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Ultra-accelerated quantum chemical molecular dynamics study of sintering dynamics of Pt/CeO_2 interface

Md. Khorshed Alam[1], Ryuji Miura[1], Ai Suzuki[2], Hideyuki Tsuboi[1], Nozomu Hatakeyama[1], Akira Endou[1], Hiromitsu Takaba[1], Momoji Kubo[1] and Akira Miyamoto[1,2]

[1] Department of Chemical Engineering /Applied Chemistry, Graduate School of Engineering, Tohoku University, (6-6-10-205 Aoba, Aramaki, Aoba-ku, Sendai, 980-8579, Japan)

[2]New Industry Creation Hatchery Center, Tohoku University, (6-6-10-205 Aoba, Aramaki, Aoba, Sendai 980-8579, Japan)

Introduction: A severe problem affects many applications due to loss of active surface area. It is a particularly pernicious problem in its applications under high temperature. Sintering causes a decrease in the catalytic activity. The lifetime of a vehicle catalyst is especially important in maintaining low exhaust gas emissions. Little work has been devoted to the theoretical evaluation of catalyst sintering behavior. Supported precious metal such as Pt, Rh and Pd are used to facilitate many industrial catalytic processes. We demonstrated macro-scale sintering behavior of $Pt/\gamma\text{-}Al_2O_3$, $Pt/ZrO_2$and Pt/CeO_2 catalysts using a developed 3D simulator and concluded that Pt on CeO_2 demonstrated the highest stability against sintering [1]. In order to develop more efficient and durable TWC, it is really meaningful to investigate and clarify the relationship between supported precious metals and ceria as a support with defect and without defect.

Method: Ultra accelerated quantum chemical molecular dynamics (UA-QCMD) have been used for this calculation. Tight binding (TB) used to obtain two body potentials and charges that were updated to perform MD simulation. TB and New-Ryudo were repeated for number of times for accuracy.

Results and Discussion: In the present study, we simulated Pt cluster supported on the CeO_2 surface to study the adsorption properties, dynamical properties of Pt/CeO_2 interface with defect and without defect ceria surface. We considered the adsorption energy of Pt37 on the reduced and unreduced ceria surfaces. The calculated adsorption energy of Pt37 for ceria surface is –1936.4 kcal/mol other hand, for defect ceria surface is –2142.3 kcal/mol. Molecular dynamics (MD) simulation were carried out for $1.0X10^5$ steps with a 1.0 fs time increment under a realistic durability temperature condition, 1073 K as shown in Fig.1. We also considered diffusion characteristics of Pt for defect free surface as well as defect surface and compared bond energies, bond lengths and bond population of Pt-O, Pt-Ce and Pt-Pt. These differences in binding energies of Pt-O bonds lead to the difference of sintering behavior for different types of support.

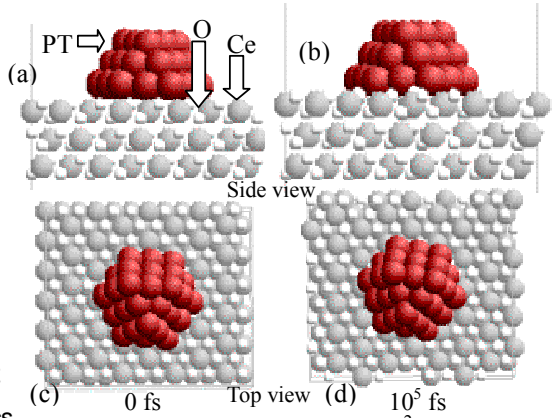

Fig.1 Side and top view of Pt/CeO^2 interface at 0fs (a), (c) and after 10^5 fs (b), (d).

1) A. Suzuki, et al. *Surface science*, **603,** 3049(2009)