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Dealloyed platinum-copper with isolated Pt atom surface: facile synthesis and promoted dehydrogenation pathway of formic acid electro-oxidation

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Abstract

In order to obtain the catalyst with Pt isolated-atom surface and high performance towards formic acid electro-oxidation (FAEO), cyclic potential dealloying method was employed for preparing dealloyed PtCu/C (D-PtCu/C) catalysts, and the effect of dealloying parameters on the activity of D-PtCu/C catalysts also were investigated. XRD results show that both of PtCu and D-PtCu catalysts are face-centered cubic (fcc) structure. TEM, XPS and ICP results demonstrate the dissolution of Cu atoms and the formation of core-shell structure. And HRTEM results further exhibit the etched surface of D-PtCu/C catalyst which should be made up of isolated atom and atomic clusters of Pt. By electro-chemical analysis, the performance towards FAEO is extremely enhanced. The onset potential of FAEO on D-PtCu/C is at -0.1 V (vs. SCE) and only one peak is confirmed at 0.5 V which negatively shifts about 0.19 V compared with the peak of commercial Pt/C (at 0.69 V). The mass activity of optimized D-PtCu/C reaches up to 1253.89 mA mg_{Pt}⁻¹ at 0.5 V, which is 6.20 times as high as that of Pt/C (202.40 mA mg_{Pt}⁻¹). And the specific activity (2.36 mA cm⁻²) is 8.43 times of Pt/C (0.28 mA cm⁻²). The increased activity should be ascribed to the electronic effect and ensemble effect, which reduce the chemisorption of CO and promote the dehydrogenation process.

Keywords: Formic acid, Electro-oxidation, Platinum-copper, Isolated atom, Dehydrogenation

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