

# Covalent functionalization of non-aluminium layered double hydroxide with tripodal ligands.

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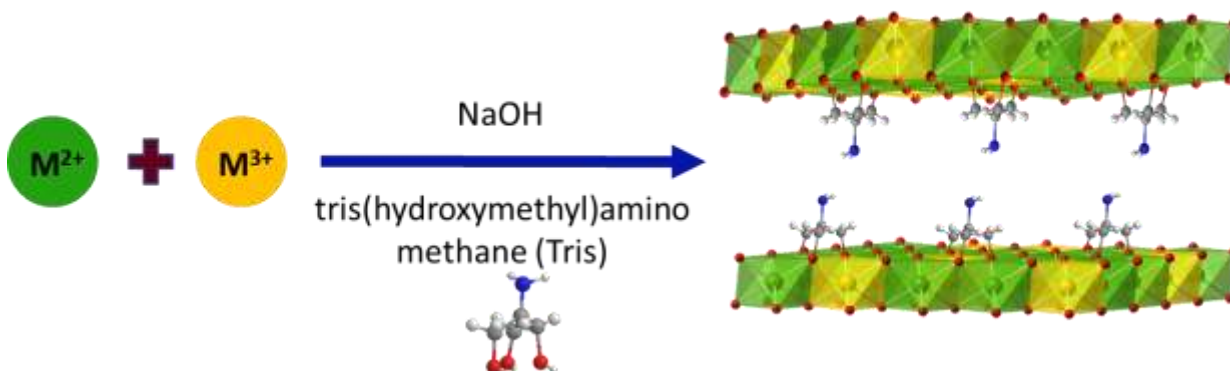
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Layered double hydroxides (LDHs) are a class of cationic layers with exchangeable anions in the interlayer space that exhibit a hydrocalcite-like structure. The high tunability regarding metallic composition, stoichiometry, and the interlayer anion gives LDHs a wide versatility; resulting in different applications such as catalysis, sensing, magnetism or energy storage, to name a few.[1] Furthermore, the anionic behaviour of these 2D materials allows the design of several hybrid heterostructures through electrostatic interactions.[2,3] In this field, a further step would be the covalent functionalization of LDH, allowing these attractive materials to be endowed with new properties and creating more complex hybrid heterostructures. With this goal in mind, the covalent functionalization of LDHs is being investigated by our group, indeed we developed a pH-reversible covalent functionalization with aminopropyl triethoxysilanes (APTES) of the NiFe-LDH.[4] However, in view of the poor stability at basic pH (typical pH for electrochemical applications) of this covalent bond, we have performed a new stable functionalization at basic pH with tris(hydroxymethyl) aminomethane (Figure 1) of LDHs with different compositions (NiFe, CoFe and NiV-LDHs), using an modified hydrothermal method from Kuroda et al.,[5]. This new family of LDHs has been studied by means of X-ray diffraction, infrared spectroscopy, thermogravimetric analysis coupled with mass spectrometry, X-ray photoelectron spectroscopy, transmission electron microscopy and energy-dispersive X-ray spectroscopy, among others. In addition, its magnetic and electrochemical behaviour was explored. Reactivity studies have been carried out on the terminal amino group, as well as protonation and deprotonation, to modify the behaviour in terms of the polarity of these materials. This new level of manipulation in LDHs can be used to design novel heterostructures on-demand and improve the exfoliation of these appealing materials.

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**Figure 1:** Synthesis of covalently functionalized LDHs with tripodal ligands.