	1.846	2.8483
205	2.394	-0.48	0.002	2.397	1.7618	2.8413
206	2.396	0.427	0.001	2.397	1.7525	2.841

207	2.395	1.063	0.003	2.398	1.8202	2.8461
208	2.389	-3.053	0.009	2.398	1.8824	2.8394
209	2.398	1.545	0.001	2.399	1.8401	2.8399
210	2.4	1.849	0	2.399	1.9287	2.8397
211	2.392	-2.935	0.007	2.4	1.806	2.8399
212	2.399	1.172	0.001	2.4	1.9796	2.7873
213	2.4	0.282	0.001	2.401	1.7295	2.8503
214	2.399	0.615	0.002	2.401	1.809	2.8408
215	2.396	-2.498	0.005	2.402	1.88	2.8472
216	2.405	3.818	-0.003	2.402	1.7461	2.842
217	2.395	-2.307	0.007	2.402	1.9776	2.8443
218	2.397	0.53	0.006	2.403	1.8144	2.8472
219	2.396	-2.152	0.008	2.403	1.9606	2.841
220	2.405	2.685	-0.001	2.404	1.8838	2.8521
221	2.401	0.523	0.003	2.404	1.8087	2.8401
222	2.395	-3.03	0.009	2.405	1.8812	2.8414
223	2.403	2.29	0.002	2.405	1.95	2.5597
224	2.401	-1.36	0.005	2.406	1.8765	2.7972
225	2.404	1.101	0.002	2.406	2.0084	2.8398
226	2.403	-1.699	0.003	2.406	1.8031	2.8108
227	2.409	1.825	-0.002	2.407	1.8728	2.807
228	2.407	1.348	0	2.407	1.8601	2.7975
229	2.4	-1.7	0.008	2.408	1.9438	2.7949
230	2.399	-2.129	0.009	2.408	1.9966	2.8058
231	2.408	2.605	0	2.409	2.0374	2.8027
232	2.406	-1.76	0.003	2.409	1.9728	2.8011
233	2.411	4.056	-0.002	2.409	1.9368	2.8189
234	2.399	-4.004	0.011	2.41	1.9117	2.8095
235	2.404	2.388	0.006	2.41	1.8931	2.6769
236	2.399	-3.438	0.012	2.411	1.9563	2.8226
237	2.408	1.291	0.003	2.411	2.089	2.7901
238	2.412	0.773	-0.001	2.411	1.9115	2.8044
239	2.413	1.678	-0.001	2.412	2.018	2.8097
240	2.406	-1.178	0.006	2.412	1.8901	2.8215
241	2.405	-0.767	0.008	2.413	2.0521	2.8164
242	2.407	-1.372	0.006	2.413	2.2354	2.8054
243	2.414	2.955	-0.001	2.413	1.9821	2.8064
244	2.409	-1.185	0.004	2.414	1.8799	2.8016
245	2.409	-1.643	0.005	2.414	1.8995	2.8114
246	2.416	2.747	-0.002	2.415	1.9773	2.6928
247	2.412	-0.04	0.003	2.415	1.9583	2.8136
248	2.408	-1.011	0.008	2.415	1.9565	2.8119
249	2.407	-1.323	0.008	2.416	1.8716	2.8199
250	2.413	0.507	0.003	2.416	1.9414	2.8089
251	2.416	2.35	0.001	2.416	1.8711	2.8081
252	2.409	-2.298	0.007	2.417	1.9785	2.8036
253	2.412	1.733	0.005	2.417	2.1984	2.8152
254	2.408	-1.443	0.009	2.418	1.8236	2.8187
255	2.41	0.946	0.008	2.418	1.9655	2.8165

256	2.408	-2.173	0.01	2.418	2.0632	2.8137
257	2.415	2.341	0.004	2.419	2.0615	2.6895
258	2.412	-0.582	0.007	2.419	2.0551	2.8087
259	2.412	0.839	0.008	2.419	1.9697	2.8189
260	2.408	-2.273	0.012	2.42	2.0322	2.8173
261	2.413	0.261	0.007	2.42	1.9531	2.821
262	2.417	3.694	0.003	2.42	2.0222	2.8195
263	2.407	-4.895	0.013	2.421	1.9912	2.822
264	2.416	2.584	0.005	2.421	1.8861	2.8254
265	2.415	-1.123	0.007	2.421	1.8384	2.8241
266	2.418	5.166	0.004	2.422	2.0924	2.8172
267	2.402	-8.41	0.021	2.422	2.0279	2.8172
268	2.417	3.971	0.006	2.422	1.8984	2.6986
269	2.417	1.925	0.006	2.423	2.0506	2.8212
270	2.41	-2.247	0.013	2.423	2.0385	2.8213
271	2.412	-1.759	0.012	2.423	2.0864	2.8125
272	2.42	2.056	0.004	2.424	2.0563	2.8258
273	2.42	-0.239	0.004	2.424	2.0789	2.8241
274	2.421	1.28	0.003	2.424	2.1547	2.8249
275	2.418	-1.597	0.007	2.425	2.0757	2.8168
276	2.42	0.968	0.005	2.425	2.1079	2.8139
277	2.419	2.029	0.006	2.425	2.093	2.8205
278	2.411	-4.289	0.015	2.426	2.2127	2.8234
279	2.418	1.1	0.008	2.426	2.056	2.6738
280	2.421	0.667	0.005	2.426	1.9647	2.8325
281	2.422	0.974	0.005	2.427	2.1498	2.8263
282	2.419	-1.014	0.008	2.427	1.9426	2.8232
283	2.42	-0.292	0.008	2.427	2.1954	2.8248
284	2.422	1.855	0.006	2.428	2.0281	2.823
285	2.417	-2.817	0.011	2.428	2.1992	2.8168
286	2.422	1.623	0.006	2.428	2.0689	2.8251
287	2.422	-0.896	0.007	2.429	2.2374	2.8146
288	2.424	2.951	0.005	2.429	1.9727	2.8258
289	2.417	-3.996	0.013	2.429	2.1763	2.83
290	2.423	1.275	0.007	2.429	2.0878	2.7021
291	2.425	3.062	0.005	2.43	2.1015	2.8137
292	2.416	-5.35	0.014	2.43	2.0519	2.8284
293	2.426	5.238	0.005	2.43	1.9965	2.8305
294	2.417	-2.993	0.013	2.431	2.092	2.8297
295	2.419	-0.598	0.012	2.431	2.1588	2.8309
296	2.423	0.72	0.008	2.431	2.1246	2.8304
297	2.425	0.85	0.007	2.432	2.0117	2.8214
298	2.423	-1.594	0.009	2.432	2.0612	2.8324
299	2.427	1.819	0.005	2.432	2.0539	2.8307
300	2.425	-	0.007	2.432	2.3497	2.8296

Table S4 Experimental result values obtained by GA, PSO, RPSO for d=30 through 50 independent runs

F	GA					PSO					RPSO				
	Best	Worst	Mean	S.D.	T(s)	Best	Worst	Mean	S.D.	T(s)	Best	Worst	Mean	S.D.	T(s)
1	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	47.14	1.11E-06	3.67E-06	2.13E-06	7.04E-07	36.44	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	0.1586
2	1.30E-04	8,72E-04	4.23E-04	1.59E-04	53.18	1.20E-02	7.42E-02	3.42E-02	1.73E-02	42.33	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**2.071**
3	2.89E+00	9.04E+01	7.90E+01	1.85E+01	62.66	1.12E+01	2.17E+02	5.14E+01	5.15E+01	51.47	**0.00E+00**	**3.99E+00**	**1.51E+00**	**1.95E+00**	**22.98**
4	**0.00E+00**	1.25E+03	2.64E+01	9.12E+02	68.54	0.00E+00	5.76E+04	3.45E+03	6.67E+02	54.78	**0.00E+00**	**1.02E+02**	**5.21E+01**	**3.24E+01**	**12.77**
5	**0.00E+00**	1.00E-01	2.00E-02	1.41E-01	55.46	9.95E+00	2.49E+01	1.58E+01	3.54E+00	45.49	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.06321**
6	5.56E-05	2.62E-04	1.13E-04	4.36E-05	67.83	1.31E-03	2.01E+01	9.70E+00	9.88E+00	57.53	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**5.464**
7	7.40E-03	3.30E-01	1.03E-01	7.33E-02	67.62	3.95E-07	8.10E-02	1.50E-02	1.98E-02	56.28	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**4.253**
8	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	124.1	1.96E-07	1.51E+01	2.25E+00	2.60E+00	108.8	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**16.31**
9	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	123.1	1.87E-06	7.73E-06	3.81E-06	1.22E-06	109.0	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**3.391**
10	**0.00E+00**	8.62E-07	0.00E+00	1.50E-07	57.48	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	0.6021	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.1101**
11	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	61.08	1.18E-06	4.52E-06	2.06E-06	6.30E-07	49.63	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.1674**
12	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	75.91	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	0.1987	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.007233**
13	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	51.72	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	42.51	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.5478**
14	9.90E-02	2.72E-01	1.83E-01	**4.32E-02**	69.77	6.82E-02	2.72E-01	1.39E-01	4.64E-02	59.35	**1.90E-02**	4.57E-01	**4.68E-02**	8.53E-02	47.48
15	2.67E-05	2.70E-03	7.64E-04	6.12E-04	69.63	1.78E-01	2.74E+00	7.96E-01	5.78E-01	58.90	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.00E+00**	**0.6101**

S.D., standard deviation, T, mean CPU time. The best experimental result values are marked bold.

Supplementary Figures

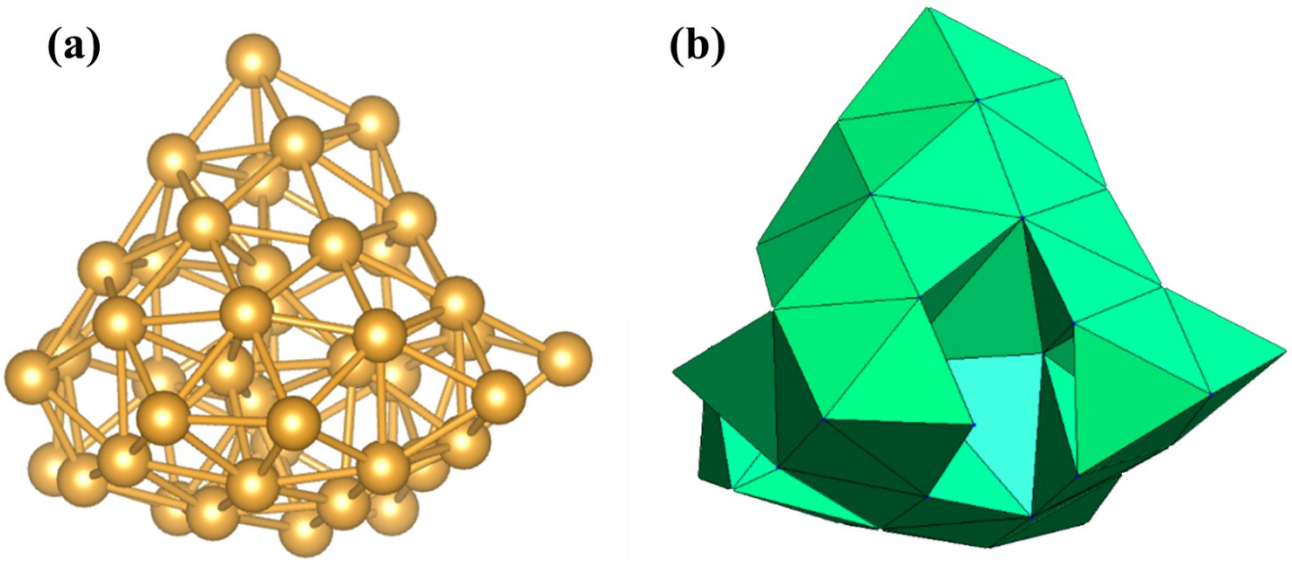

Figure S1. (a) Au_{42} cluster in ball-and-stick view. (b) Alpha shape corresponding to Au_{42} cluster.

Figure S2 (a). Snapshots of structures of Au_N cluster (N=21-119) obtained by RPSO.

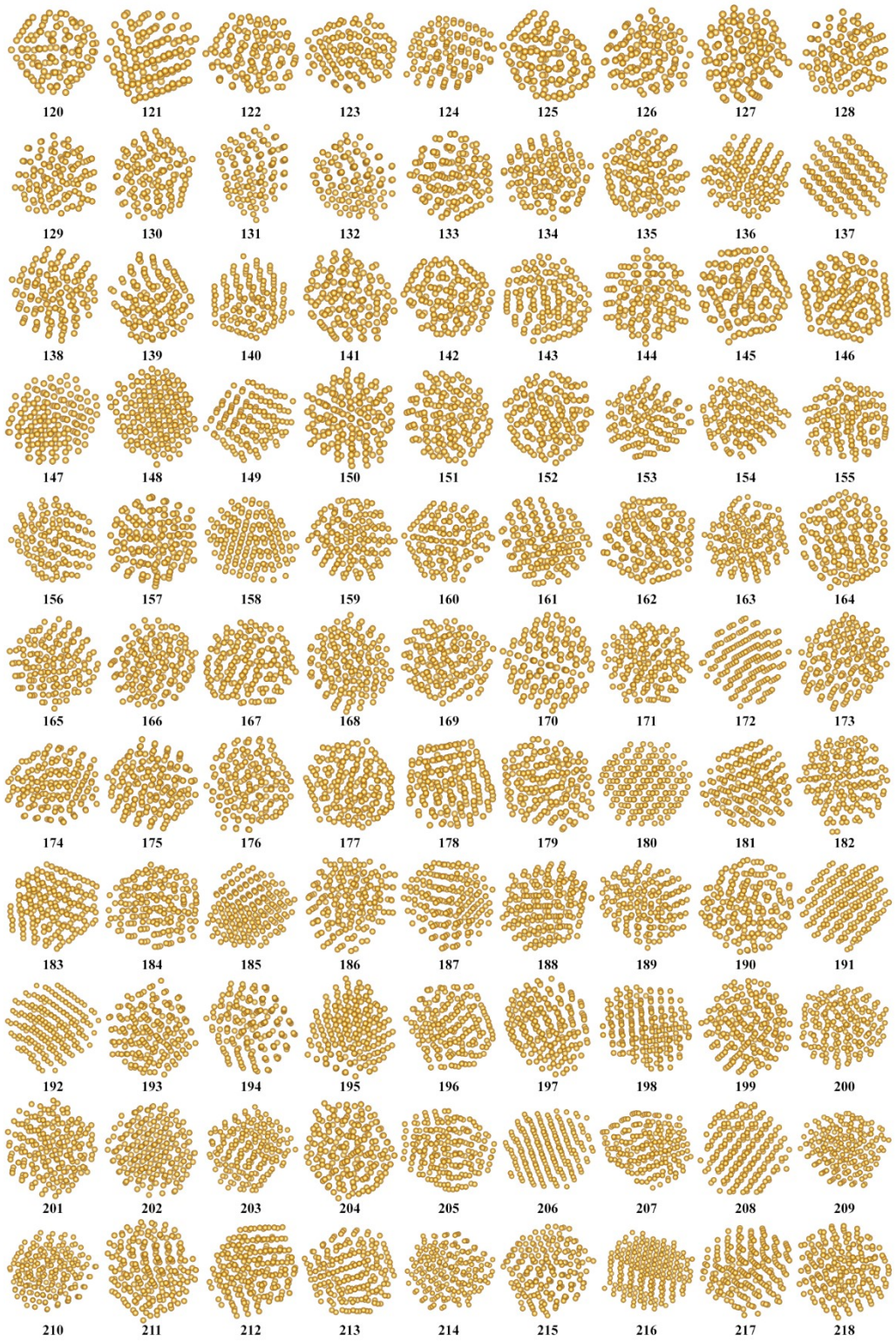

Figure S2 (b). Snapshots of structures of Au_N cluster (N=120-218) obtained by RPSO.

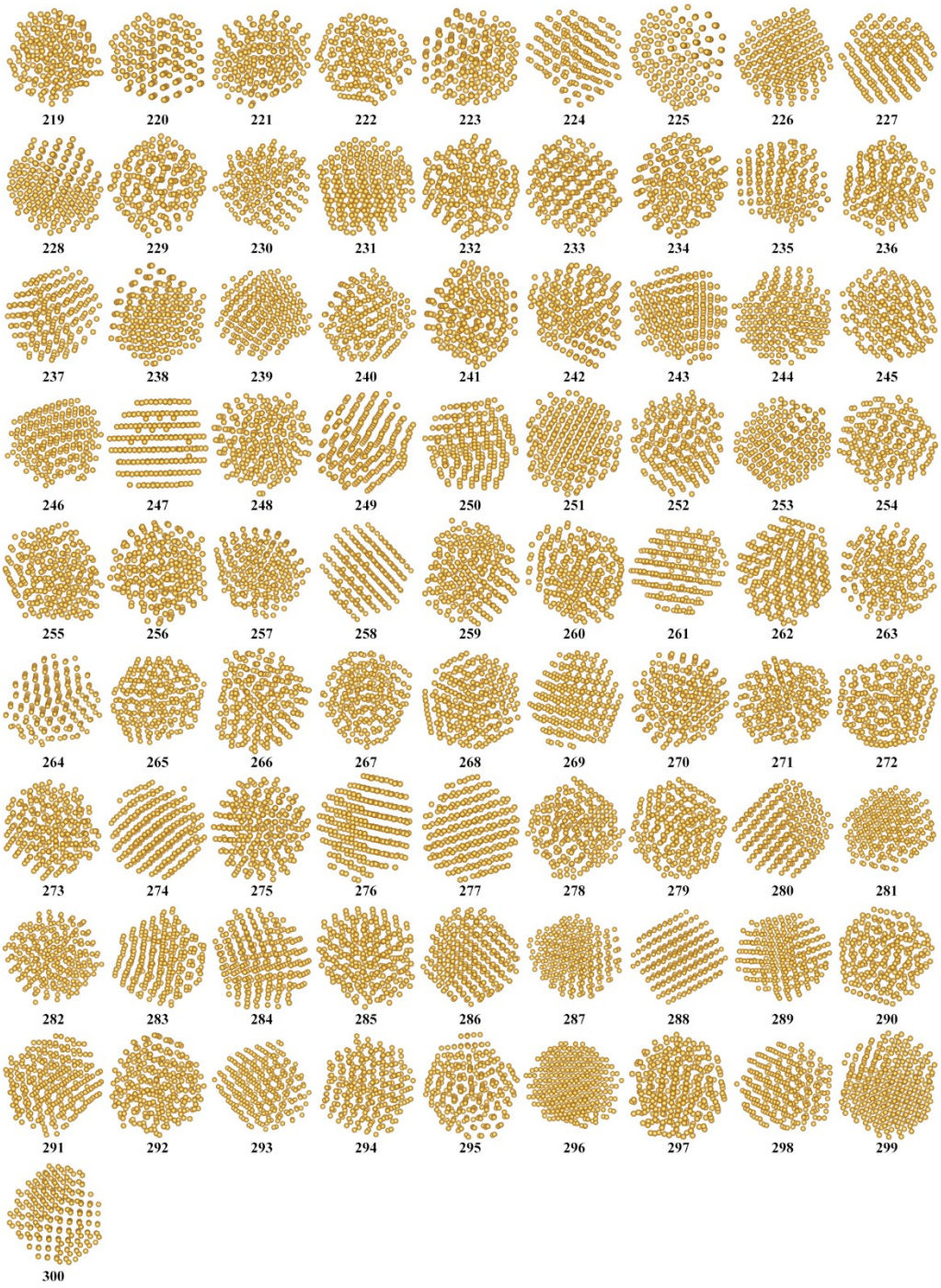

Figure S2 (c). Snapshots of structures of Au_N cluster (N=219-300) obtained by RPSO.

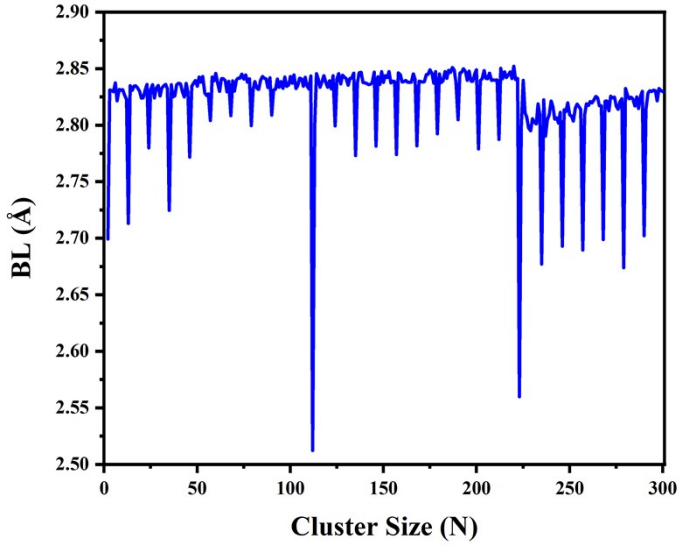

Figure S3. Average bond length for Au_N(2-300) clusters obtained by RPSO.

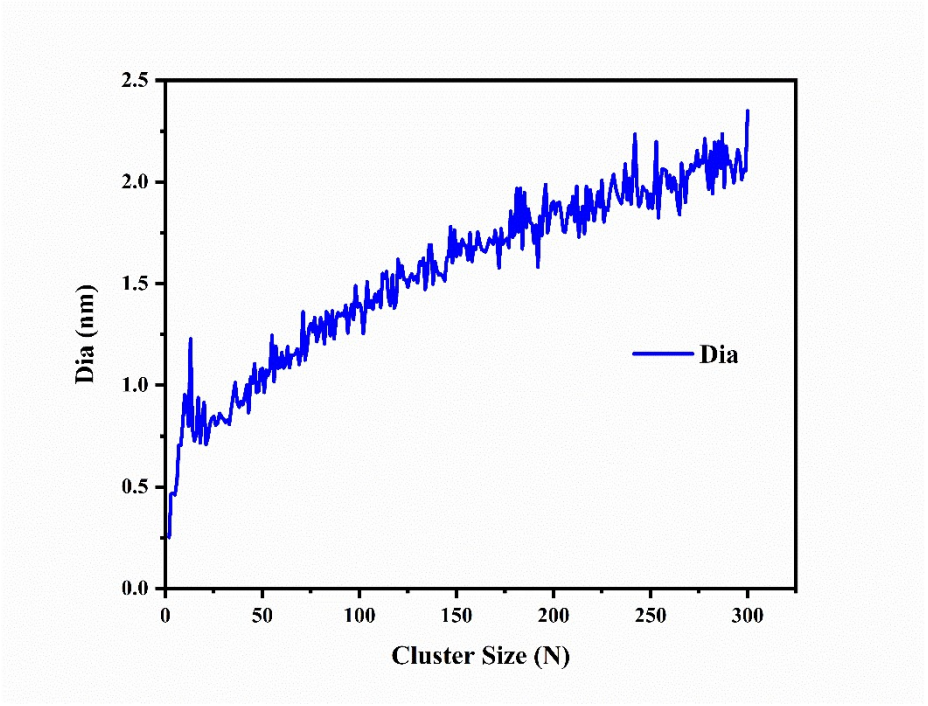

Figure S4. Dia (effective diameters) of Au_N (N=2-300) obtained by RPSO.

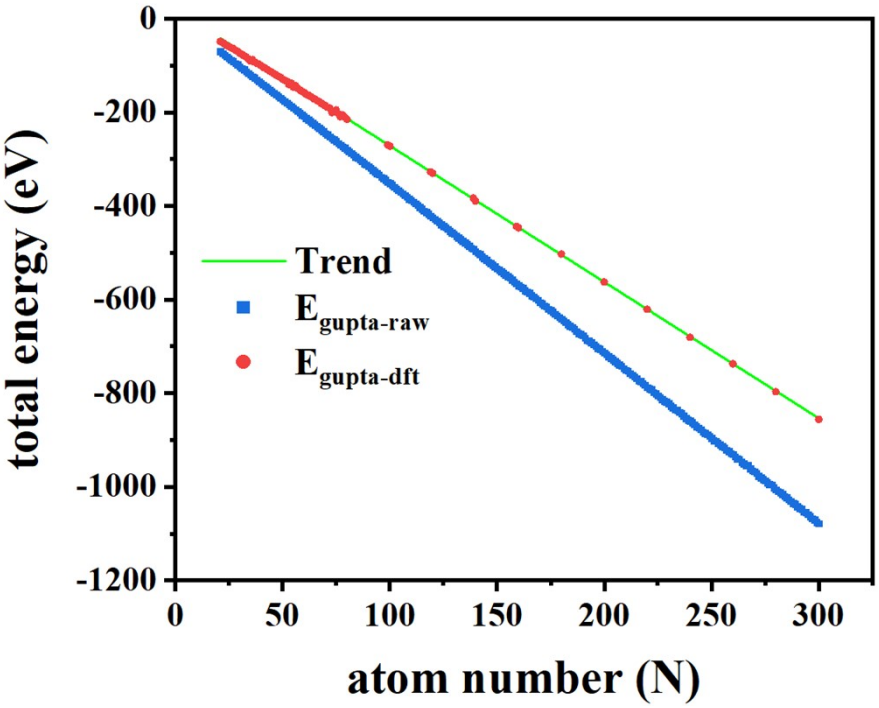

Figure S5. Schematic diagram of system energy correction. Blue square ($E_{gupta-raw}$) means the original energy of Au_N(N=2-300) obtained by RPSO at the Gupta level. The red dot ($E_{gupta-dft}$) means recalculated energy data point of the same structural configurations using VASP, selected Au_N scatter size is 21-80, 99, 100,119,120,139,140,159,160,200,220,240,260,280,300. The green trend line reflects the revised system energy trend.

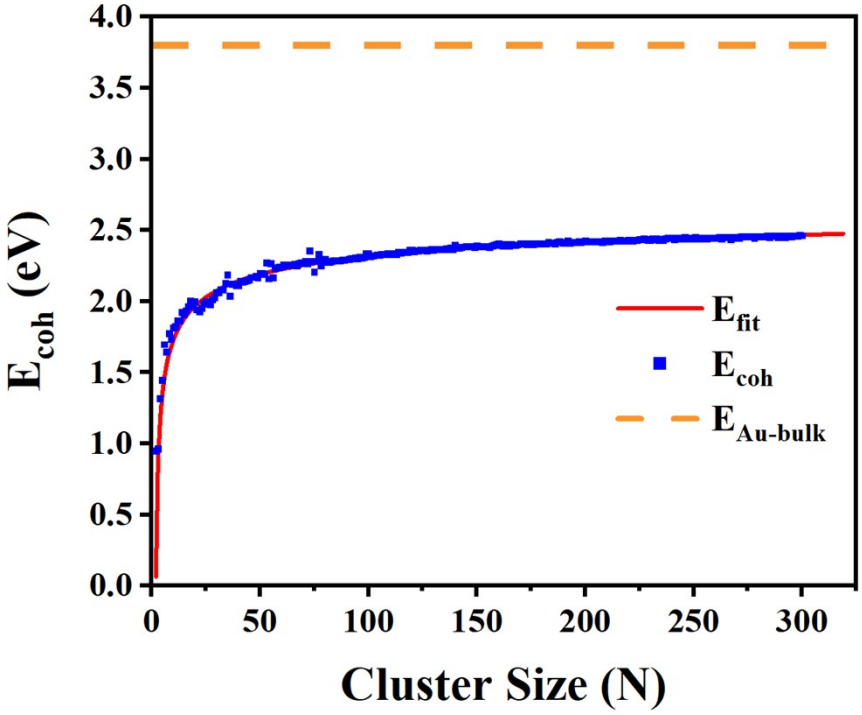

Figure S6. Cohesive energy of Au_N(N=2-300). Horizontal dashed line represents the experimental cohesive energy of bulk gold, which is 3.8 eV^2.

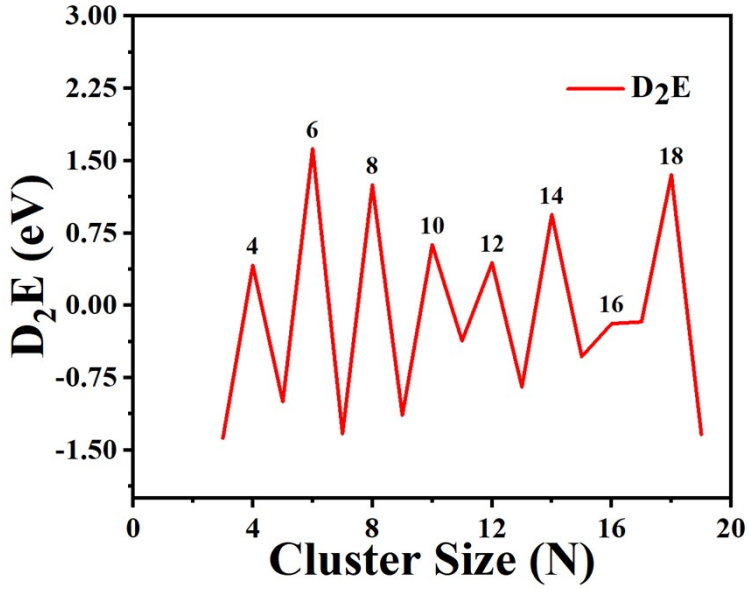

Figure S7. D_2E (Second finite difference) of Au_N (N=2-20). Black label numbers correspond to more stable sizes.

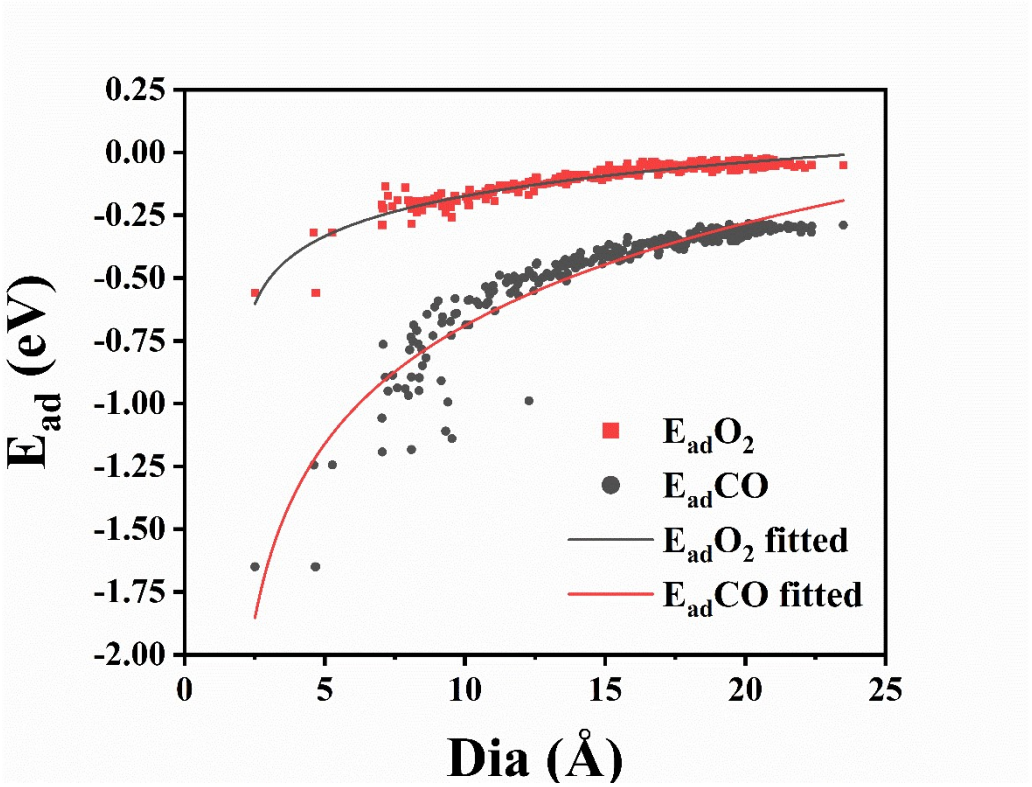

Figure S8. Average adsorption energy of CO oxidation upon Au_N cluster with effect diameter denoted as Dia. Red dot represents adsorption energy of CO, which follows linear logarithmic relationship as $y = -1.793 + 0.519\ln(x - 1.623)$ with $R^2 = 0.88$. Black dot represents adsorption energy of molecular O_2, which follows linear logarithmic relationship as $y = -0.523 + 0.167\ln(x - 1.887)$ with $R^2 = 0.88$.

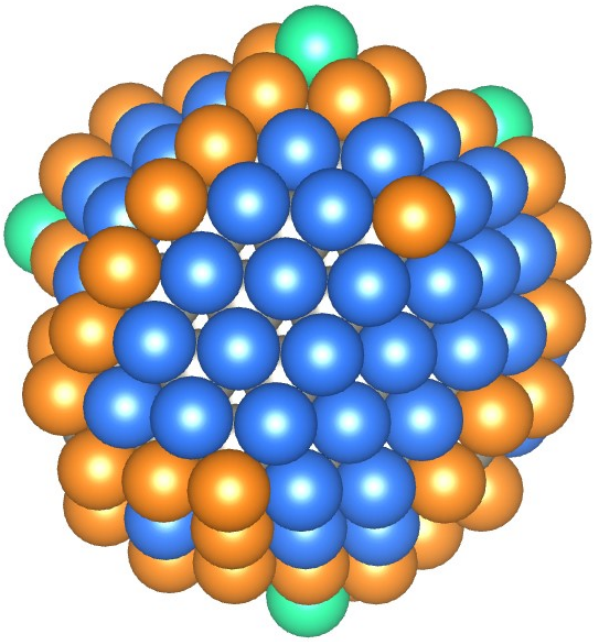

Figure S9. Schematic diagram of geometric site distribution using Au_{199} as a demo system. Blue atoms denote *Face* sites, orange atoms denote *Edge* sites, and green atoms denote *Kink* sites.

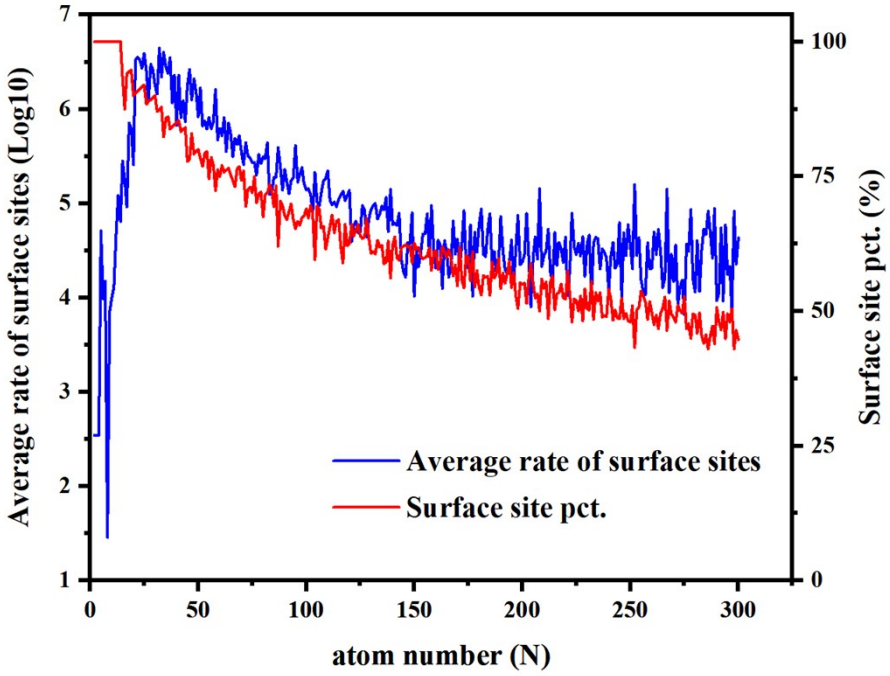

Figure S10. Blue line: average predicted reaction rate of surface sites. Red line: percentage of surface sites to total number of atoms in cluster.

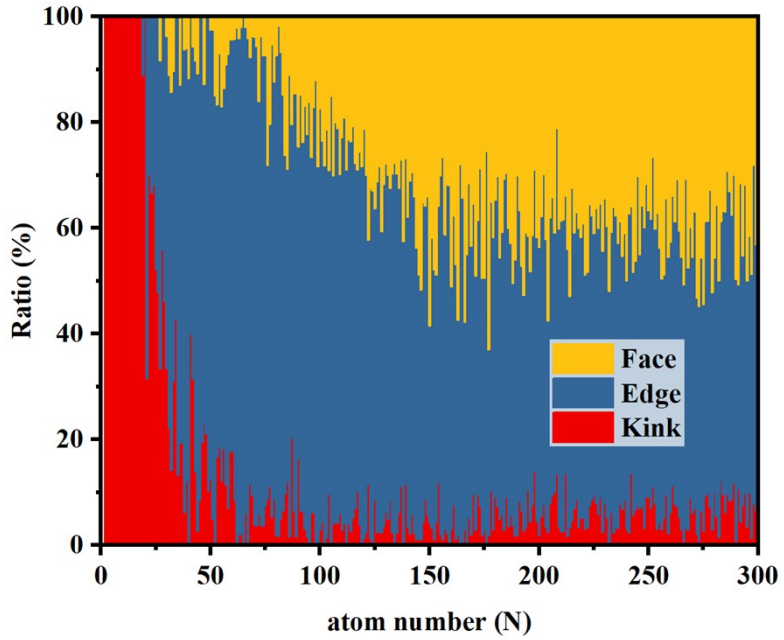

Figure S11. The ratio of different types of sites (*Kink*, *Edge* and *Face*) to the total number of surface sites.

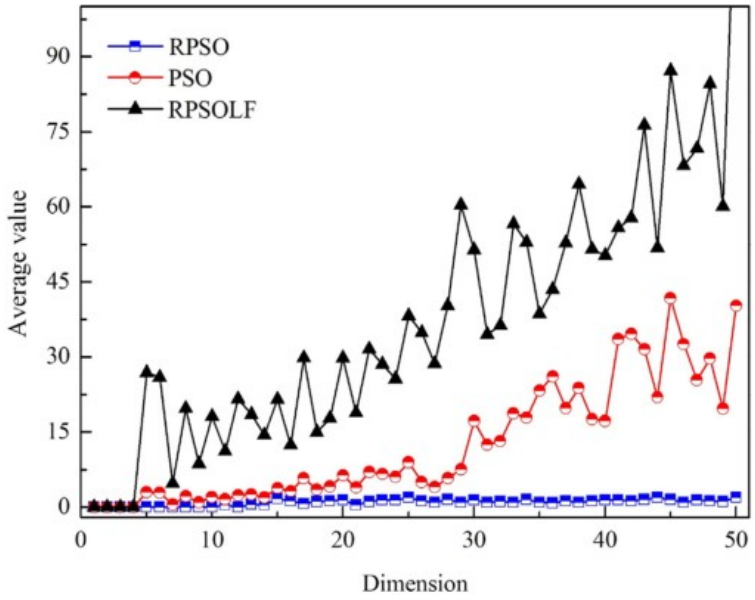

Figure S12. The comparison of RPSO, RPSOLF and PSO algorithms results for different dimensions of Rosenbrock function. Each point represents the mean values of the given dimension of the Rosenbrock function obtained by three algorithms starting from the same initial solution. Cited from ref [3].

Reference

(1) Tailoring of Pd-Pt Bimetallic Clusters with High Stability for Oxygen Reduction Reaction.Cheng D, Wang W.

(2) Assadollahzadeh, B.; Schwerdtfeger, P. A Systematic Search for Minimum Structures of Small Gold Clusters Aun (N=2–20) and Their Electronic Properties. *The Journal of Chemical Physics* **2009**, *131* (6), 064306.

(3) Zhou, Y.; Zhao, Z.; Cheng, D. Cluster Structure Prediction via Revised Particle-Swarm Optimization Algorithm. *Computer Physics Communications* **2020**, *247*, 106945.