2nd World Congress and Expo on
Nanotechnology and Material Science
April 04-06, 2016 at Dubai, UAE

Density Functional Theory Study of the Adsorption and Dissociation of H_2S on the Bimetallic Ni/Pd(111) Surfaces

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Hydrogen sulfide (H_2S) molecule is the most common sulfur-containing compound presented in fossil derived fuels, chemical feedstocks, and syngas, and the removal of H_2S is a significant concern because it is directly related to the preventing the catalysts from poisoning, protecting pipeline from corrosion and stemming from the stringent environmental regulations.

We have performed periodical density-functional theory calculations to investigate the adsorption structures and dissociative reaction pathways for H_2S on different monometallic and bimetallic Ni/Pd(111) surfaces. For the most energetically favorable chemisorption structures on each surface, the corresponding adsorption energies arrange in the following order: Ni-Pd-Ni(111) > Pd(111) > Pd-Ni(111) > Ni(111) > Pd-Ni-Pd(111) > Ni-Pd(111).

For the complete dissociations of H_2S on the various surfaces, except the pure Pd(111) , Pd-Ni-Pd, Pd-Ni surfaces, the dissociations of H_2S on the pure Ni(111) and other bimetallic surfaces are quite exothermic, and the values of E_{ads} are larger than 2.0 eV. Meanwhile, because of the overlap between the $2b_1/5a_1$ states(namely, two lone pairs of S atom) and the surface d band, the DOS peaks of $2b_1$ and $5a_1$ orbitals disappeared, while two other sharp features associated $4a_1$ and $2b_2$(namely, two S-H σ bonds) orbitals still exist and move away from the fermi level. With the climbing-image nudged elastic band method, we have obtained the minimum energy path for H_2S on the various surfaces. Two elementary steps ($H_2S_{(ads)} \rightarrow SH_{(ads)} + H_{(ads)}$; $H_{(ads)} + SH_{(ads)} \rightarrow S_{(ads)} + 2H_{(ads)}$) are found for all monometallic and bimetallic Ni/Pd metal surfaces. Correspondingly, two transition states which are defined as TS_1 and TS_2 are identified. The first dissociation barrier on Ni-Pd-Ni(111) surface is the smallest (0.16eV), indicating that the dissociative adsorption reaction is most likely to be occurred. In addition, for the case of Ni-Pd(111) surface, H_2S tends to be desorbed before decomposing reaction happened because of the energy barrier of TS1 (0.31 eV) is larger than the binding energy of H_2S molecule (0.21 eV), indicating that Ni-Pd(111) surface will exhibit a good sulfur resistance performance.

References

1 L.Yang, Z.Cheng, M.L.Liu, L.Wilson, Energy Environ. Sci. 3 (2010)1804-1809.

2 E.I.Kauppia,J.M.Kanervoa,J.Lehtonena, L.Lefferts, Appl. Catal. B: Environ.164 (2015) 360- 370.

3 E. Ozdogan, J. Wilcox, J. Phys. Chem. B. 114 (2010) 12851-12858.