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Recent advances in layered double hydroxide-based nanomaterials for the removal of radionuclides from aqueous solution☆

Pengcheng Gu [a], Sai Zhang [a], Xing Li [a], Xiangxue Wang [a], Tao Wen [a], Riffat Jehan [a], Ahmed Alsaedi [c], Tasawar Hayat [c, d], Xiangke Wang [a, b, c, *]

[a] College of Environmental Science and Engineering, North China Electric Power University, Beijing, 102206, PR China
[b] Collaborative Innovation Center of Radiation Medicine of Jiangsu Higher Education Institutions and School for Radiological and Interdisciplinary Sciences, Soochow University, 215123, Suzhou, PR China
[c] NAAM Research Group, Faculty of Science, King Abdulaziz University, Jeddah, 21589, Saudi Arabia
[d] Department of Mathematics, Quaid-I-Azam University, Islamabad, 44000, Pakistan

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ABSTRACT

Layered double hydroxides (LDHs), one of the most important two-dimensional layered compounds, have enabled massive developments in effective pollution treatments. Their derivative materials have also attracted multidisciplinary attention owing to the intrinsic advantages of their moderate chemiostability, low cost and nontoxicity. Over the past few decades, significant advances have been made in the synthesis of novel LDH-based composites and the optimization of characterization techniques. In this review, we give an overview of the recent advances in LDH-based nanomaterials, from a brief introduction to their preparation and modification methods to an overview of their application in the removal of radionuclides and an exploration of their underlying adsorption mechanisms. In the end, a summary and outlook are also briefly addressed. This review intends to provide deep insight into the design of high-performance LDH-based materials for the potential elimination of radionuclides from aqueous solutions during environmental pollution cleanup.

1. Introduction

With the rapid development of modern industries, environmental ecosystems and human health are seriously threatened by multifarious pollutants such as organic pollutants, heavy metals and radionuclides (Gao et al., 2017; J. Li et al., 2018; Song et al., 2017; Wang et al., 2017a). In the frontier field of environmental ecology, contamination treatment has become a paramount issue due to the widespread emergence of erosion-environmental effects (Yu et al., 2016a, 2018; Wang et al., 2015). As a distinguished new energy source, nuclear power, frequently referred to in "solving the energy crisis", has recently received attention for its ability to satisfy basic energy requirements and relieve energy pressures. Nevertheless, the extensive operation and utilization of nuclear energy will undisputedly produce radioactive pollution, thereby resulting in different levels of environmental pollution and potential toxicological effects. Therefore, it is of great urgency to develop highly efficient and environmentally friendly adsorbents for the preconcentration and solidification of radionuclides from aqueous solutions (Wen et al., 2013).

To date, various conventional and modern technologies, such as chemical precipitation (Kulkarni et al., 2013),ion-exchange (Ladeira et al., 2005), membrane separation (Jr et al., 1958) and adsorption (Gu et al., 2018; Yin et al., 2018) have been developed to separate and preconcentrate radionuclides from contaminated water. Each of these methods has been systematically studied by experimental and theoretical techniques for the separation of radionuclides. However, most of these methods still present limitations in terms of their high installation costs, secondary pollution and complex operation. Impressively, the adsorption technique has recently received increasing attention for its merits as a highly efficient, versatile and easy-to-operate technique for eliminating trace levels of radionuclides (X. Li et al., 2018).

In recent years, many kinds of adsorbents, including clay minerals (Choung et al., 2014), carbon-based materials, graphene

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* Corresponding author. College of Environmental Science and Engineering, North China Electric Power University, Beijing, 102206, PR China.
E-mail address: xkwang@ncepu.edu.cn (X. Wang).

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oxides (Yu et al., 2015), zeolite-based materials (Akyil et al., 1998), metal-organic framework material (Li et al., 2017; Liu et al., 2017; Sheng et al., 2017) and layered double hydroxides (LDHs), have been extensively studied for treating various pollutants. Among these adsorbents, LDHs represent a class of anionic clay composites that exist as naturally occurring minerals (Wang et al., 2016). The discovery of "natural layered double hydroxides" can be dated back to the 1840s. Since then, researchers have gradually found potential uses for LDHs, such as in redox reactions, photocatalysis and photoelectrocatalysis in the field of catalysis (Zhao et al., 2010). LDHs, as typical anionic layered structure compounds, are composed of positively charged host layers and guest interlayer anions, which are assembled by the non-covalent bond interaction. Generally, LDHs are expressed as the formula $[M^{2+}_{1-x}M^{3+}_{x}(OH)_2]^{x+}(A^{m-})_{x/m}\cdot nH_2O$, where M^{2+} and M^{3+} represent the divalent (Mg^{2+}, Zn^{2+}, Co^{2+}, Mn^{2+}, Ni^{2+}, and Ca^{2+}) or trivalent (Al^{3+}, Fe^{3+}, and Cr^{3+}) metal cations, A^{m-} is the interlayer charge-balancing anion (e.g., Cl^-, ClO_4^-, NO_3^-, CO_3^{2-}, and SO_4^{2-}), and x is regarded as the molar ratio of $M^{2+}/(M^{2+} + M^{3+})$ (Zhu et al., 2016). It is well accepted that the structural property of LDHs displays outstanding characteristics such as the following: (1) The existence of multiple alternative metal cations allows the chemical compositions of host layers to be controlled precisely. (2) The type and number of high-activity, interlayer anions can be substituted, which can contribute to the enhanced anion exchange capacity. (3) The layered property allows for tuning of their dimension and distribution in a wide range by intercalation of suitable anions. (4) A unique memory effect endows LDHs with the restorability to their original layered structures (Huang et al., 2017).

Owing to the special structural merits of LDHs, remarkable advances in catalysis have been achieved along with the simultaneous development of novel LDH-based materials and the expansion of new application fields (Choudary et al., 2004; Jiang et al., 2013; Yao et al., 2017). For example, MgCoAl-LDHs with a high catalytic activation energy (60.5 kJ/mol) were prepared by Li et al. (2009). Since then, two-dimensional (2D) LDH materials have received considerable attention in the field of wastewater treatment (Wen et al., 2013). In the early stage, various research efforts were devoted to investigate the adsorption of small molecules (Pavan et al., 1998). By virtue of their unique structure and impressive ion exchange capacity, some exciting improvements have been made in wastewater treatment based on LDH materials, manifested by a continuous increment in the literature publication numbers and the advancement of the understanding of intrinsic adsorption mechanisms. (Zhao et al., 2011). Despite of those remarkable advances achieved, as the advanced research topic in environmental ecology, functionalized-LDHs materials and their corresponding wastewater control still face some key challenges: (i) the fundamental aspects of design strategies for high performance and recycling of LDH-based nanomaterials are still unclear; (ii) to really realize the low-cost, high yields as well as environmentally friendly fabrication processes are all challenging tasks to be solved.

Considering these fundamental scientific issues and the general interest in environmental pollution management regarding to LDH-based materials, together with the substantial progress recently achieved in this field, a thematic review is absolutely desired. Moreover, a discriminating summary about the adsorption properties, interaction mechanism and application of LDH-based nanomaterials in radionuclides removal is still scarce. Herein, the main aim of this review is to provide a systematic overview of LDH-based materials and their application in radionuclide removal from aqueous solutions (Fig. 1). First, we will focus on their preparation and modification methods to offer an intuitive understanding of LDHs. Then, we will review the application of LDH-based materials in the removal of radionuclides from aqueous systems.

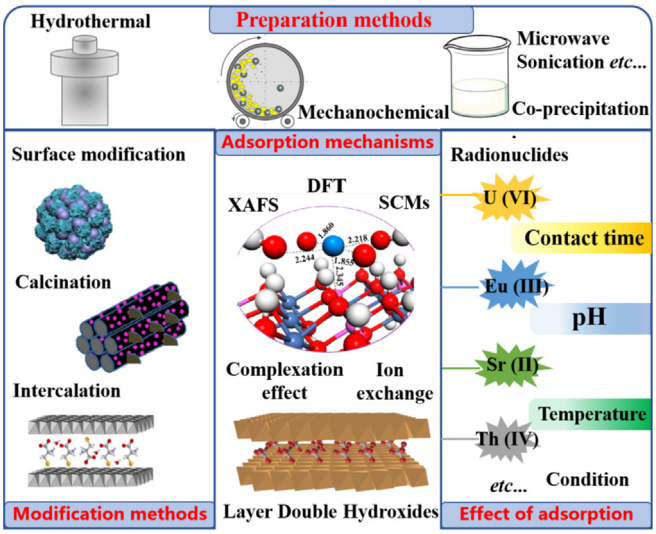

Fig. 1. An overall content of this review.

Subsequently, the effect of different environmental conditions is also summarized for various factors, i.e., pH, temperature and contact time. Finally, the interaction mechanisms of radionuclides and concise conclusions are briefly demonstrated. It is expected that this systematic summary will make a useful guide for the fabrication of highly efficient LDH-based adsorbents and provide key clues for the elucidation of the adsorption behaviors of radionuclides on LDH-based materials.

2. Preparation of LDHs

In the past few years, numerous LDHs with different morphologies have been successfully formulated *via* various strategies, including hydrothermal methods, co-precipitation and mechanochemical synthesis *etc.* (Crepaldi et al., 2000). To give a comprehensive understanding of LDHs, in this section, we will briefly outline the preparation methods of LDHs.

2.1. Hydrothermal method

Hydrothermal synthesis is a mostly fundamental method to prepare LDHs, wherein the autogenous pressure generated by an autoclave is frequently employed as the reaction medium. Experimentally, hydrothermal treatment at high temperature will result in the growth of regular crystallite shapes by virtue of the distribution of cations in the layers. Different morphologies and dimensions can be achieved by controlling the aging temperature and reaction time. A representative paradigm was reported by Lv et al. (2015). It was demonstrated that diverse LDH nanostructures, including nanofibers, nanosheets and nanoscrolls, could be formed by varying the reaction conditions. Nanoscrolls with irregular edges, 1D LDH nanoscrolls, and further evolution into regular nanosheets were observed under different aging times (6 h, 12 h and 24 h) (Fig. 2A). Interestingly, the originally nanosheets gradually evolved to perfect nanoscrolls with the increase of reaction temperature (Fig. 2B).

2.2. Co-precipitation approach

Co-precipitation provides a traditional approach to synthesize LDHs. In a typical coprecipitation process, a substrate with cations is immersed into a mixed solution of two metal salts, and then the