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SECM Screening of the Catalytic Activities of AuPd Bimetallic Patterns and their Heterogeneous Electron Transfer Rates Calculation

Mengwei Ye[1], Yi Li[1], Juan-Wu[1], Jie Zhang[2], Jing Tang[1]
[1]*Fuzhou University, Fuzhou, Fujian Province, China*
[2]*Xiamen University, Xiamen, Fujian Province, China*

Scanning electrochemical microscopy (SECM) is a good technique for evaluating the local electrocatalytic activity of different catalysts such as some multimetal catalysts for oxygen reduction, hydrogen oxidation, and hydrogen peroxide reduction [1]. Moreover, SECM is also a powerful electrochemical tool for investigating heterogeneous electron-transfer (HET) reactions at interfaces between an electrode and an electrolyte[2]. Now we use SECM to batch screening the catalytic activities of different AuPd catalysts for H_2O_2 and FcMeOH reduction reaction and also confirm the results of SECM by calculated HET rates for the respective AuPd electrodes.

The sample of electrocatalyst arrays with various AuPd compositions was prepared by an electrochemical wet-stamping technique. Micropatterned high-strength agarose containing different solutions of chloroauric acid and chloropalladic acid has been used to electrodeposit and generate patterns of AuPd nanoparticle arrays on ITO. The electrochemical activities of the AuPd alloys for H_2O_2 and FcMeOH reduction reactions have been investigated in both redox-competition and feedback modes by means of SECM. Then we used the SECM approach curves to study the kinetic of heterogeneous reactions on AuPd array substrates.

In the following images, Au_xPd_y represents a simplified form of the AuPd bimetal nanoparticles that were deposited from electrolytes containing mixed salts that C_{HAuCl4}:$C_{H2PdCl4}$=x:y. Fig. 1(a) shows the RC-SECM mode images of H_2O_2 reduction on a series of AuPd electrocatalyst arrays in H_2O_2 solution. The tip and the substrate were both biased at -0.2 V, at which H_2O_2 reduction can take place. In this image, the Au_3Pd_1 band showed the lowest current among all of the bands which demonstrate the Au_3Pd_1 band have the highest activity for the H_2O_2 reduction. Fig. 1(b) presents an SECM feedback mode image of FcMeOH reduction and the Au_3Pd_1 band presents the highest current among all the bands in the respective arrays. Contrary to the RC-mode, the higher current represents a better catalytic activity in feedback mode, so the Au_3Pd_1 band showed the highest activity for FcMeOH reduction. Fig. 1(c) is the comparison of the experimental (solid lines) and theoretical (symbols) SECM approach curves on AuPd arrays with different compositions in FcMeOH solution. The finite element modeling method was used to simulate the tip current response as a function of the standard HET rate constant (k_0) through COMSOL Multiphysics 4.3b, and the simulated probe approach curve with different values of k_0 could be readily fitted to the experimental current curves. Because a higher value of k_0 indicates better electrochemical activity, the Au_3Pd_1 showed the highest electrocatalyst activitie, this result is in accordance with the SECM image.

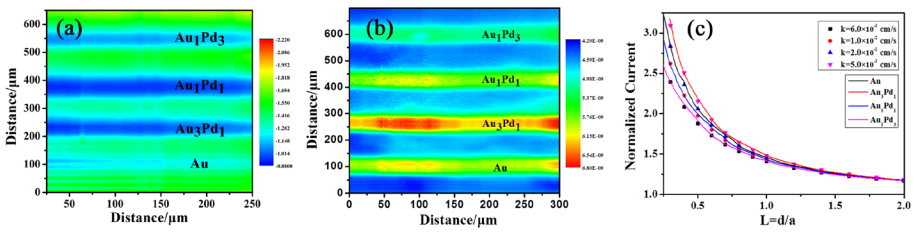

Figure 1 (a) The SECM RC-mode image of AuPd band arrays in H_2O_2 solution. (b) The SECM feedback mode image of the AuPd band arrays in FcMeOH solution. (c) Comparison of the experimental (solid lines) and theoretical (symbols) SECM approach curves on AuPd arrays.

References

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