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## Layered double hydroxide coatings on magnesium alloys: A review

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## ABSTRACT

Layered double hydroxides (LDHs) as a class of anionic clays have extensive applications due to their unique structures. Nowadays, the emphasis is laid on the development of LDH coatings for corrosion resistance and medical applications. Thus, this review highlights synthetic methods of LDH coatings and LDH-based composite coatings on magnesium alloys. Special attention is focused on self-healing, biocompatible and self-cleaning LDH-based composite coatings on magnesium alloys.

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## 1. Introduction

Magnesium (Mg) alloys as the lightest constructional materials, have been extensively applied in a wide range of areas, such as lightweight structure materials and biomedical materials [1–3]. In particular, Mg and its alloys provide a great potential for the applications in computers, communications and consumer electronics (3C) products, and aerospace and automotive industries [4–6]. In a typical car, an overall weight saving of 10% yields fuel savings on an order of 20%–30%, without marked changes in design [7].

In addition, ~60% of Mg in the human body is stored in bone [8,9]. Possessing mechanical properties similar to those of natural bone, as well as greater mechanical behavior than polymer biomaterials, Mg alloys are employed as biodegradable cardiovascular stents and orthopedic implants [10–13].

Nevertheless, Mg alloys are active due to their low standard potential, which leads to poor corrosion resistance and restricts their industrial applications on a larger scale [14–17]. Then along with the fast degradation of Mg alloys, increasing pH and degradation in mechanical strength may result in local inflammation and destruction of surrounding tissue, limiting the medical applications

to some extent [18,19]. Up to now, various surface modifications have been adopted to enhance corrosion resistance of Mg alloys, including chemical conversion coatings [20–23], polymer coatings [24–26], micro-arc oxidation (MAO) [27–30], and saline coatings [31–35] as well as their composite coatings [36–40]. Among the different films or coatings, layered double hydroxides (LDHs) have been widely investigated due to their unique structures as well as corrosion protection on metals.

LDHs, also known as hydrotalcite-like (HT) compounds, are a class of anionic clays with a highly tunable brucite structure [41–45]. Natural hydrotalcite was first discovered in Sweden around 1842 and synthesized by Feitknecht 100 years later [46]. As shown in Fig. 1, a general molecular formula of LDHs is  $[M^{2+}_{1-x}M^{3+}_x(OH)_2]^{x+}(A^{m-})_{x/m}\cdot nH_2O$ , where  $M^{2+}$  and  $M^{3+}$  represent divalent metallic cations (e.g.  $Mg^{2+}$ ,  $Ca^{2+}$ ,  $Cu^{2+}$ ,  $Mn^{2+}$ ,  $Zn^{2+}$ ) and trivalent metallic cations (e.g.  $Al^{3+}$ ,  $Cr^{3+}$ ,  $Fe^{3+}$ ,  $Co^{3+}$ ), respectively [47,48].  $A^{m-}$  is the interlayer anion (e.g.  $NO_3^-$ ,  $Cl^-$ ,  $CO_3^{2-}$ ,  $PO_4^{3-}$ ),  $m$  is the charge of the interlayer anion; whereas, water molecules are generally embedded between the hydroxides layers during the synthesis process.  $X$  indicates the molar ratio of  $M^{3+}/(M^{2+}+M^{3+})$ , ranging from 0.20 to 0.33 [49–51]. Basically, size, charge and ratio of metal cations, charge and orientation of anions as well as relative amount of crystal water determine crystal structure parameters of LDHs, bond strength and anion exchange capacity [52–54].

So far, LDH materials have been largely used as supercapacitors [55], adsorbents [56], anion exchangers [57], catalysts [58],

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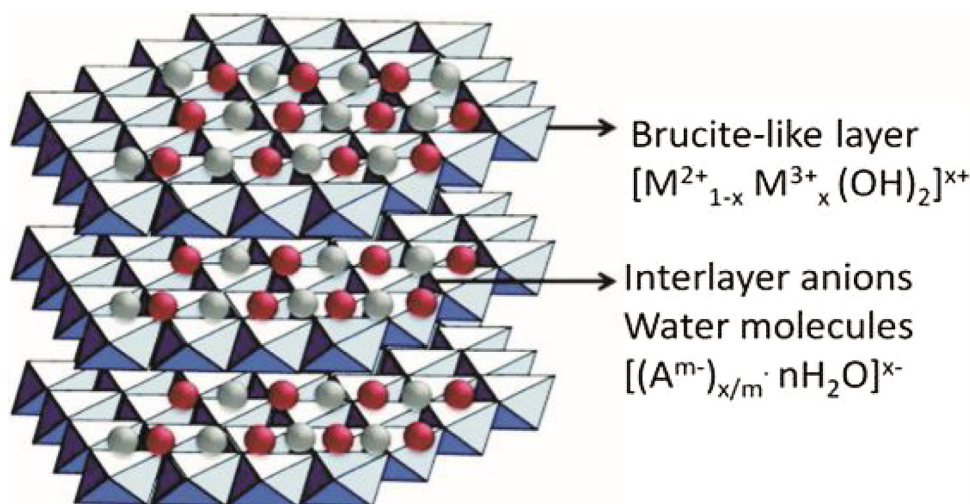


Fig. 1. Schematic illustration of LDH structure and chemical components [48].

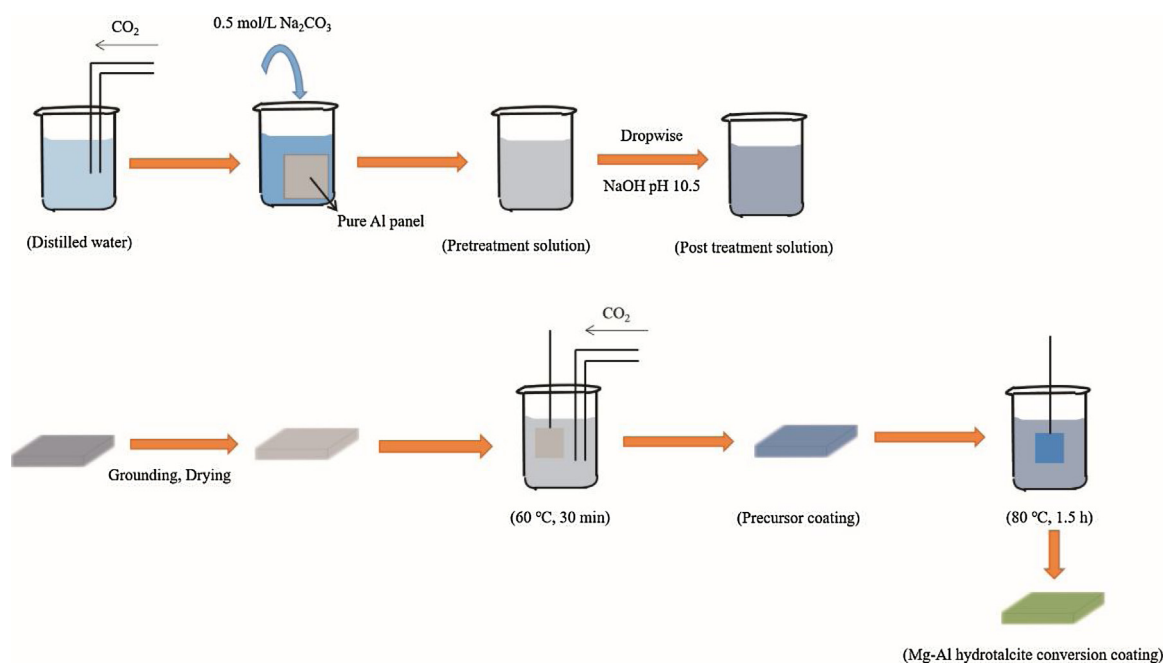


Fig. 2. Solutions used in the two-step in situ method.

polymer additives [59], drug delivery [60] and corrosion protection agents [61] due to their diversity in composition and structures. Substantial studies have been focused on the fabrication of LDH coatings on aluminum and zinc alloys by means of in situ methods [54,62,63]. To date, different LDH coatings have been developed on Mg alloys.

The development of LDHs can trace back to recent decades. Rives et al. [64] reviewed the systems of LDH intercalated with metal coordination compounds and oxometalates in 1999. Yang et al. [65] summarized the utilization of LDH-based materials as potential adsorbents and photocatalysts for decontamination of dyes wastewater due to their high anion exchange capacity. Basu et al. [46] discussed the synthesis, structure, properties and applications of elastomer/(LDH) composites. Recently, more and more attention has been paid to the applications of LDH coatings, especially on the surface of Mg alloys. This review aims to summarize the synthetic methods of single LDH coatings and their composite coatings on Mg alloys, and to take insight into the corrosion performance of LDH coatings.

## 2. Single LDH coatings

### 2.1. In situ growth methods

In situ growth techniques have been regarded as a promising alternative method due to their simplicity and versatility in practice and strong adhesion of coatings to metal surface by chemical bonds. And size of LDH particles and density of the coatings can be adjusted by changing processing time and pH during the reactions. It is noteworthy that Buchheit et al. developed a series of LDH coatings and made great contributions on the development of in situ growth methods [66,67]. As early as 1997, his team applied a patent "Method for increasing the corrosion resistance of aluminum and aluminum alloys" [68] and suggested that a protective hydroxalcite film formed on the surface of aluminum or aluminum alloys immersed in an alkaline lithium or alkaline Mg salt solution. This is of instructive significance. It is, however, more challenging to in situ grow LDH coatings on Mg alloys due to the fact that magnesium hydroxide is much easier to form in such alkaline solutions

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