Preparation of N-doped Carbon/Metal Phosphides as Promising Trifunctional Electrocatalysts Toward the OER, ORR and HER

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INTRODUCTION

Sustainable energy storage and conversion technologies, such as electrochemical water splitting and fuel cells, attracts increasing attention as alternative processes to advance toward a global decarbonation. However, the high cost, scarcity, and poor stability of the most active electrocatalysts, mainly based on noble metal (Pt, RuO_2, IrO_2,…), difficult severely their large-scale production and use.[1] In this regard, the development of earth-abundant electrocatalysts, with high activity for the different processes, is needed.[2]

EXPERIMENTAL/THEORETICAL STUDY

We propose the use of imidazole-containing metal etidronates, a type of coordination polymers, as precursors of transition metal phosphides (TMPs) under pyrolytic treatments. The presence of imidazole may lead to the formation of heteroatom-doped carbon composites, which have been found to be essential to enhance the electrochemical properties.[3] In this work, the electrocatalytic performances of the resulting N-doped carbon/TMPs have been investigated toward the oxygen evolution reaction (OER), oxygen reduction reaction (ORR), and hydrogen evolution reaction (HER).

RESULTS AND DISCUSSION

Several imidazole-containing metal etidronates, **MLIm-n** (M^{2+}= Fe, Co, Ni and solid solutions; L= ETID; Im= Imidazole; n= [0, 3]), were prepared using different synthetic procedures. Thermal reduction of **MLIm-n** in 5%H_2–Ar at different temperatures resulted on core–shell N-doped carbon/TMPs with variable content of MP and M_xP as crystalline phases. Their electrocatalytic activities have been widely studied by cyclic and linear sweep voltammetry, impedance spectroscopy, transmission electron microscopy, and XPS analysis. Preliminary results reveal that factors such as the presence of the imidazole and the metal coordination environment in the precursor samples determine the final composition and electrochemical properties of the resulting pyrolyzed derivatives. So, the Co^{2+}- derivative, **CoLIm-0@800**, with a composition CoP/Co_2P= 80/20 wt. %, exhibited the best electrocatalytic properties toward OER/ORR/HER as well as good capabilities as anode for overall water splitting in comparison to the expensive reference RuO_2 electrocatalyst.[4]

CONCLUSION

N-doped carbon/TMPs (M^{2+}= Fe, Co, and Ni and solid solutions) have been prepared as promising electrocatalysts from imidazole-containing metal etidronates by pyrolytic treatment. In general, those TMPs obtained from **CoLIm-n** exhibited better performances due to the formation of more active crystalline phases, and the presence of imidazole has been demonstrated to be detrimental for enhancing the electrocatalytic activity.

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