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Study of Surface Reaction Mechanisms over CeO_2 (111) using Ultra Accelerated Quantum Chemical Molecular Dynamics.

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【Introduction】

Ceria (CeO_2) is an important catalyst in various industrial and environmental applications such as three-way catalyst (TWCs), which simultaneously convert hydrocarbon, CO and NO_x into CO_2 , H_2O and N_2. Ceria based oxide are being currently considered as strong candidates for low temperature water gas shift reaction, which converts CO and water to CO_2 and hydrogen. Most of these technological applications rely upon the characteristics feature of Ce ions that easily switch oxidation number between +4 to +3. In this study we use our ultra accelerated quantum chemical molecular dynamics system to investigate the surface reduction mechanism of CeO_2 (111) by atomic hydrogen.

【Method】

Recently we have successfully developed a novel tight-binding quantum chemical molecular dynamics program “New-Colors” based on our original tight-binding theory. This program is more than 10,000,000 times faster than the other conventional methods. Our Colors program enables us to examine huge catalyst system containing both ultra fine metal particle and metal oxide. We obtained the potentials between pair of atoms using the New-Colors program. Further molecular dynamics simulation was performed using the Ryudo program.

【Result and Discussion】

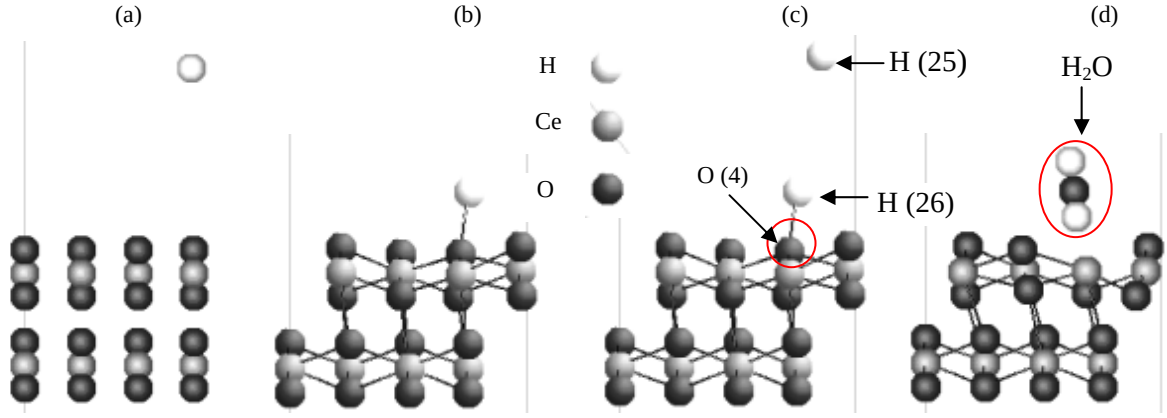

Figure 1. (a) Initial structure, (b) Snapshot at 175 fs the H atom has come in contact to the ceria surface, (c) Another one atomic H atom has been added to the model, (d) Water formation extraction of an oxygen atom from ceria surface.

The structure used in the simulation contains eight Ce atoms, sixteen oxygen atoms and one hydrogen atom. Initial structure Fig.1 (a) a hydrogen atom has been placed on the free surface above Ce and O atom. Distance between the H atom and the ceria surface is 6.17 Å, the calculation temperature was 873 K and given velocity of H atom was -3.9 km/s. In molecular dynamics simulation after 175 fs H atom interacts with ceria surface Fig1 (b). After 190 fs another H atom has been added to the model Fig 1(c) with same condition. After 250 fs both H (25), H (26) have come cantacts to ceria surface and bond formation with O (4), simultaneously they were trying to eliminate the oxygen atom. After1190 fs extraction of an oxygen atom from ceria surface form water Fig.1 (d) creating oxygen vacancies in the ceria surface. The bond energy of O-H is 98.22 kcal/mol, 109.11 kcal/mol and bond length is 1.09 Å, 1.005 Å, respectively in the water that form in this calculation.