

STUDI KOMPUTASI MEKANISME REAKSI OKSIDASI ETANOL PADA KATALIS PdCu (111) MENGGUNAKAN PENDEKATAN TEORI FUNGSI KERAPATAN

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INTISARI

Telah dilakukan kajian kimia komputasi mekanisme reaksi oksidasi etanol pada katalis PdCu (111) menggunakan pendekatan teori fungsi kerapatan dengan tujuan mengetahui posisi interaksi terstabil dari spesi reaksi oksidasi etanol pada permukaan PdCu (111), pengaruh variasi *doping* Cu terhadap sifat elektronik katalis PdCu (111), dan pengaruh persebaran *co-catalyst* (Cu) pada katalis PdCu (111) untuk mekanisme pembentukan asam asetat dan pemutusan ikatan C–C dalam reaksi oksidasi etanol. Model katalis PdCu (111) didesain menggunakan ekspansi 3'3 sel satuan periodik dengan tiga lapisan Pd. Variasi PdCu(111)-A dimodelkan permukaan tertutup atom Cu, sedangkan PdCu(111)-B dimodelkan atom Cu dan Pd terdistribusi merata pada dua lapisan pertama. Perhitungan dilakukan dengan fungsi korelasi-pertukaran PBEsol dalam basis *ultrasoft* (US) *pseudopotentials* (PPs). Keadaan transisi ditentukan menggunakan metode *nudged elastic band* (NEB). Hasil penelitian menunjukkan posisi interaksi yang stabil dari semua spesi reaksi oksidasi etanol pada permukaan PdCu (111), yaitu spesi CH*, OH* dan CO* berada di posisi *hollow*, CH₃CO* dan CH₃COOH* di posisi *on-top*. Untuk CHCO* berada *fcc* (atom C dari CH*), *bridge* (atom C dari CO*), dan *on-top* (atom O) dari permukaan katalis. Sifat elektronik PdCu(111)-A lebih baik daripada PdCu(111)-B, tetapi menunjukkan aktivitas katalitik yang kurang baik. Analisis kerapatan muatan menunjukkan atom Cu bermuatan parsial negatif dan atom Pd bermuatan parsial positif. Katalis PdCu (111) dengan Cu dan Pd terdistribusi merata pada dua lapisan pertama menunjukkan aktivitas katalitik yang baik untuk oksidasi etanol dengan energi aktivasi pembentukan asam asetat terbesar (0,61 eV) dan pemutusan ikatan C–C terkecil (0,96 eV).

Kata kunci: adsorpsi, energi, katalis, PdCu (111)

***THE COMPUTATIONAL STUDY OF ETHANOL OXIDATION REACTION
MECHANISM ON PdCu (111) CATALYST USING DENSITY FUNCTIONAL
THEORY APPROXIMATION***

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ABSTRACT

The mechanism of ethanol oxidation reaction on the PdCu (111) catalyst had performed using density functional theory approximation. The purpose of this study was to determine the stable interaction position of ethanol oxidation reaction species on the PdCu (111) surface, the effect of variation doping Cu on electronic properties of PdCu catalyst (111), and the distribution effect co-catalyst (Cu) on the PdCu catalyst (111) for the acetic acid formation mechanism and the breaking C–C bonds in the ethanol oxidation reaction. Models of PdCu (111) designed by a periodic 3'3unit cell with three Pd monolayers. In the PdCu(111)-A, the surface was covered by Cu atoms, while PdCu(111)-B models Pd and Cu atoms had uniformly distributed in the uppermost two layers. All calculations performed by the PBEsol correlation-exchange function based on ultrasoft (US) pseudopotentials (PPs). The transition state had determined using the nudged elastic band (NEB) method. The results showed a stable interaction position of ethanol oxidation reaction species on the surface of PdCu (111), namely CH*, OH*, and CO* species were on the hollow position, CH₃CO* and CH₃COOH* on-top position. For CHCO* on fcc (C atom of CH*), bridge (C atom of CO*), and top (O atom) of the catalyst surface. The electronic properties of PdCu(111)-A is better than PdCu(111)-B, but has less good catalytic activity. Analysis of charge density showed Cu atoms partially negatively charged and the Pd atoms partially positively charged. PdCu catalyst (111) with Cu and Pd evenly distributed in the uppermost two layers showed good catalytic activity for ethanol oxidation with the highest activation energy of acetic acid formation (0.61 eV) and the breaking of the lowest C–C bond (0.96 eV).

Keywords: adsorption, catalyst, energy, PdCu (111)