

DFT Study of Adsorption Properties of Reaction Intermediates under Electric Potential in Solid Oxide Fuel Cell

○Shixue Liu, Teppei Ogura, Takayoshi Ishimoto, Michihisa Koyama (Kyushu Univ., JST-CREST)
 Motooka 744, Nishi-ku, Fukuoka 819-0395, Japan
 Phone: +81-92-802-6970, Fax: +81-92-802-6970
 sxliu@ifrc.kyushu-u.ac.jp

Methane reforming reaction under electric potential is studied using DFT method. SIESTA and effective screening medium (ESM) [1] method are used. Different from conventional DFT calculations, an electric potential is applied by adding net charge to the catalyst surface, and an effective metal electrode with opposite charge on the surface. The stable adsorption structures can be compared for different net charge, referring to different electric potential. In this report, we focus on the adsorption properties of reaction intermediates.

Generalized gradient approximation (GGA) with Perdew-Burke-Ernzerhos (PBE) exchange and correlation functionals are used in the DFT calculations, with spin polarized. Fig. 1 shows the pure nickel slab with the schematic effective metal electrode. The nickel lattice parameter is optimized firstly, 3.517 Å, which fits well with experimental value of 3.524 Å. The nickel catalyst is simulated by three layers slab of (111) surfaces, with 6 Å top vacuum layer and 2 Å bottom layer. Every nickel layer include 2x2 unit cell. Different net charge numbers will be used to simulate various SOFC operating voltage. The electric potential is performed and calculated by ESM method. The adsorption structures of intermediates of methane reforming reaction, such as CH₂, CH, CO, are fully optimized on the nickel surface.

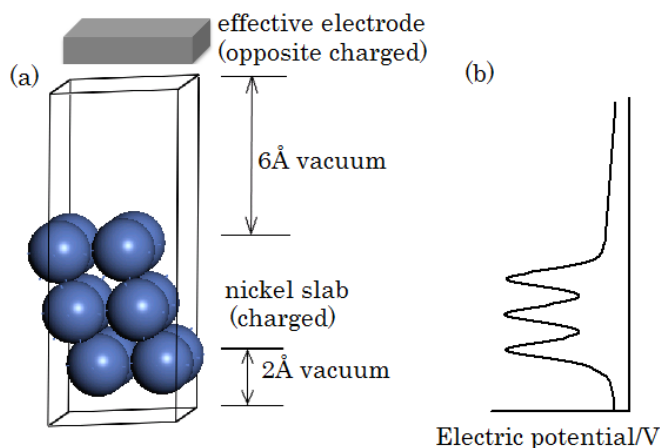


Fig. 1 (a) Nickel slab model with schematic illustration of effective metal electrode and (b) schematic of electric potential by ESM method.

Table 1 shows the binding energy of eight intermediates calculated by SIESTA with zero net charge. All the intermediates are adsorbed on the hcp site as shown in Fig. 2.

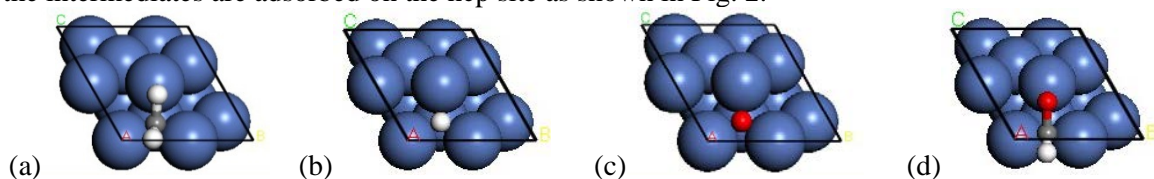


Fig. 2 Top view of the adsorbed intermediates on hcp site of Ni(111) (a) CH₂, (b) CH, H, (c) O and (d) CHO.

Table 1. Binding energy (eV) calculated by SIESTA (Net charge=0).

	CH ₂	CH	C	O	H	CO	CHO
SIESTA	4.27	7.08	7.66	4.90	2.75	2.39	3.54

Reference:

[1] M. Otani, O. Sugino, Phys. Rev. B 73 (2006) 115407.