Exploring the Stable Structure of Cerium Oxide Nanoclusters using High-Dimensional Neural Network Potential

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The high-dimensional neural network[1, 2]

The main idea of this neural network is to represent the total energy as the sum of the energies of all the atoms,

$$E = \sum_i E_i$$

.

The E_i represent the energy of atom i. The atomic energies are trained by atomic neural network using the symmetry functions to represent the structural and chemical environment information of each atom. In this work, we choose the following three symmetry functions to represent local chemical environment of cerium atoms and oxygen atoms from Cartesian coordinates,

Radial symmetry function

$$G_i^{radial} = \sum_{j \neq i} e^{-\eta(r_{ij} - r_s)^2} f_c(r_{ij})$$

Narrow angular symmetry function

$$G_i^{ang.n.} = 2^{1-\zeta} \sum_{\substack{j,k \neq i \\ j<k}} (1 + \lambda \cos\theta_{ijk})^{\zeta} e^{-\eta((r_{ij} - r_s)^2 + (r_{ik} - r_s)^2 + (r_{jk} - r_s)^2)} f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk})$$

Wide angular symmetry function

$$G_i^{ang.w.} = 2^{1-\zeta} \sum_{\substack{j,k \neq i \\ j<k}} (1 + \lambda \cos\theta_{ijk})^{\zeta} e^{-\eta((r_{ij} - r_s)^2 + (r_{ik} - r_s)^2)} f_c(r_{ij}) f_c(r_{ik})$$

.

$f_c(r_{ij})$ represents the cutoff function, which has the form

$$f_c(r_{ij}) = \begin{cases} 0.5 \times \left[\cos\left(\frac{\pi r_{ij}}{r_c}\right) + 1\right] & for\ r_{ij} \leq r_c, \\ 0 & for\ r_{ij} > r_c. \end{cases}$$

r_{ij} represents the distance between atom i and atom j. r_c represents the cutoff radius.

Table S1: Parameters of symmetry functions used for neural network performed using the n2p2 code[3]

Radial			Angular				
η	r_s	r_c	η	λ	ζ	r_s	r_c
2	1.5	6	0.2222	-1	1	0	6
2	2	6	0.2222	1	1	0	6
2	2.5	6	0.2222	-1	6	0	6
2	3	6	0.2222	1	6	0	6
2	3.5	6	0.0408	-1	1	0	6
2	4	6	0.0408	1	1	0	6
2	4.5	6	0.0408	-1	6	0	6
2	5	6	0.0408	1	6	0	6
2	5.5	6	0.0165	-1	1	0	6
			0.0165	1	1	0	6
			0.0165	-1	6	0	6
			0.0165	1	6	0	6

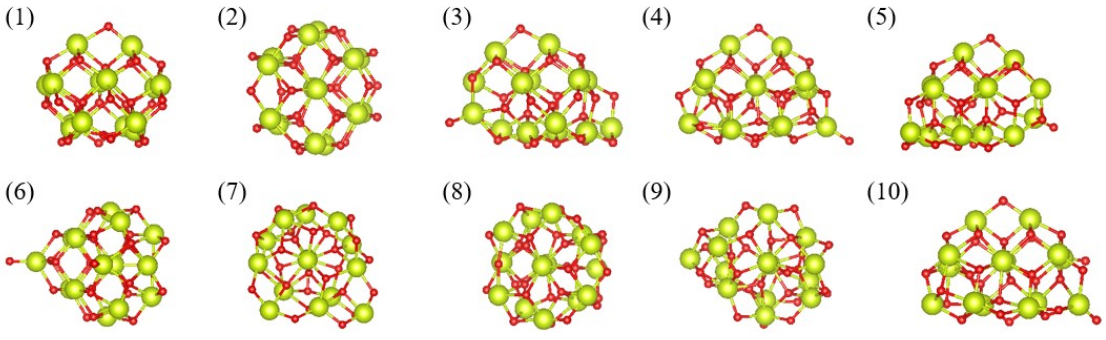

Fig.S1 The top 10 minimums of nanoclusters $Ce_{14}O_{27}$. (red: oxygen atoms, green: cerium atoms)

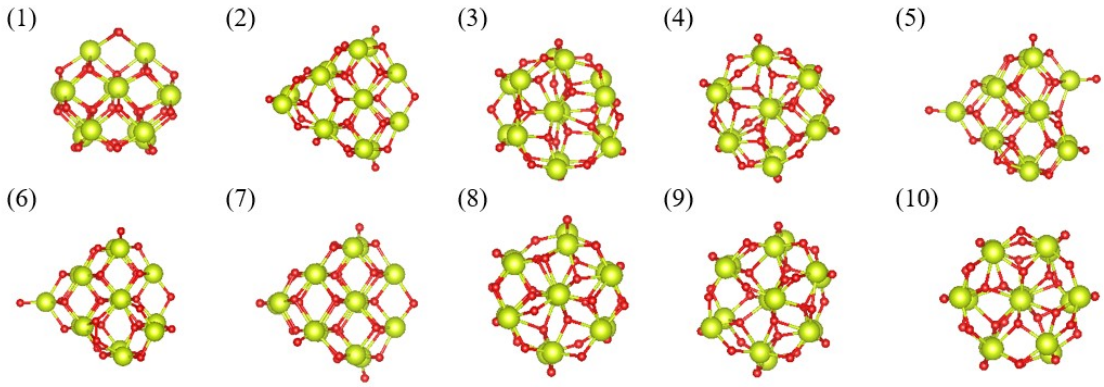

Fig.S2 The top 10 minimums of nanoclusters $Ce_{14}O_{28}$. (red: oxygen atoms, green: cerium atoms)

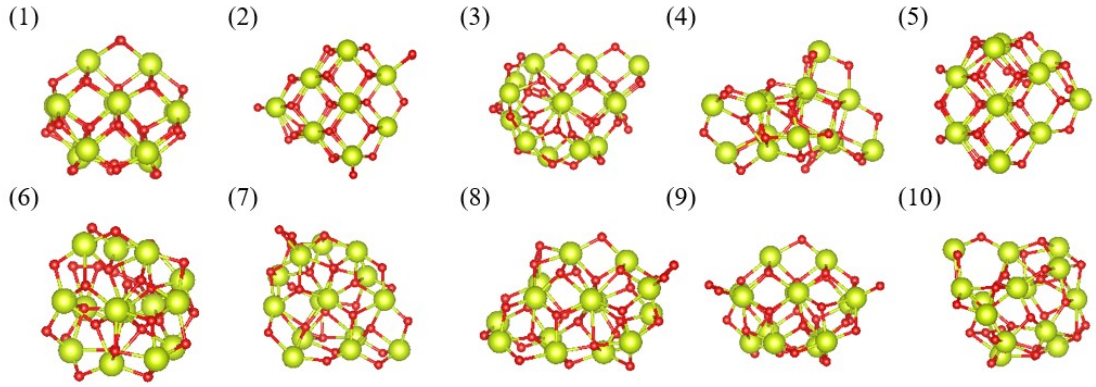

Fig.S3 The top 10 minimums of nanoclusters $Ce_{14}O_{29}$. (red: oxygen atoms, green: cerium atoms)

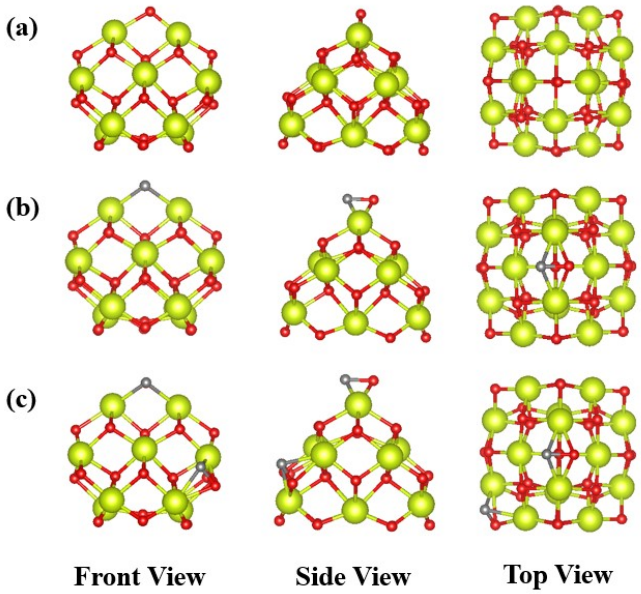

Fig.S4 The structures comparison of $Ce_{14}O_{27}$, $Ce_{14}O_{28}$, and $Ce_{14}O_{29}$. (red: oxygen atoms, grey: addition oxygen atoms, green: cerium atoms)

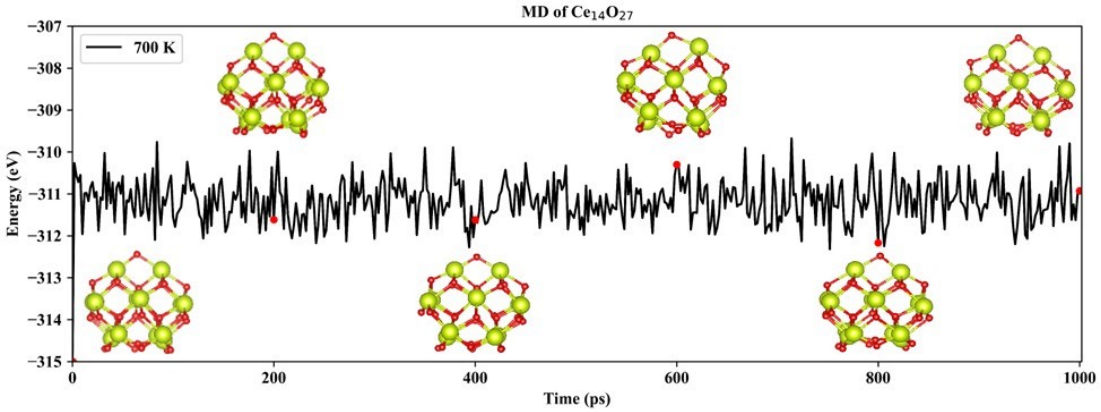

Fig.S5 Snapshots of the most stable nanocluster $Ce_{14}O_{27}$ extracted every 200 ps from 700 K MD trajectories.

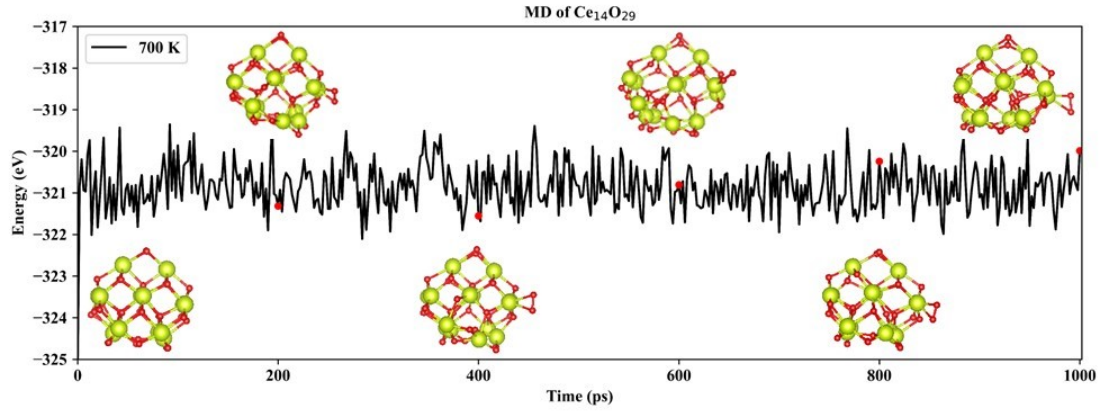

Fig.S6 Snapshots of the most stable nanocluster $Ce_{14}O_{29}$ extracted every 200 ps from 700 K MD trajectories.

The coordinates of the most stable $Ce_{14}O_{27}$ (.xyz format)

41

O Ce

O	7.663066	9.886884	8.103913
O	11.654997	11.078156	9.731121
O	9.681866	9.432853	4.612806

O	7.711274	8.107805	5.915162
O	9.573336	10.029844	11.371962
O	11.457870	8.050962	6.266395
O	11.667717	13.059427	11.707449
O	11.250403	6.227804	4.245559
O	13.670287	9.744797	11.261906
O	9.669910	12.858351	8.896226
O	8.077448	12.420979	5.787269
O	11.558014	8.327799	12.479060
O	11.568368	14.271835	7.654282
O	15.609321	9.182742	8.524478
O	7.955402	7.041839	10.587491
O	14.887578	6.567616	10.280367
O	13.097531	8.192351	9.012982
O	9.854687	8.388128	9.118485
O	10.015035	10.888994	6.975340
O	13.023679	8.937119	4.200226
O	15.139023	7.309918	5.964425
O	13.113317	10.570622	7.006850
O	13.156794	6.066869	7.220040
O	13.878901	12.633309	9.404819
O	9.465807	6.323635	7.189558
O	11.250365	12.421795	5.075002
O	11.414563	6.492488	10.462792
Ce	11.404906	6.876673	8.230675
Ce	10.049023	13.222273	6.607650
Ce	11.790135	10.331182	5.384389
Ce	9.652025	7.252866	5.105847
Ce	11.895367	13.441673	9.660338
Ce	15.037662	7.229935	8.122348
Ce	8.072752	10.347711	5.899452
Ce	11.735417	10.808419	12.058903
Ce	9.484694	10.720157	9.208440
Ce	7.803768	7.559018	8.305671
Ce	9.715111	7.770212	11.370380
Ce	13.314571	7.588308	11.215500
Ce	13.914300	10.533986	8.997531
Ce	13.209753	7.047938	5.062960

The coordinates of the most stable $Ce_{14}O_{28}$ (.xyz format)

42

O Ce

O	10.115682	5.811046	9.956292
O	12.990373	13.383770	8.654510

O	12.610801	6.580429	8.866866
O	8.528120	11.418891	11.179386
O	14.773922	9.809621	9.221152
O	14.169817	7.568423	11.813012
O	9.682199	14.009515	8.130670
O	12.272331	12.544620	5.431630
O	10.772294	12.019706	9.665606
O	6.712742	8.447321	10.577548
O	9.518198	8.410007	4.978011
O	11.358785	5.859905	5.785236
O	11.093703	8.196656	7.199659
O	14.411002	12.216060	10.652491
O	12.267140	9.694798	3.918391
O	8.699368	6.975352	7.958113
O	8.998705	9.363416	9.372544
O	9.812107	11.026721	6.893133
O	10.624299	14.276491	11.220259
O	12.194277	9.066133	10.211775
O	13.548058	8.626337	5.983358
O	6.912906	9.854389	7.668502
O	12.091511	11.300218	11.994450
O	9.926711	8.073148	11.641401
O	13.191690	8.114167	12.814864
O	12.867047	10.851174	7.492783
O	15.244422	11.681194	6.408547
O	7.051506	12.614040	9.318725
Ce	10.350452	10.212980	11.042562
Ce	6.960058	10.491956	9.666339
Ce	12.369885	13.195621	10.815004
Ce	13.154785	8.650676	8.136471
Ce	8.412161	7.295896	10.169769
Ce	10.725871	6.006104	7.781203
Ce	11.554510	7.924052	4.912853
Ce	8.845906	9.134656	6.935284
Ce	14.676706	11.908537	8.417803
Ce	13.512980	10.866552	5.231270
Ce	11.965221	6.888490	11.164495
Ce	8.926458	13.433464	9.995328
Ce	14.013224	9.939650	11.499090
Ce	11.163474	12.603255	7.321141

The coordinates of the most stable $Ce_{14}O_{29}$ (.xyz format)

43

O Ce

O	10.096267	5.802702	9.963305
O	13.023787	13.338359	8.661959
O	12.605649	6.561892	8.877320
O	8.538922	11.421655	11.175012
O	14.755538	9.784024	9.258139
O	14.151260	7.548267	11.848949
O	9.705625	13.975443	8.092520
O	12.241113	12.474038	5.395965
O	10.786941	12.008877	9.667855
O	6.694128	8.453777	10.571016
O	9.513947	8.404178	4.994242
O	11.339269	5.842449	5.801012
O	11.090933	8.182919	7.214348
O	14.427209	12.193623	10.681068
O	12.295611	9.674783	3.924985
O	8.681298	6.969016	7.967079
O	8.985276	9.358759	9.383656
O	9.839733	10.995024	6.917371
O	10.647105	14.280490	11.187314
O	12.188402	9.053157	10.212802
O	13.526678	8.566593	5.989969
O	6.918421	9.882491	7.667952
O	15.639878	10.960236	6.501083
O	12.092049	11.285251	12.005931
O	9.913522	8.064806	11.645858
O	13.159906	8.096968	12.841043
O	12.935908	10.846620	7.466771
O	15.174282	12.326839	6.214855
O	7.063369	12.623341	9.324018
Ce	10.346512	10.201652	11.038873
Ce	6.956644	10.497947	9.667779
Ce	12.386978	13.174287	10.826112
Ce	13.158199	8.624398	8.135381
Ce	8.395638	7.291010	10.176323
Ce	10.708621	5.994251	7.789719
Ce	11.554372	7.910556	4.925321
Ce	8.839733	9.122964	6.942354
Ce	14.696910	11.871054	8.460087
Ce	13.545364	10.845901	5.212686
Ce	11.950598	6.876740	11.170578
Ce	8.949909	13.422896	9.970196
Ce	13.999246	9.915459	11.523300
Ce	11.190536	12.579628	7.306636

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3. A. Singraber, T. Morawietz, J. Behler and C. Dellago, *J Chem Theory Comput*, 2019, **15**, 3075-3092.