

Layered Double Hydroxides

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The impact of layered double hydroxides (LDHs) within the multidisciplinary fields of materials sciences, physics, chemistry, and biology is rapidly growing, given their easiness of synthesis, flexibility in composition, tunable biocompatibility and morphology. LDHs constitute a versatile platform for the realization of new classes of functional systems, showing unique enhanced surface effects and unprecedented properties for application in very different fields, namely, surface chemistry and catalysis, storage and triggered release of functional anions, flame retardants, drug delivery and nanomedicine, remediation, energy storage and conversion. These systems can be synthesized as self-assembled hierarchical nanosheet thin films by means of low temperature solution-based approaches, which are accessible by many laboratories and have the advantages of low cost, mild conditions, and environmental friendliness. In addition, the possibility of LDHs to be exfoliated into 2D nanosheets has been demonstrated to further improve their performance in many applications, as well as to be an attractive route to achieve building blocks for fabricating a wide plethora of hybrid functional architectures. LDHs are therefore a playground for exciting new research covering all of the most intriguing features of 2D materials and more. This Special Issue on “Layered Double Hydroxides” gathers a multidisciplinary collection of original contributions and review articles from authors with diverse scientific backgrounds and who employ LDHs for very different applications, permitting the demonstration of their versatility. Along with LDH-focused papers, this Special Issue also includes some research in which materials different to LDHs resulted in a convenient choice for selected purposes.

A study of particular interest is the report from Teixeira et al. [1], where the extreme flexibility of LDH matrices in changing both interlayer and metal components to tune their physicochemical properties is explored. The memory effect in thermally treated LDHs, i.e., the restoration of their lamellar structure by rehydration in aqueous solutions containing anions, is leveraged for the synthesis of rare earth Eu^{3+} doped luminescent LDHs intercalated with 1,3,5-benzenetricarboxylate anion. The latter acts as an anionic photosensitizer for Eu^{3+} ions, increasing the total observable luminescence by means of the so-called antenna effect. Such a combination of the two effects also provided a useful tool to monitor the rehydration process of the calcined LDHs.

This Special Issue has received many contributions from the field of pollutant remediation, highlighting the key role of LDH-based compounds for this particular application. Specifically, it is important to consider LDHs as outstanding candidates for selective adsorption of anionic contaminants, taking advantage of anion exchange with the interlayer anions, or anion trapping in the interlayer during rehydration of mixed metal oxides from calcined LDHs and subsequent reconstruction of the lamellar structure via the “memory effect”. In this context, Dore et al. [2] showed a clear example of the usefulness of calcined LDHs, namely, mixed MgAlFe oxides and mixed ZnAl oxides from hydrotalcite-like and zaccagnaite-like compounds, respectively, to remove Sb(V) , in the Sb(OH)_6^- form, from aqueous solution. The authors also demonstrated the feasibility of LDH-based removal of Sb(OH)_6^- from the slag drainage in an abandoned mine in Sardinia, Italy. Another excellent application

in the field of water decontamination comes from Golban and coworkers [3], who proposed a new and convenient method to synthesize Mg_4Fe -LDHs from iron-containing acidic residual solution of the hot-dip galvanizing process, obtaining a material suitable for the effective decontamination of MoO_4^{2-} from aqueous solutions. Similarly, the selective recovery of Cu(II) from metal mixtures was conveniently achieved by Yang et al. [4] with calcium alginate beads, which are well-known green sorbents for the biosorption of heavy metals. The fabricated alginate beads showed also excellent retainment of their properties after five cycles of sorption–desorption procedures.

Along with heavy metal pollution, antibiotics increasingly pose a serious concern for environmental water. Panplado et al. [5] demonstrated a simple strategy to very rapidly remove tetracycline (TC) antibiotic molecules from contaminated water. They propose an *in-situ* adsorption method which involves the utilization of Mg^{2+} and Al^{3+} containing LDH precursors to promote the precipitation of mixed metal hydroxides (MMHs), which act as fast sorbents for capturing TC from aqueous solution. The strong interactions between the charged MMH surface and the TC molecules, consisting of electrostatic attraction and hydrogen bonding, were leveraged to achieve instantaneous adsorption, which is superior to the use of LDH as sorbent in a conventional route.

This Special Issue also contains original contributions in the field of biology-related applications. Chang et al. [6] demonstrated the ability of Mg_2Fe -LDHs to significantly enhance the production of surfactin in bacterial cells of a *Bacillus subtilis* ATC 21,322 culture. Surfactin is a cyclic lipopeptide of seven amino acids and acts as an excellent biosurfactant. However, in order to obtain sufficient levels to allow for its commercial use, its production needs to be enhanced. Another interesting contribution is provided by Sun and coworkers [7], who leveraged solid lipid nanoparticles for efficient resveratrol loading, with the aim to obtain a substantial improvement of the mitochondrial function in mice, in comparison with control resveratrol supplementation in the absence of nanoparticle loading.

Finally, the role of LDHs as catalysts in relevant organic chemistry reactions and their emerging application in cellular biology were extensively reviewed by Arrabito et al. [8], highlighting the conspicuous studies focusing on the synthesis, characterization, and applications of LDH-based systems. In a second review article [9], the same authors reviewed the role of LDHs in the scenario of bioinspired nanomaterials research and applications thereof. This work provides a possible link between the role of LDHs in the origin of life and the formation of pre-biotic molecules, to inspire the fabrication of artificial LDH-based compartments that mimic prebiotic assemblies. The design of LDH-based systems for life-like and life-inspired devices was also reviewed.

In summary, the present Special Issue on “Layered Double Hydroxides” can be considered as a status report that gathers and reviews different contributions summarizing the progress of many different LDH-related research and applications in the past several years.

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