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Perspective

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Ultrafast capture of per- and polyfluoroalkyl substances by nitrate-intercalated layered double hydroxide through cationic geometry and functional interlayers

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Abstract

Per- and polyfluoroalkyl substances (PFAS) pose serious environmental risks because of their persistence and toxicity, with perfluorooctanoic acid (PFOA) being a widely studied example. By constructing a CuAl layered double hydroxide (LDH) with nitrate intercalation and cationic disorder, ultrafast PFAS capture capacity exceeding the theoretical limit was achieved, along with simultaneous pollutant degradation via thermal regeneration cycles, thereby addressing the issues of low capacity, slow kinetics, and secondary pollution inherent to traditional adsorbents. This report introduces a highly crystalline nitrate-intercalated Cu_xAl LDH that effectively removes PFOA under neutral pH and ambient conditions, achieving a remarkable adsorption capacity of 1,702 mg/g. The disordered cationic structure enhances the local charge density, facilitating rapid anion exchange without disrupting the LDH framework. Given its efficacy in continuous fixed-bed systems and real water samples, the material provides an integrated capture–destruction–recycling strategy, offering a scalable solution for mitigating PFAS contamination.

Keywords: PFAS, PFOA, Adsorption, Layered double hydroxide

Per- and polyfluoroalkyl substances (PFAS) are a class of persistent organic pollutants widely utilized in plastics, textiles, food packaging, and firefighting foams. These compounds are pervasive in the environment and present a serious threat to surface water, groundwater, and drinking water resources[1]. The pervasive environmental presence of perfluorooctane sulfonate, stemming from its wide range of applications, has led to inevitable human exposure. Perfluorooctanoic acid (PFOA), a notable emerging pollutant, is of specific concern because of its documented carcinogenic, neurotoxic, and immunotoxic effects. Its prevalence is underscored by its detection in 95% of human blood samples in US national surveys. There is increasing evidence that dietary sources and drinking water are the primary exposure pathways, thereby posing a substantial threat to both human health and environmental safety[2]. Recycling waste biomass and converting it into carbon-based adsorbents, such as biochar or activated carbon, represents a sustainable approach for capturing environmental pollutants, including PFOA[3]. However, these materials inherently suffer from low adsorption capacity and the generation of secondary waste, which limits their practical viability[4]. Layered double hydroxides (LDHs) possess a unique layered structure that enables them to chemically immobilize the target adsorbates within their lattice via anion exchange. This property has led to their widespread application for the adsorption and removal of diverse compounds. However, in the context of PFOA adsorption by LDH, our current understanding of how anion exchange within the LDH structure critically influences both adsorption capacity and the kinetics remains limited. Therefore, there is a pressing need to develop efficient, eco-friendly, and cost-effective methods through technological innovations to overcome these limitations. Ensuring the effective and sustainable removal of PFOA from the environment is essential to safeguard ecosystems and public health.

Kim et al.[5] recently reported an innovative strategy by developing an ultrafast capture–thermal destruction–recycling (CTR) process based on a highly crystalline and pure nitrate-intercalated Cu_xAl LDH. The core of their innovation lies in engineering the LDH structure to maximize both the capacity and kinetics. The weak interlayer electrostatic forces in this nitrate-intercalated material facilitate rapid anion exchange with PFOA under neutral pH conditions, resulting in an ultrahigh capacity of 1,702 mg/g, governed mainly by chemical exchange rather than physical adsorption. Rapid and efficient exchange of the intercalated nitrate ions with PFAS anions allows for ultrafast capture and immobilization of the

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pollutants, constituting the key functionality of the material's interlayer. By employing an optimized urea hydrolysis method, the authors precisely regulated both the OH^- concentration and the Cu/Al ratio to maintain a low pH environment, thereby achieving higher crystallinity and phase purity. Moreover, when PFOA exceeds its critical micelle concentration, the formation of a semi-gel or gel on the LDH surface triggers additional physical adsorption, causing the total uptake to exceed the theoretical ion exchange limit. They reported that the CTR process for the removal of PFOA utilizing the anion exchange capability of LDH. Anion exchange in LDH relies on electrostatic interactions between the interlayer's cations and anions, the strength of which depends on the anion species. For instance, carbonate ions in the LDH interlayer exhibit strong electrostatic attraction, making them unfavorable for anion exchange. In contrast, nitrate ions experience weaker electrostatic binding, which promotes efficient anion exchange with PFOA, especially at pH levels where PFOA exists in its anionic form.

The hydrogenation behavior of Cu_xAl-NO_3 LDH makes it amenable to characterization by ^2H magic angle spinning nuclear magnetic resonance (MAS-NMR). By comparing the peak intensity ratios across three distinct sites in all synthesized LDH samples, it becomes evident that the $Cu_{2.35}Al$ LDH exhibits a higher degree of structural disorder relative to the other compositions. This disorder appears to correlate with the observed kinetics of PFOA adsorption. Thus, the cation layer disorder in LDH enhances the local positive charge density, thereby facilitating the rapid attraction of anions into the interlayer space during the anion exchange process. The multiple lattice spacings observed in the interlayer region suggest a random orientation of PFOA molecules, resulting from a mixture of PFOA and hydroxide ions at various angles rather than a unidirectional arrangement. This implies that the anion exchange between nitrate and PFOA does not significantly alter the cationic layers of the LDH.

To further verify that PFOA adsorption does not induce structural changes, the authors employed Cu K-edge wavelet-transformed extended X-ray absorption fine structure (WT-EXAFS) analysis. The spectral features remained unchanged after PFOA adsorption, providing direct evidence that the local electronic structure and overall framework of the LDH were preserved. The adsorption capacity of the PFOA-saturated Cu_2Al-NO_3 LDH was successfully restored via thermal regeneration. Although the initial adsorption rate decreased from 10.0 h^{-1} in the first cycle to 4.0 h^{-1} by the fifth cycle, the thermal treatment effectively regenerated the LDH structure, demonstrating the robustness and reusability of the adsorbent.

The performance of the adsorbent in a continuous flow system was evaluated further using a fixed-bed adsorption column. The breakthrough curves were analyzed at three empty bed contact times (EBCTs), corresponding to flow rates of 0.5, 1, and 2 mL/min. Through accelerated tests using a 75 mg/L PFOA influent, the authors demonstrated that higher flow rates directly correlated with reduced breakthrough and exhaustion times. These results demonstrate that Cu_2Al-NO_3 LDH is a promising adsorbent for the continuous removal of PFOA from aqueous systems.

To evaluate the practical applicability of Cu_2Al-NO_3 LDH in PFAS treatment, the authors spiked various real water samples with 0.1 g/L PFOA. These samples included the tertiary effluent from a municipal wastewater treatment plant (WWTP), as well as the influent and effluent from a drinking water treatment plant (DWTP). The impact of coexisting anions was examined using $CaCl_2$ and Na_2CO_3. PFOA adsorption was only mildly suppressed by high concentrations of $CaCl_2$ (≥50 mM) but was severely hindered by Na_2CO_3. This pronounced inhibition by Na_2CO_3 stems from a competitive anion exchange process: Bicarbonate (HCO_3^-) exhibits strong electrostatic attraction to the LDH interlayer, outcompeting PFOA at a neutral pH. As bicarbonate is the primary form of alkalinity in neutral water, the elevated alkalinity levels found in real-world waters, such as the influent of wastewater treatment plants and tertiary effluent, are likely to diminish the adsorption efficiency of the Cu_2Al-NO_3 LDH, in contrast to its performance in the lower-alkalinity effluent of a DWTP. The results indicate that using Cu_2Al-NO_3 LDH for the post-treatment of effluents from advanced or secondary treatment plants could effectively enhance the removal of PFOA within conventional treatment systems.

Practical deployment of Cu_xAl-NO_3 LDH is facilitated by its ultrafast adsorption and high capacity, which break through conventional limits, and by its capture–pyrolysis–regeneration cycle for complete removal of PFAS. Although the successful capture in the laboratory is undeniable, the destruction and regeneration phase warrants critical assessment. The thermal treatment at 500 °C for regeneration, co-heated with $CaCO_3$, achieved a defluorination of only approximately 54%. This may be attributed to the significant reduction of organic fluorine compounds (e.g., PFOA), as indicated by the weakened F1s peak after heating, along with the decreased binding energy associated with the metal fluoride peak. This pivotal yet cautious finding implies that nearly half of the captured fluorine could be released as other potentially harmful organofluorine compounds.

Before this can be considered a true "destruction" technology, comprehensive analysis of gaseous and solid byproducts is essential to avoid simply transforming one pollutant into another. Moreover, the energy-intensive nature of regeneration at 500 °C and the relatively high cost of copper compared with other metals pose techno-economic hurdles for the method's large-scale application. Future research should therefore focus on (1) enhancing the defluorination efficiency to > 99.9% by exploring advanced catalytic systems or alternative energy sources; (2) improving the material's design for better selectivity against competing anions like sulfate and for long-term cycling stability; and (3) conducting pilot-scale trials to bridge the gap from laboratory to field.

We believe that the capture and destruction of PFAS by LDHs will open a new avenue for water security through the continuous development of materials science and engineering.

Author contributions

The authors confirm their contributions to the paper as follows: investigation: Wang Y, Chen C; writing – original draft: Wang Y; Writing – review and editing: Wang X. All authors reviewed the results and approved the final version of the manuscript.

Data availability

All data used in this article are derived from public domain resources.

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Declarations

Competing interests

The authors declare that they have no conflict of interest.

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