Processing of Layered Double Hydroxides for Energy Storage Applications

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Layered double hydroxides (LDHs) are a class of anionic clays consisting of positive charged brucite-like layers spaced by water molecules and counterbalancing anions. In particular, NiAl-, NiFe- and NiCo-LDHs have drawn attention for energy storage and conversion applications[1] because of their electrocatalytic properties for oxygen evolution reaction (OER)[2] and hydrogen evolution reaction (HER)[2], as well as for pseudocapacitive behaviour[3][4]. In contrast with other layered materials, including graphene and transition metal dichalcogenides (TMDs), LDH layers are held in place by electrostatic forces and a dense network of hydrogen bonds[5]. For these reasons, a careful choice of solvent is pivotal for an efficient exfoliation of the LDHs. One of the most efficient solvent in term of exfoliation yield is formamide.[6][7] However, formamide has toxicity issues and high boiling point, making the search for other solvents needed for practical LDH processing[8]. Although, dispersions of LDH nanoplates in water or alcohols are stable, they result in an ineffective material exfoliation. Therefore, surfactants are typically required to assist the LDH exfoliation into monolayer forms[6]. In our work, we will rationalize the properties of optimal solvent:surfactant mixtures in term of surface energy and dielectric constant, which are primary parameters determining the LDH exfoliation. Moreover, we will take into account the viscosity, the boiling point and the toxicity of the solvent:surfactant in order to enable an effective and unhazardous LDH handling[9]. We will show that NiFe-LDH hexagonal nanoplates, synthesized with Jaśkaniec's method[10], can be treated with aqueous acetate solution to obtain a stable dispersion in ethanol. Transmission electron microscopy analysis reveals a partial exfoliation of the LDH nanoplatelets, whilst the ultraviolet-visible light spectroscopy measurements suggest the presence of the acetate ion bonded the LDH surface, enhancing the LDH/ethanol affinity. The as-produced LDH dispersion can be deposited on proper substrates (*e.g.*, graphite paper, nickel foam) to form catalytic films for electrochemical applications. Finally, LDHs can be combined with graphene materials, and TMDs creating superlattice materials with superior electrochemical properties[1].

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