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Adsorption characteristics of nitrite on Friedel's salt under the landfill circumstance

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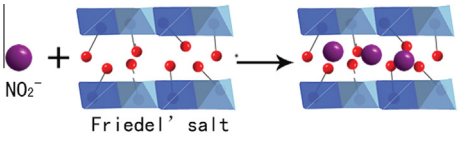
HIGHLIGHTS

- The adsorption capacity of the nitrite on Friedel' salt is 3.817 mg g^{-1}.
- The adsorption can be well described by the Lagergren-first-order kinetics.
- The adsorption agrees well to the Langmuir–Freundlich model.
- The adsorption can be affected by the variation of the landfill circumstance.
- The migration of nitrite in the landfill can be affected by Friedel's salt.

GRAPHICAL ABSTRACT



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ABSTRACT

Landfill sites contain abundance of Friedel's salt. In this study, Friedel's salt was prepared by a coprecipitation method and characterized by the X-ray diffraction (XRD) pattern and Fourier transform infrared (FTIR) spectroscopy. The adsorption characteristics of nitrite on Friedel's salt were investigated to reveal the effect of Friedel's salt on the migration of nitrite in the landfill. The adsorption was an exothermic process and could be well described by Lagergren-first-order kinetics and Langmuir–Freundlich model, with a maximum adsorption capacity of 3.817 mg g^{-1}. Low pH was beneficial to the adsorption process. However, the Cl^-, SO_4^{2-}, PO_4^{3-} and organic matter in the leachate could restrict the adsorption to a low level. The results suggested that Friedel's salt could affect the migration of nitrite in the landfill, which was related with the variation of the landfill circumstance.

1. Introduction

Landfill is still the most widely used technology for the treatment of municipal solid waste (MSW) in lots of countries. For instance, more than 90% of MSW is disposed by landfill in China [1]. Among the solid waste disposed at landfill sites, the construction and demolition (C&D) waste frequently accounts for 10–30% around the world [2,3]. Except for the C&D waste, Municipal solid waste incinerator (MSWI) bottom ash is also allowed to be disposed in landfill sites in several countries and areas, such as China, Japan and Taiwan [4,5]. It is used as the liner, protection layer and drainage layer instead of natural minerals in the landfill [6,7]. The C&D waste contains Friedel's salt, which is formed as soon as the Portland cement was attacked by the chloride ions. Several studies have indicated that Friedel's salt is the most abundant compound in the C&D waste [8,9]. MSWI bottom ash is also reported to contain considerable amounts of Friedel's salt. For example, Ito et al. [10] found that the content of Friedel's salt in MSWI bottom ash was 4.16%. Besides, Talero [8] and Grishchenko et al. [11] pointed out that the neoformation of Friedel's salt was predicable with the existence of reactive alumina, iron oxide and chloride ions. As a matter of fact, these minerals prevail in the landfill. The neoformation of Friedel's salt can further increase the level of Friedel's salt in the landfill.

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Friedel's salt ($3CaO{\cdot}Al_2O_3{\cdot}CaCl_2{\cdot}10H_2O$) is a Layered double hydroxide (LDH) composed of hydroxide layers of edge-sharing octahedra occupied by Ca^{2+} and Al^{3+}, with an interlayer composed of Cl^-. The chloride ion occupied in the interlayer could exchange with other ions. Due to the high exchange capacity of chloride ion and specific area, Friedel's salt was identified as a superior adsorbent for the anionic contaminants [12,13]. Several researches have been done to use Friedel's salt to remove the anionic contaminants from the liquid solution. For example, Dai et al. [14] use the Friedel's salt to remove CrO_4^{2-} from the waste water, the removal efficiency could be up to 99%. Zhang et al. [15] found Friedel's salt had the great adsorption capacity for AsO_3^-. Zhang et al. [16] found Cd in the waste water could be successfully removed by Friedel's salt. Ma et al. [17] found Friedel's salt could remove 95% silicates from the sodium aluminate solution. Lots of studies have revealed that Friedel's salt in the concrete could protect the steel from the attack of Cl^- and extended the longevity of the concrete, owing to the adsorption of Friedel's salt for Cl^- [18,19].

Landfill leachate contains abundant anions, including NO_2^- [20]. NO_2^- may be adsorbed on Friedel's salt when the leachate flows through it. Therefore, the migration and transformation of NO_2^- in the landfill will be altered. NO_2^- is the intermediate product of the transformation of nitrogen, including nitrification and denitrification [21]. The uptake of NO_2^- by Friedel's salt may affect the nitrogen degradation of the landfill, which has turned out to be the research focus in the last decades [21–25]. Besides, nitrites can combine with amines to produce N-nitroso compounds, which are mutagenic, teratogenic, and carcinogenic. A high level of nitrite may inhibit the vitality of the microorganism and affect the stabilization process of the landfill. The release of nitrite from landfill can also cause the blue baby syndrome among children. The uptake of nitrite by Friedel's salt may mitigate the migration of nitrite and reduce its toxicity to the surrounding environment. Thereby, it is necessary to study the adsorption characteristics of nitrite on Friedel's salt, to reveal its effect on the nitrite migration in the landfill. Unfortunately, to our knowledge up to the date, there are few studies on the adsorption characteristics of nitrite on Friedel's salt, especially under the landfill circumstance.

In this study, Friedel's salt were synthesized by a coprecipitation method and characterized by the X-ray diffraction (XRD) pattern and Fourier transform infrared (FTIR) spectrum. The kinetics and isotherms of the adsorption were investigated. The effects of the environmental parameters, such as the pH, temperature and anions competition on the adsorption were also examined. We aim to provide insights into the effect of Friedel's salt on the migration of the nitrite in the landfill.

2. Materials and methods

2.1. Chemicals and materials

All chemicals used in this study were of analytical grade and purchased from Sinopharm Chemical Reagent Co. Ltd. Distilled water was used all through the experiments. All the glassware was cleaned by 5% HNO_3 and rinsed three times with the distilled water. Nitrite stock solution was prepared by $NaNO_2$.

2.2. Preparation of Friedel's salt

Friedel's salt was prepared by a coprecipitation method, as described elsewhere [15,17]. Briefly, two solutions were first prepared, one contained 0.5 M $CaCl_2$ (500 mL), which was preheated to 50 °C. Another one contained 0.25 M $NaAlO_2$. Then, the $NaAlO_2$ solution was added to the $CaCl_2$ solution at the rate of 5.0 mL min^{-1} by a peristaltic pump and the mixture was vigorously stirred at 300 rpm. The synthesis of Friedel's salt was performed under the nitrogen protection. After 1 h reaction, the precipitate was collected by the filtration, washed three times with distilled water to remove residual substances and dried in a vacuum oven at 50 °C for 10 h. The obtained sample was ground and stored in a desiccator for the further use.

2.3. Characterization of Friedel's salt

XRD measurements were carried out using an XRD diffractometer (D8-Advance, Bruker). The patterns with the Cu Ka radiation (k = 0.15406 nm) at 40 kV and 30 mA were recorded in the region of 2 h from 5° to 80°. The FTIR spectrum was obtained on a Shimadzu IR Prestige-21 spectrometer with the resolution of 4 cm^{-1}. The KBr/sample discs were prepared by mixing 1% of samples in KBr.

2.4. Batch adsorption studies

Batch adsorption experiments were conducted and equilibrated using a model KYC-1102 air-temperature-controlled shaker (Ningbo Jiangnan Instrument Factory, China) at 100 rpm. The solution was then continually flushed with N_2 to avoid the contact with the atmosphere. Batch experiments were conducted in triplicate to ensure the accuracy of the obtained data.

According to our previous research, the level of NO_2^- in landfill leachate ranged from 0.38 to 2.24 mg L^{-1} (see Fig. S1). Therefore, the initial concentration of nitrite used in this study was 2.0 mg L^{-1}. Besides, the dosage study showed that the removal efficiency of NO_2^- reached the platform when the dosage of Friedel's salt increased to 2.0 mg (see Fig. S2). Thus, 2.0 mg of Friedel's salt was added during the adsorption study.

The adsorption kinetics of nitrite on Friedel's salt was studied by adding 100 mL nitrite solution (2.0 mg L^{-1}) and 2.0 g Friedel's salt to 100 mL beaker for the contact time ranging from 5 to 150 min at 25 °C.

Isotherm studies were conducted with a constant Friedel's salt dosage (2.0 g) and 100 mL nitrite solution with the initial concentrations varying from 1.0 to 8.0 mg L^{-1}. The mixture was equilibrated at 25 °C for 120 min.

The temperature of the practical landfill sites varied from 22 to 52 °C [21]. Thereby, the effect of the temperature on the adsorption was conducted over 20–50 °C. 100 mL nitrite solution (2.0 mg L^{-1}) and 2.0 g Friedel's salt were added and equilibrated at the desired temperature for 120 min.

The effect of pH on the adsorption was conducted at the range from 5.0 to 10.0, which was corresponding to the variation of leachate pH [25]. 100 mL nitrite solution (2.0 mg L^{-1}) and 2.0 g Friedel's salt were added and equilibrated under the desired pH condition for 120 min.

The coexisting ions, such as Cl^-, SO_4^{2-} and PO_4^{3-} were imposed to explore their impacts on the adsorption. The concentration of Cl^-, SO_4^{2-} and PO_4^{3-} ranged from 0 to 5000 mg L^{-1}, 0 to 2000 mg L^{-1} and 0 to 10 mg L^{-1}, respectively, which were set according to our previous research (see Figs. S3 and S4) and literature report [20]. The mixture was equilibrated at 25 °C for 120 min.

Besides, the acetic acid with the concentration ranged from 0 to 40,000 mg L^{-1} was imposed to investigate the effect of organic acid on the adsorption. The concentration of the acetic acid was corresponding to the variation of COD in the leachate (see Fig. S5, 1.0 g acetic acid equal to 1.06 g COD). The mixture was equilibrated at 25 °C for 120 min.

After the adsorption, the samples were centrifuged and NO_2^- concentration in supernatant was determined immediately by ion chromatograph (Dionex ICS-2000, USA).

The amount of nitrite absorbed on the Friedel's salt (q_t) and the removal percentage of nitrite (η%) were calculated according to Eqs. (1) and (2), respectively: