Topochemical synthesis and anion exchange of iron (II, III) layered hydroxides toward oxygen evolution reaction (OER)

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Layered double hydroxides (LDHs), especially Ni-Fe, Co-Fe LDHs, have displayed significant oxygen evolution reaction (OER) electrocatalytic activity in alkaline conditions.[1,2] In these cases, Fe plays an essential role in enhancing the electrocatalytic performance. Green rust (GR), a Fe^{2+}-Fe^{3+} LDH, is considered a promising single-iron electrocatalyst. However, GR synthesized by conventional methods is generally of low crystallinity. It is required to develop an alternative and reliable method to synthesize GR with a high quality.

In the current work, an innovative topochemical oxidative intercalation method was explored for preparing micrometer-sized hexagonal platelets of GR under rigorous exclusion of oxygen.[3] Highly crystalline hexagonal platelets of brucite-like ferrous hydroxide ($Fe(OH)_2$) were first prepared by homogeneous precipitation. After treatment with stoichiometric iodine (I_2) for 72 h, I^--intercalated GR was formed via oxidative intercalation. The interlayer spacing was correspondingly expanded from 4.6 Å to 8.2 Å. Using dehydrated dimethylformamide (DMF) as a solvent, I^--intercalated GR was anion exchanged into dodecyl sulfate (DS^-) form with an interlayer spacing of 25.6 Å. During the topochemical transformation and anion exchange processes, the high quality of precursor crystals was perfectly inherited. As synthesized GR can be further exfoliated into monolayer nanosheets and used as electrocatalysts in water splitting, which is of great importance to understand the reaction mechanism since Fe is widely recognized as the active sites for efficient OER catalysis.

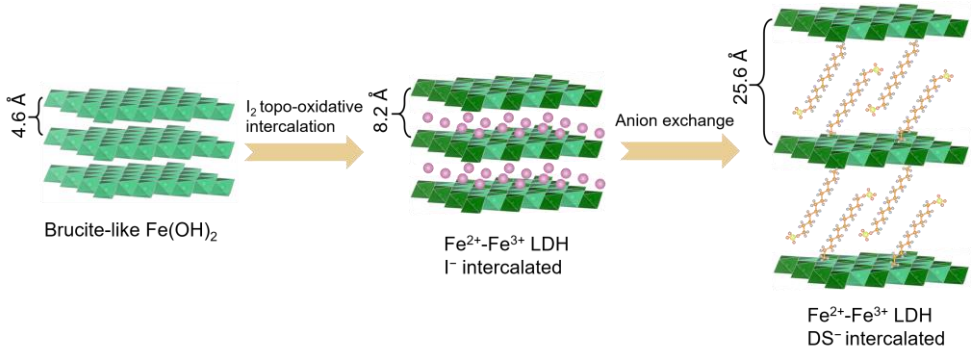

Fig. 1 Schematic illustration of topochemical synthesis and anion exchange of iron layered hydroxides.

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