



Synthesis and function of nanosheets doped with multi-elements in equal molarity

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Abstract

Water pollution by dyes is one of the biggest environmental problems. Adsorption technology has been widely used in wastewater treatment. Here, high-entropy concept is used to design surface defective hydroxides realizing the rapid removal of dyes from water. Multi-element hydroxides containing five, i.e., Mn, Fe, Co, Ni, and Zn, transition metal cations are successfully synthesized through a bottom-up polyol process (Fig.1). Along with the increase in compositional complexity, the thickness of the nanosheets in the hydroxides decreases, while the degree of surface defects increases. These surface defects are proved to be the active sites for anionic dyes adsorption. These multi-element hydroxide nanosheets are also promising electrocatalysts for oxygen evolution reaction.

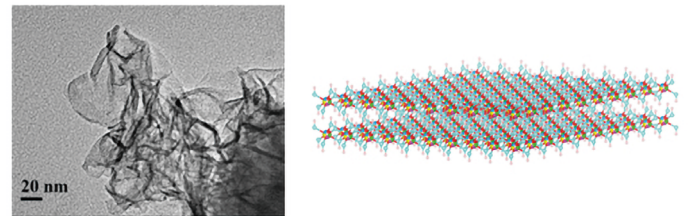


Fig.1 multi-element hydroxides nanosheet

Background & Results

High-entropy alloys describe a class of single-phase solid solution containing five or more elements in relatively high concentrations (5-35 at.%). This high-entropy concept expands to the whole materials field since 2015. The concept of high-entropy design opens a door to develop novel materials with vast compositional space. The multi-element feature of high-entropy materials leads to several advantages, which are the so-called four core effects, namely the high-entropy effect in thermodynamics, the lattice distortion effect in crystallography, the sluggish diffusion effect in kinetics, and the "cocktail" effect in material performance. The key issue in synthesizing high-entropy materials is how to mix homogeneously the multiple elements. High-energy ball milling followed by high-temperature heat treatment are commonly used approaches. However, it is very difficult to control the formation of low-dimensional nanomaterials, such as two-dimensional (2D) high-entropy materials.

Here, we report a simple polyol process for the synthesis of 2D layered multi-element (Mn, Fe, Co, Ni, Zn) hydroxides (MEHs), involving the hydrolysis and inorganic polymerization of metal-containing species in ethylene glycol. Hydrolysis and reduction of metal cations are two competitive reactions in our polyol process. By optimizing the synthesis conditions, the MEHs nanosheets can be successfully obtained after solvothermal treatment at $\sim 200^\circ\text{C}$ (Fig.2). The surface of these MEHs nanosheets are highly defective. These MEHs nanosheets can adsorb anionic dyes (e.g., Congo red) from wastewater, possessing adsorption kinetics that are two orders of magnitude higher than that of commercial activated carbon (Fig.3). These defects can also be active sites for catalyzing oxygen evolution reactions, giving an overpotential of 298 mV at a current density of 10 mA cm^{-2} in 1.0 M KOH electrolyte.

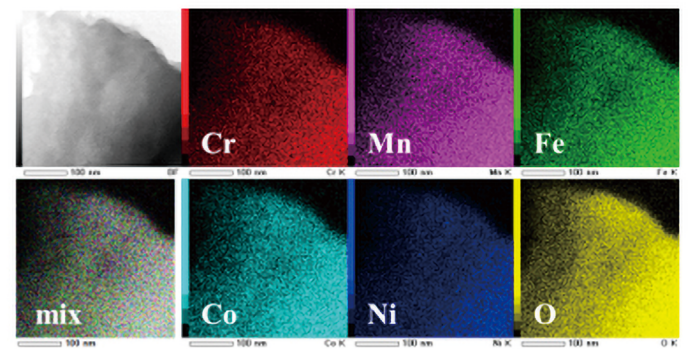


Fig.2 elemental maps of multi-element hydroxides nanosheet

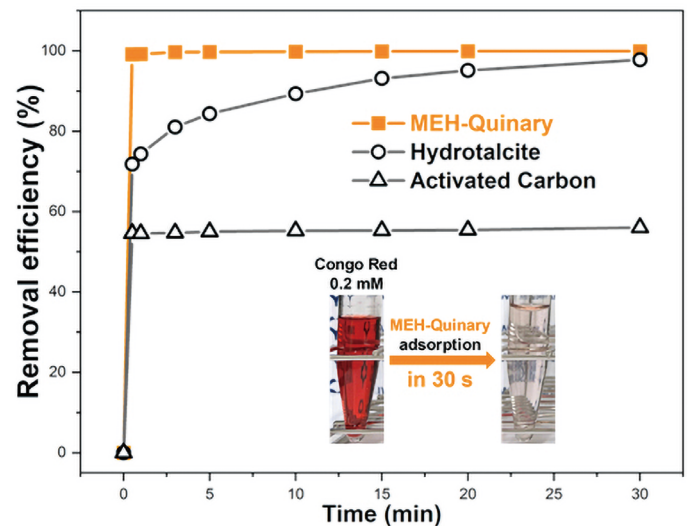


Fig.3 Anionic dye removal functionality of multi-element hydroxide nanosheets

Significance of the research and Future perspective

The combination of unique surface features of 2D nanomaterials and advantages of high-entropy materials brings great possibilities to discover novel multifunctional materials. Our observations reveal that utilizing configurational entropy to modulate the electronic structures and defects would be an effective approach to obtain novel materials with charming applications.

Patent

Treatise

URL

Keyword

Li, Fei; Maruyama, Jun; Abe, Hiroya et al. Bottom-up synthesis of 2D layered high-entropy transition metal hydroxides. *Nanoscale Advances*. 2022, 4, 2468-2489. doi: 10.1039/d1na00871d
 Li, Fei; Maruyama, Jun; Abe, Hiroya et al. Defective multi-element hydroxides nanosheets for rapid removal of anionic organic dyes from water and oxygen evolution reaction. *J. Hazard. Mater.* 2023, 447, 130803. doi: 10.1016/j.jhazmat.2023.130803

multi-element, compound, environment