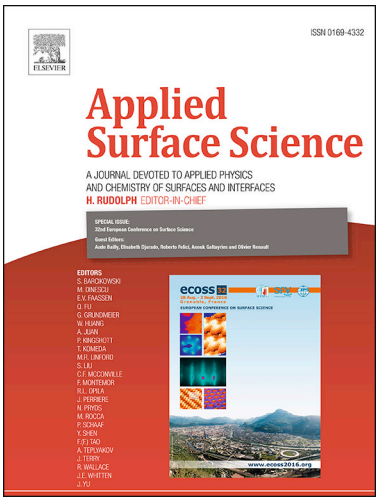
Accepted Manuscript



Full Length Article

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PII: S0169-4332(18)30141-7
DOI: https://doi.org/10.1016/j.apsusc.2018.01.129
Reference: APSUSC 38268

To appear in: *Applied Surface Science*

Received Date: 31 August 2017
Revised Date: 6 January 2018
Accepted Date: 13 January 2018

Please cite this article as: O. Dmitriyeva, S.C. Hamm, D.L. Knies, R. Cantwell, M. McConnell, The impact of surface composition on Tafel kinetics leading to enhanced electrochemical insertion of hydrogen in palladium, *Applied Surface Science* (2018), doi: https://doi.org/10.1016/j.apsusc.2018.01.129

The impact of surface composition on Tafel kinetics leading to enhanced electrochemical insertion of hydrogen in palladium

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Abstract

Our previous work experimentally demonstrated the enhancement of electrochemical hydrogen insertion into palladium by modifying the chemical composition of the cathode surface with Pb, Pt and Bi, referred to as surface promoters. The experiment demonstrated that an optimal combination of the surface promoters led to an increase in hydrogen fugacity of more than three orders of magnitude, while maintaining the same current density. This manuscript discusses the application of Density Functional Theory (DFT) to elucidate the thermodynamics and kinetics of observed enhancement of electrochemical hydrogen insertion into palladium. We present theoretical simulations that: (1) establish the elevation of hydrogen's chemical potential on Pb and Bi surfaces to enhance hydrogen insertion, (2) confirm the increase of a Tafel activation barrier that results in a decrease of the reaction rate at the given hydrogen overpotential, and (3) explain why the surface promoter's coverage needs to be non-uniform, namely to allow hydrogen insertion into palladium bulk while simultaneously locking hydrogen below the surface (the corking effect). The discussed DFT-based method can be used for efficient scanning of different material configurations to design a highly effective hydrogen storage system.

Keywords

DFT, NEB, hydrogen storage, HER, surface promoter, Tafel poison