



# Effect of structure and bonding on the interfacial properties and the reactivity of layered double hydroxides and Zn hydroxide salts

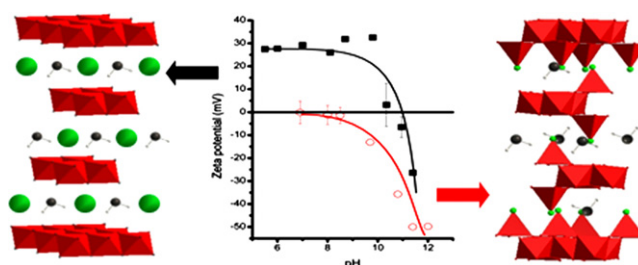
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## HIGHLIGHTS

- Correlation between structure/binding and interfacial properties/ reactivity is described.
- LDHs and LHSs exhibited anion and ligand exchange reactions, respectively.
- Layers structure and chemical interactions between aromatic anions determined particle charge.
- Aromatic anions intercalation induced a hydrophobic character increase.
- The interlayer anion ionizable group influenced binding strength to LDH/ LHS layers.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 30 October 2012

Received in revised form

27 November 2012

Accepted 3 December 2012

Available online 11 December 2012

### Keywords:

Intercalation compounds

Zeta potential

Hydrophobic/hydrophilic character

Anion/ligand exchange

## ABSTRACT

Both layered double hydroxides (LDHs) and Zn layered hydroxide salts (LHSs) are intercalation compounds with potential applications as pollutant sorbents and antimicrobial, drug, and pesticide carriers, among others. Their reactivity and interfacial properties are essential to these applications and dependent on the layer structure and interlayer anion nature. In this work, the structure and bonding of Zn–Cr LDHs and Zn–LHSs intercalated with chloride, 4-chlorobenzoate, and 4-chlorobenzenesulfonate were analyzed using X-ray diffraction and infrared spectroscopy. Afterwards, their effect on interfacial properties and reactivity was assessed by zeta potential measurements, contact angle determinations, and hydroxide anions uptake. Due to their different structure, LDHs presented electrostatic interactions between their layers and the intercalated anions, while LHSs portrayed coordinated binding. The latter exhibited neutrally or negatively charged particles and ligand exchange reactions, while LDHs presented positively charged particles and anion exchange reactions. The interlayer anion nature also modulated the behavior of these solids: the introduction of aromatic anions strongly modified the hydrophilic/hydrophobic character of the surface, while their ionizable group determined the anion binding strength and consequently the anion/ligand exchange equilibrium of LDHs and LHSs.

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## 1. Introduction

Layered double hydroxides (LDHs) and Zn-layered hydroxide salts (LHSs) are intercalation compounds with different

bi-dimensional structures derived from that of brucite ( $\text{Mg}(\text{OH})_2$ ). The structure of this hydroxide can be described as a compact packing of hydroxide anions, where divalent ions are placed in half of the octahedral sites, resulting in layers of edge-sharing octahedra. LDHs structure is derived by isomorphic substitution of divalent by trivalent ions in octahedral sites [1,2]. LDH layers present a positive charge excess leading to intercalation of anions that share the interlayer domain with solvent molecules (usually water). On the

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