

Synthesis of a hydrotalcite-like compound from oil shale ash and its application in uranium removal



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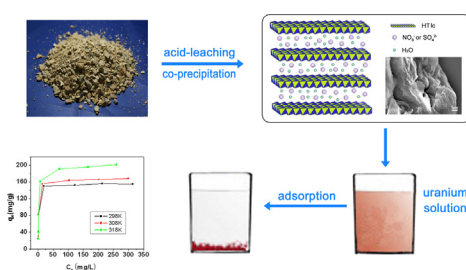
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HIGHLIGHTS

- Waste product of oil shale industry was utilized as sorbent for uranium.
- Hydrotalcite-like compound was synthesized using oil shale ash.
- Composition, microstructure and absorptive property were investigated.
- It was an Mg–Al-based hydrotalcite compound with a lamellar morphology.
- The maximum adsorption capacity toward uranium is 156 mg/g at 298 K.

GRAPHICAL ABSTRACT



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ABSTRACT

Oil shale ash (OSA) was utilized to synthesize a hydrotalcite-like compound (HTlc) via a facile acid-leaching and co-precipitation process. Chemical component, structural and morphological properties of HTlc were characterized by ICP-AES, FTIR, XRD, HRTEM, SEM, and XPS. The results showed that HTlc is Mg–Al based hydrotalcite with a lamellar morphology, incorporating other metal cations in its structure. Building on the synthetic HTlc, adsorption of uranium from aqueous solution was performed in a single component system with the variation of pH, adsorbent dosage, contact times, temperatures and initial concentrations of uranium. It is found that the adsorption process conforms to Langmuir isotherm model. Upon raising temperature from 298 to 318 K, the maximum adsorption capacity increases from 156 to 200 mg/g. The adsorption of uranium is revealed to follow the pseudo-second-order rate model by adsorption kinetics studies. Obtainable thermodynamic parameters showed that the uranium adsorption is endothermic and spontaneous in nature. Comparison with similar adsorbents demonstrates that HTlc is an effective and economic adsorbent to remove uranium from aqueous solution. And thus, the present work is expected to provide a new strategy for disposal of both wastewater of nuclear industry and waste product of oil shale industry.

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1. Introduction

Uranium plays a central role in the nuclear industry, which has emerged as pollutant of the environment because of its long life time, as well as highly radiological and biological toxicity [1,2]. Adsorption is an essential approach to remove uranium from wastewaters, where adsorbent is a key factor to determine the final effects of adsorption process. A great number of materials have been investigated for their adsorbing properties, such as coir pith [3], zeolite [4–6], akaganeite [7], goethite [8], modified kaolin [9], modified diatomite [10], modified pine sawdust [11], and dried tea and coffee wastes [12]. Among them, adsorbents fabricated from waste materials have gained a significant interest due to its double benefits of both resource re-generation and pollution abatement.

As one of the largest energy resources in the world, oil shale has been utilized as a source of liquid fuel. Its by-product of oil shale processing, OSA, might be generated from oil shale reserves run and estimated to be about 3.2×10^{11} tons. [13] It has shown that only in China, the total discharge of OSA is estimated to be more than 800,000 tons annually. [14] The dumped OSA is considered as a serious environmental pollutant, even 'hazardous waste' because of the high alkalinity of their leachates [15]. Therefore, it is necessary to develop new approach to reduce accumulation of OSA and further explore its new application. In a variety of reported application, the conversion of this ash into an adsorbent to remove heavy metals provides a two-fold advantage with respect to energy and environmental management. The dumped OSA is capable of being, to some extent, reduced due to its conversion into more useful and highly value-added adsorbents, which contributes to the further development of the oil shale industry. On the other hand, the cost-effective adsorbent from OSA may treat wastewaters with reasonable cost, which partially solves wastewater treatment problem. [16] It has been reported that OSA was converted into zeolite and used for removal of copper, cadmium and lead from wastewater [17,18]. To the best of our knowledge, study on the uranium removal using OSA or its converted products remain rare.

OSA is mainly composed of SiO_2 , Al_2O_3 , MgO , CaO , Fe_2O_3 , and extremely few other metallic oxides [19], allowing for OSA suitable for the raw material to produce HTlc. Layered double hydroxides (LDHs) have attracted considerable attention in recent years because of their promising applications in areas of nuclear waste disposal and storage. [20] LDHs have an anion-exchange capability and feature with $[\text{M}_{1-x}\text{M}_x^{n+}(\text{OH})_2](\text{An}^-)_{x/n} \cdot m\text{H}_2\text{O}$ (M^{2+} contains Mg^{2+} , Ca^{2+} , Zn^{2+} , etc.; M^{n+} is Al^{3+} , Fe^{3+} , Ti^{4+} , Ce^{4+} etc.; An^- represents interlayer anions; and x typically ranges from 0.20 to 0.33) [21,22]. The typical lamellar structure and the interlayer anion-exchange behavior make LDHs useful for absorption of uranium from aqueous solution. [2] In our previous study, we prepared Mg-Al LDHs and Ca-Al LDHs with co-precipitation method, which exhibit high adsorptive capacity of uranium [21,23]. After modified by citrate acid, Mg-Al LDHs showed higher adsorption capacity for uranium [21]. The maximum adsorption capacity of Ca-Al LDHs with nano-hydroxyapatite is 207.9 mg/g at 298 K [2]. Although being effective adsorbents for uranium, the use of LDHs have been greatly restricted due to their cost and loss of adsorption efficiency after regeneration. Therefore, exploration of cheaper and more effective adsorbents is highly demanded. In this respect, we have developed some approaches to fabricate LDHs by means of natural materials and waste materials as starting raw materials. [24,25]

In this work, HTlc has been prepared from OSA. A facile combined leaching and precipitation treatment has been employed. In this process, effect of various factors possibly existing in natural condition on uranium sorption as well as relevant adsorption kinetics and thermodynamics have been systematically evaluated. The results demonstrated that HTlc, composed of Mg-Al based hydroxalcalite, is an effective adsorbent. The study is not only to develop

a low-cost and effective adsorbents to remove uranium from the solution, but also to explore a new and proper strategy for OSA handling, disposal and utilization.

2. Materials and methods

2.1. Treatment of oil shale ash

Samples of oil shale stones were collected from Yinan area in the northeast of China. These samples were crushed and sieved to a particle sizes less than 2 mm, burned in muffle furnace at 950°C for 8 h to remove all incorporated hydrocarbons, and the remaining ash was grinded and stored. [26] The chemical compositions of the oil shale and OSA were determined by XRF.

OSA (20 g) was mixed with sodium chloride (8 g), and heated at 700°C for 3 h then air-dried. The ash samples were acid-leached with 30 wt.% sulfuric acid solution (200 mL) at 80°C for 6 h with constant stirring. [27] After the reaction was completed, the leach liquor was separated from the residue by vacuum filtration. [28]

2.2. Synthesis of HTlc

The nitrate magnesium solution (50 ml, 4 M) was added to the leaching liquor in order to adjust $\text{M}^{2+}/\text{M}^{3+}$ molar ratios. The mixed solution was adjusted to pH 10.5 with a saturated solution of sodium hydroxide, then the reaction mixture was further aged for 8 h at 80°C . HTlc was obtained by filtering the resultant suspension, washed with deionized water and dried in vacuum at 60°C for 24 h. The chemical compositions of HTlc were determined through acid decomposition.

2.3. Characterization

The elements contents of oil shale and OSA were measured on a Bruker S4 Explorer X-ray fluorescence spectrometer. The concentrations of metal ions in the mineral leachates solution and HTlc were analyzed using ICP-AES (Optima-7000DV). Qualitative chemical structure assessment was done by FT-IR analysis with an AVATAR 360 FT-IR spectrophotometer using a standard KBr pellet technique. The Crystal structure of samples was analyzed by XRD patterns of the solid products, using a X'Pert Pro XRD analysis with Cu-K α radiation ($k = 0.154178$ nm). Morphology was characterized using HRTEM and SEM. Powder samples for the HRTEM observation were dispersed in ethanol by ultrasound and mounted on a carbon-coated copper microgrid, TEM images were taken by HRTEM (JEM-2100) with an acceleration voltage of 200 kV. SEM images were measured on a JEOL JSM-6480 with an energy-dispersive X-ray spectroscopy (EDS) instrument. XPS measurements were performed using a PHI 5700 ESCA spectrometer with monochromated Al K α radiation ($h\nu = 1486.6$ eV). All XPS spectra were corrected by the C1s line at 284.5 eV.

2.4. Removal of uranium

All the sorption experiments were performed in a series of conical flasks (100 ml). A given dose of adsorbents were shaken together with the uranium solution (50 mL) of given concentration and pH value in a thermostatic water shaker using shaking rate of 200 rpm. After the sorption reached the equilibrium, the solid material was separated by centrifugation. The concentration of uranium in the solution was analyzed using a Trace Uranium Analyzer. The adsorption capacity (q_e mg/g), the uranium loading per unit mass of HTlc, was calculated using the following equation:

$$q_e = \frac{(C_0 - C_t)V}{W} \quad (1)$$

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