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Removal of strontium ions from aqueous solution using hybrid membranes: Kinetics and thermodynamics☆



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ABSTRACT

The removal of radionuclide from radioactive wastewater has captured much attention. Strontium-90 is one of the major radionuclides. To develop a new type of adsorbents to remove strontium ions from the radioactive wastewater, in this study, novel hybrid membranes were prepared and characterized. The adsorption kinetics, thermodynamic parameters of ΔG , ΔH and ΔS , as well as surface SEM and EDS images were used to investigate the removal of strontium ions from stimulated radioactive wastewater using the previously prepared hybrid membranes as efficient adsorbents. The study of kinetic model confirmed that the adsorption of strontium ions on these hybrid membranes followed the Lagergren pseudo-second order model. Moreover, it was proved that the adsorption of strontium ions on these samples was solely controlled by intraparticle diffusion. The negative values of ΔG and the positive values of ΔH indicated that the adsorption of strontium ions on samples A–D is a spontaneous and endothermic process in nature. Furthermore, surface SEM and DES images give significant evidence to confirm the existence of strontium ions on the surface of the adsorbed samples. These findings demonstrate that these hybrid membranes are promising adsorbents for the removal of strontium ions from aqueous solution and can be potentially applied in the adsorptive separation of radionuclides from the radioactive wastewater.

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1. Introduction

With the rapid development of modern industry and technologies, fossil fuels such as petroleum, coal, natural gas and other non-renewable energy resources will be exhausted in the next decades. Researching new energy supply route to substitute these non-renewable energy resources is thus vital importance. Hopefully, nuclear electric power generation is regarded as one of the most promising approaches to substitute non-renewable energy resources for new energy supply. As a result, the construction of nuclear power station is escalated to a new level in many developed and developing countries, especially in China. Along with the rapid growth of nuclear power station, radioactive wastes containing various radionuclides, such as strontium (^{90}Sr), cobalt (^{60}Co) and cesium (^{137}Cs), etc., are unavoidably produced during the operation of nuclear plant [1,2]. Among which, both chemical and radiological toxicity caused by these radionuclides threat not only the inhabited surroundings, but also the health of human body. Especially, the accidents at the Fukushima nuclear plant (Japan) caused by the earthquake has sounded the alarm to the world and worried the

inhabitants [3,4]. Consequently, the contamination from these radioactive wastes attracts much public attention in recent years. How to dispose the radioactive wastewater from the nuclear power plants challenges the researchers and the administrators. Developing new techniques to remove or delete these radionuclides from the polluted aqueous solution is thus turned into an utmost urgent issue.

As one of the major radioactive isotopes, strontium (^{90}Sr) has similar properties to calcium and can easily replace it in the human body to induce cancer and some other diseases [1,2]. Consequently, it is urgent to delete radioactive isotopes, such as strontium (^{90}Sr) from the radioactive wastewater. However, strontium (^{90}Sr) is extremely dangerous and will endanger the health of researchers. The researches on the removal of strontium (^{90}Sr) from the radioactive wastewater are thus turned into challenge. Hopefully, this trouble can be removed using the stimulated radioactive wastewater containing strontium (Sr^{2+}) ions to replace the real process, i.e. selecting strontium (Sr^{2+}) ions as the replacement of strontium (^{90}Sr) to perform the stimulated theoretical explanation. This is because strontium (Sr^{2+}) ions have the same number of the outer electrons as that of strontium (^{90}Sr) except its difference in the number of neutrons. Thus the performance characteristics of strontium (Sr^{2+}) ions are identical to those of strontium (^{90}Sr) [5]. Due to these reasons, the removal of strontium ions from aqueous media is therefore significantly important.

To remove or eliminate strontium ions from aqueous solution, various innovative strategies, such as adsorption using materials or particles

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[1,2], biosorption [6], extraction [7], ion exchange [8], and membrane process [9], etc., have been recently designed. Among them, adsorption using functionalized materials as adsorbents is regarded as one of the most effective techniques to remove strontium ions from water. As a result, a variety of functionalized materials have been prepared for strontium ion removal using adsorption technique and many researchers have conducted such investigations [1,2]. For example, Cheng *et al.* [1] prepared a type of magnetic Fe₃O₄ particles modified sawdust (MCS) using chitosan as the bridging reagent to remove strontium ions from aqueous solutions. It was found that the adsorption was a physical process and followed the pseudo first-order model. Wang and co-worker [2] synthesized the magnetic chitosan beads to remove Sr²⁺ ions from aqueous solution. It was reported that –NH₂ was mainly involved in Sr²⁺ ion sorption by magnetic chitosan. From these examples, it can be discovered that the functionalized groups play an important role in the adsorption separation of strontium ions from aqueous solutions. Unfortunately, the application of these functionalized materials as adsorbents also encounters many technique problems, such as frangibility and hard to be recovered due to its tiny particle size. Such problems will block their further utility in industrial processes. Thus adsorption for strontium ions using membrane-shape adsorbents containing functionalized groups will be more attractive than the use of particle-shape adsorbents.

Presently, inorganic/organic hybrid materials and the related hybrid membranes have been used as solid adsorbents for the removal of heavy-metal ions from aqueous solutions and revealed high removal efficiency [10–13]. These hybrids not only combine the advantages of organic and inorganic materials, but also exhibit some distinguished properties, such as structural flexibility, thermal and mechanical stability. Their unique properties make them have high advantages over others in the separation and recovery of heavy-metal ions from aqueous media. However, little work is done so far to remove or recover strontium (Sr²⁺) ions from aqueous solution *via* adsorption separation using hybrid membrane as a solid-state sorbent [14].

Recently, much effort was made to prepare functionalized hybrid materials and the related hybrid membranes used as adsorbents for the removal of toxic heavy-metal ions from aqueous solutions [13, 15–17]. Our continuing interest in these hybrid adsorbents makes us do more. Therefore, to develop a membrane-shaped hybrid adsorbent and extend its potential application in the removal of radionuclides strontium (Sr²⁺) ions from radioactive wastewater, herein, a novel route for the preparation of hybrid membranes via a sol–gel reaction will be reported and their adsorption behaviors for strontium ions are investigated.

Compared with the previous articles [13,15–17], the particularity of this new approach is that: (1) the adsorbent was fabricated as membrane-shaped appearance, which is different from the conventional particle-shape adsorbents, (2) the functionalized groups (–NH₂) were incorporated into the hybrid membranes via a sol–gel reaction between PVA and silicone containing –NH₂ groups, and (3) the application of hybrid membrane adsorbents will be extended to a new field of radionuclide removal. Moreover, their adsorption performances for strontium (Sr²⁺) ions were examined as a typical model for the removal of radionuclides from the stimulated radioactive wastewater.

2. Experimental

2.1. Materials

N-[3-(trimethoxysilyl)propyl] ethylene diamine (TMSPEDA, purity ≥ 95.0%) was purchased from Silicone New Material Co. Ltd of Wuhan University (Wuhan City, China) and used without further purification. Poly(vinyl alcohol) (PVA, DP: 1750 ± 50) was purchased from Shanghai Chemical Reagent Co., Ltd. (Shanghai City, China) and used as received. Other reagents were of analytical grade and used as received.

2.2. Fabrication of hybrid membrane adsorbents

The fabrication of these hybrid membrane adsorbents could be described briefly as follows. First, 25 g PVA was dissolved in 475 ml deionized water and stirred vigorously for 2 h at 90 °C to produce 5% aqueous PVA solution. Second, prescribed amount of TMSPEDA (the amount of TMSPEDA in samples A–D was 2.5 g, 5.0 g, 10.0 g and 20.0 g, respectively) was added dropwise into 100 g of the above-prepared 5% aqueous PVA solution within 2 h. Third, the mixed solution was stirred vigorously for additional 5 h to conduct the sol–gel reaction so as to form the coating solution. Following that, a homogeneous sol could be obtained and the Si–O–Si bonds in the coating solution might occur between the Si and O elements. Subsequently, the previously prepared sol was aged for 24 h and spread on a teflon plate to produce the desirable hybrid membrane. Finally, it was air-dried at room temperature for 2–4 days, and then kept at 50 °C for 12 h to obtain the final hybrid membrane adsorbents. To increase the membrane thickness, this step might be repeated several times.

2.3. Adsorption experiments

The adsorption experiment of hybrid membrane adsorbents for strontium ions was conducted in a static state. Some chief influencing factors, such as initial pH, contact time and solution temperature, were investigated. The content of strontium (Sr²⁺) ions in aqueous Sr(Cl)₂ solution was determined using atom adsorption spectrum (PE, 900T). The adsorption capacity ($q_{Sr^{2+}}$) of Sr²⁺ ions can be calculated by Eq. (1):

$$q_{Sr^{2+}} = \frac{(C_0 - C_R)V}{W} \quad (1)$$

where V is the volume of aqueous SrCl₂ solution (L); C_0 and C_R are the concentration of initial and remaining Sr²⁺ ions (g·L⁻¹), respectively; W is the mass of the sample (g).

To obtain a better pH scale for strontium ion adsorption, the initial pH of the solution was firstly examined, in which the pH value of the solution was adjusted with 0.01 mol·L⁻¹ aqueous NaOH or HCl solution. For adsorption kinetic studies, the prepared sample was immersed in 0.01 g·L⁻¹ aqueous Sr(Cl)₂ solution for different adsorption times at pH 6. The adsorption experimental data were analyzed using typical two-parameter theoretical models, such as the Lagergren pseudo-first-order and pseudo-second-order kinetic equations, and intraparticle diffusion model. On the basis of adsorption data, the thermodynamic parameters, ΔG , ΔH and ΔS , at different temperatures for the removal of strontium (Sr²⁺) ions, were calculated.

3. Results and Discussion

3.1. The effect of influencing factors on adsorption capacity

3.1.1. The effect of initial pH on adsorption capacity

Initial pH of solution has a strong effect on the adsorption of metal ions on an adsorbent. To gain a better pH scale for strontium ion adsorption, the pH in the range of 2 to 10 at a concentration of 0.01 g·L⁻¹ at 40 °C for 24 h was selected to examine the effect of the initial pH on strontium adsorption. The effect of the initial pH on the adsorption capacity of strontium ions was conducted and is presented in Fig. 1.

As shown in Fig. 1, it can be found that for the individual sample, the $q_{Sr^{2+}}$ value increased from samples A to D, suggesting that the $q_{Sr^{2+}}$ value increased with an increasing content of TMSPEDA in samples A–D. Such result implies that TMSPEDA might play an important role in the removal of strontium ions from aqueous solution. Moreover, it can also be found that for samples A–D, the $q_{Sr^{2+}}$ value increased with an increase in the initial pH and reached its highest value at pH 6. Subsequently, it decreased as the initial pH was higher than 6.

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