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Removal of cesium ions from waste solution using sericite incorporated into nickel hexacyanoferrate

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Abstract−To increase adsorption capacity and selectivity for cesium ions from waste solution, sericite was chemically modified by means of nickel hexacyanoferrate (NiHCF) with a high selectivity trap agent for cesium. Scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) spectroscopy were used for the characteristic analysis of surface onto the NiHCF-sericite. The adsorption capacity of cesium ions for the NiHCF-sericite increased about 2.5 times, as compared with natural sericite at initial pH 5.0 of waste solution. Adsorption equilibrium was investigated by Langmuir and Freundlich isotherm model, respectively. Maximum adsorption capacity was estimated as 16.583 mg/g, and the Langmuir isotherm fits the adsorption data better than Freundlich model. The adsorption process was determined as an exothermic reaction and all adsorption was completed in 30 min. In addition, the adsorption capacity of cesium ions was not greatly affected by ionic strength (~0.1 M NaCl concentration) and other metals in mixed waste solution.

Keywords: Sericite, Nickel Hexacyanoferrate (NiHCF), Cesium, Adsorption, Selectivity

INTRODUCTION

Removal of toxic metals from industrial wastewater has become a most important issue. Especially, the radioactive wastes, appearing from the power nuclear reactors and the application of radionuclides such as medicine, agricultures, industry and research are a major concern to human health if they enter the environment. Since the major nuclear accident at Fukushima, Japan in 2011, 630,000-777,000 TBq (tetrabecquerels) of radioactive nuclides such as Sr, Se, I and Cs were released into water, soil and air [1,2]. Among radionuclides, ^{137}Cs has a long half-life of ($t_{1/2}$=30.17 years) and poses serious environmental threat because of its high solubility and mobility with aqueous media in the subsurface [3]. Furthermore, it can be readily assimilated by terrestrial and living organisms due to its similar chemical characteristics with potassium [4]. Thyroid cancer is well known as one of the dire consequences of ^{137}Cs adsorption via the contaminated food and water [5]. Therefore, the concentration of cesium should be reduced from waste solution, and many studies on the effective and low cost techniques to remove cesium ions in aquatic environment have been recently performed.

The separation/removal of cesium ions from waste solutions generally can be achieved by means of physical-chemical treatment such as coagulation-precipitation, solvent extraction, adsorption, ion exchange and membrane processes [6-8]. Solvent extraction, ion exchange and membrane processes, which have been widely used, are costly and unsuitable for large-scale application [9]. Also, coagulation-precipitation methods generate a large amount of sludge difficult to control [10]. In the case of adsorption, it has several advantages such as high efficiency, simplicity, cost effectiveness and the availability of a wide range of adsorbents [11,12]. Among adsorbents, clay minerals have been most widely used to remove cesium ions in water and wastewater because of superior qualities such as large specific surface area, chemical and mechanical strength, layered structure and high exchange capacity [13,14]. They have also several advantages of their abundance, low cost, and environment-friendly nature over other adsorbents [15]. Various clay minerals such as zeolite, bentonite and montmorillonite have been applied to remove cesium ions in waste solutions [16,17]. We also reported that sericite, which was obtained from a local mining industry in Korea, has good adsorption capacity for cesium ions in aqueous solution [18,19]. However, the study on increasing adsorption capacity of sericite for cesium ions is required to compete commercial ion exchange resin. Also, sericite has no selectivity to cesium ions due to the competitive interactions when cesium ions were mixed with other cations such as sodium, potassium and various metal ions in waste solution [20,21].

Transition metal hexacyanoferrate, especially nickel hexacyanoferrate (NiHCF), is well known as a highly selective agent for cesium adsorption [22]. It has a special cubic structure with a channel diameter of about 3.2 Å, through which only small hydrated ions like cesium ions can permeate while larger hydrated ions like sodium ions get blocked [23]. However, its use in particles is impossible because of the nano particle size of NiHCF; therefore, suitable support materials are necessary. Ding et al. developed the nickel haxacyanoferrate (NiHCF)-walnut shell to remove cesium from aqueous solution and successfully overcame the difficulty of nanoparticle separation [9].

In this study, sericite was used as a support material. To the best

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of our knowledge, until now, there are no studies on the synthesis of NiHCF using sericite to remove cesium ions. Therefore, the aim of this work was to develop novel adsorbent of sericite incorporated into nickel hexacyanoferrate (NiHCF-sericite) to increase adsorption capacity of cesium ions and remove selectively cesium ions only in mixed waste solutions with sodium and other metals. This study also presents adsorption characteristics of NiHCF-sericite for cesium ions under various experimental conditions such as pH, temperature, time, agitation speed and ionic strength.

MATERIALS AND METHODS

Sericite used in this study was obtained from the SG mining industry, which is located in Gangneung province in Korea. Samples were washed several times with deionized water to remove soluble impurities and then completely dried in an oven (JEIO TECH OF-22GW) at 80 °C for 1 day. They were ground and homonized by a blender with steel blades (Hanil Co., Korea) and sieved through No. 8 and 16 size meshes. All of the chemicals were of analytical grade (Sigma Aldrich, U.S.A.) and deionized water obtained from a Milli-Q system (Millipore, Bedford, MA, USA) was used to prepare all the solutions.

The manufacturing processes of incorporation of NiHCF onto the sericite are almost same as the Ding's work [9]. 10 g of sericite was soaked in 100mL of 50% (v/v %) hydrochloric acid for 10 h at 50 °C. The sericite was washed with deionized water until the eluent pH was almost 7 and then dried in an oven at 105 °C overnight. 5 g of sericite through above process was immersed in 20 mL of 0.5 M $NiCl_2 \cdot 6H_2O$ solution and placed in a shaking incubator (JEIO TECH, SI-600R, Korea) for 24 h at 25 °C. And then it was filtered by centrifuge (Gyrozen, Gyro 1236 MG, Korea) and washed with deionized water to remove excess $NiCl_2 \cdot 6H_2O$. The sample of $NiCl_2$ loaded onto sericite was added to 10 mL of 5% (wt%) $K_3[Fe(CN)_6] \cdot 3H_2O$ solution and was reacted in a water bath (JEIO TECH, BS-21, Korea) at 30 °C for 24 h. The NiHCF-sericite was separated by filtration, washed with deionized water and dried at 60 °C. All procedure was repeated three times to verify the incorporation of NiHCF onto the sericite.

To determine properties of the NiHCF-sericite, specific surface area was measured by means of multi-point adsorption data from linear segment of the N_2 adsorption isotherms using Brunauer-Emmett-Teller (BET) theory and chemical composition was analyzed by the powder X-ray diffractometry (Siemens D-5005 diffractometer, U.S.A.).

Scanning electron microscopy (SEM, Hitachi model S-4100, Japan) photograph and energy dispersive X-ray (EDX, Oxford INCA, U.S.A.) spectroscopy were applied to investigate surface condition and component onto NiHCF-sericite for before and after adsorption of cesium.

Stock cesium solution was prepared by dissolving cesium chloride (CsCl, FW: 168.36) in distilled water and used to prepare the adsorbate solution with appropriate dilution. The initial pH of the cesium solution was adjusted by negligible volume of 0.1 M NaOH and HCl solution, and all sorption experiments were not done above pH 8.0 to avoid precipitation for cesium solution. All sorption experiments were conducted under desired experimental condition by batch-type in 100 mL of solution in a shaking incubator (JEIO TECH, SI-600R, Korea). Some amount of NiHCF-sericite and 100 mL of cesium solution was added in a 250 mL Erlenmeyer flask. The suspension was shaken for a fixed time in a shaking incubator (JEIO TECH, SI-600R, Korea) and then centrifuged at 4,000 rpm for 30 min by centrifuge (Gyrozen, Gyro 1236 MG, Korea) to remove suspending NiHCF-sericite. The concentration of cesium ion in supernatant was analyzed by atomic absorption spectroscopy (Perkin-Elmer A Analyst 100/A Analyst 700, U.S.A.). All experiments were performed in triplicate. The adsorption capacity, Q (mg/g) of NiHCF-sericite for cesium ions was obtained from the following Eq. (1).

$$Q=(C_i \times V_i - C_f \times V_f)/W \tag{1}$$

where C_i (mg/L) and C_f (mg/L) are initial and equilibrium cesium concentration in solution, respectively; V_i (L) and V_f (L) are initial and final volume of waste solution; W (g) is the weight of the NiHCF-sericite.

The specific conditions for each adsorption experiment are as follows. The effect of pH on adsorption capacity was conducted in the range of 2.0-7.0, and 0.1 g of NiHCF-sericite was added to 100 mL of cesium solution including 20 mg/L of cesium ions. For the isothermal adsorption experiment, 0.1-1.0 g of adsorbent was added to 100 mL of cesium solution with 1-20 mg/L of cesium ions, respectively, and initial pH of the cesium solution was controlled as 5.0. To investigate the effect of time, each 0.1 g of NiHCF-sericite was loaded to 100 ml of cesium solution with 20 mg/L of cesium ions, and adsorption capacity was measured for 10, 30, 60, 120 and 180 min, respectively. Also, temperature effect on cesium adsorption capacity was observed for the range over 15, 25, 35 and 45 °C, respectively, at pH 5.0. The effect of agitation speed was also investigated for 100, 150, 200, 250, 300 rpm (rotations per minute) under condition of 20 mg/L of cesium concentration in 100 mL of solution at pH 5.0. In addition, effect of ionic strength on adsorption capacity was performed for the 0.01, 0.05, 0.1, 0.5, 1.0 and 2.0 M of NaCl concentration under condition of 20 mg/L of cesium ions in 100 mL of solution at pH 5.0, respectively. Multi-metal adsorption capacity and selectivity experiment was carried out under the condition of 0.15 mM of each metal concentration (cesium, lead, cadmium, nickel, zinc, potassium, calcium and magnesium ions) in 100 mL of solution at initial pH 5.0 of waste solution. Finally, to investigate the regeneration ability for NiHCF-sericite, adsorption-desorption cycles were repeated four times by the means of same adsorbent by batch process. 1.0 M of HNO_3 was used as desorbing agent.

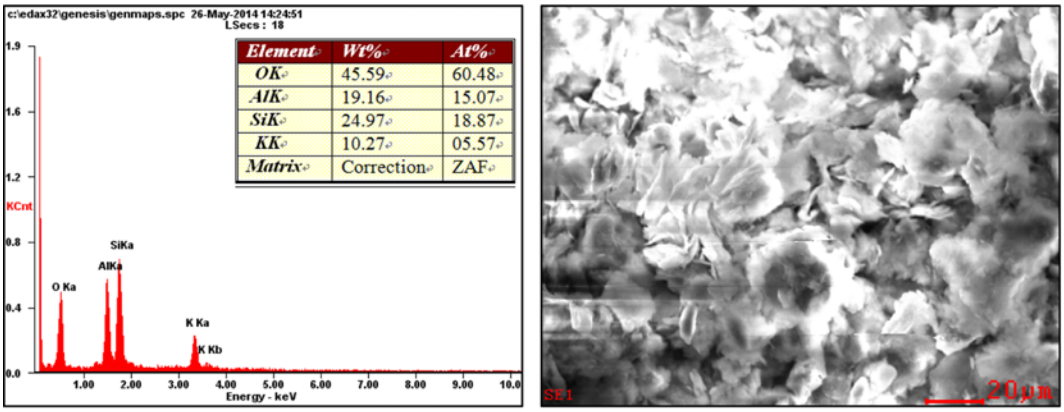
Table 1. Chemical composition of various metal oxides in NiHCF-sericite

Metal oxides	wt% Composition
SiO_2	49.20
Al_2O_3	33.41
K_2O	10.35
NiO	3.79
MgO	1.82
CaO	0.90
Fe_2O_3	0.29
Na_2O	0.03

RESULTS AND DISCUSSION

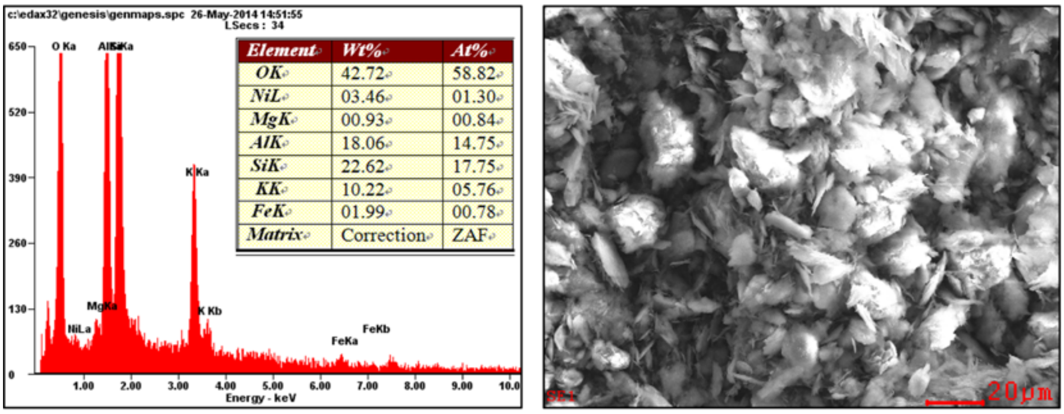
The BET surface area of NiHCF-sericite was measured as the 0.024 m^2/g and its value was almost the same as the natural sericite [18]. The chemical composition of NiHCF-sericite which was determined by X-ray diffractometry is presented in Table 1. It is mainly composed of silica as SiO_2 (49.2%), aluminum as Al_2O_3 (33.4%), potassium as K_2O (10.5%). In addition, nickel as NiO was contained as the 3.79%.

Fig. 1(a) and (b) shows the SEM images and EDX spectrum of sericite for before and after modification with NiHCF. It can be seen that the modified sericite has NiHCF particles attached on the surface of sericite and the 3.46 wt% of Ni incorporated into sericite was confirmed by the EDX spectrum. The electron dense part (more bright), which is thought to be cesium ions, appeared in Fig. 1(c), as compared with (b). To verify the existence of cesium ions, EDX was also employed and the characteristic peak for cesium ions with 0.91 wt% clearly emerged.



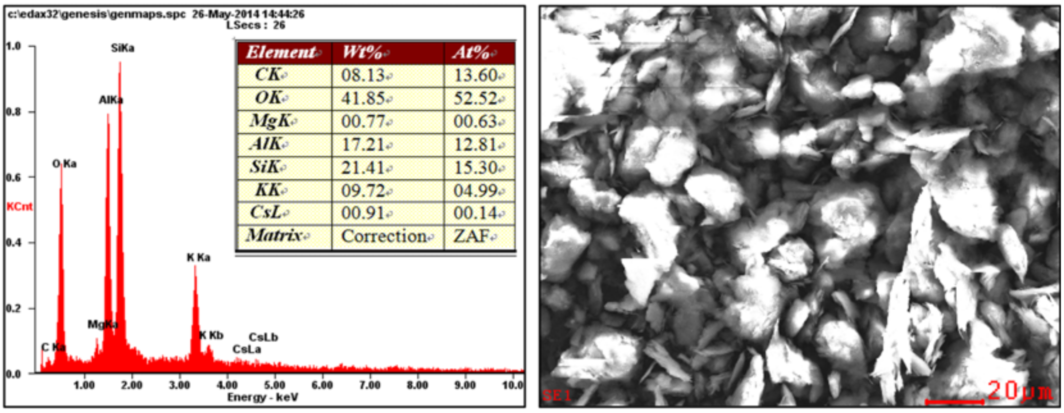
Element	Wt%	At%
OK	45.59	60.48
AlK	19.16	15.07
SiK	24.97	18.87
KK	10.27	05.57
Matrix	Correction	ZAF

(a) Natural sericite



Element	Wt%	At%
OK	42.72	58.82
NiL	03.46	01.30
MgK	00.93	00.84
AlK	18.06	14.75
SiK	22.62	17.75
KK	10.22	05.76
FeK	01.99	00.78
Matrix	Correction	ZAF

(b) NiHCF-sericite



Element	Wt%	At%
CK	08.13	13.60
OK	41.85	52.52
MgK	00.77	00.63
AlK	17.21	12.81
SiK	21.41	15.30
KK	09.72	04.99
CsL	00.91	00.14
Matrix	Correction	ZAF

(c) After cesium adsorption with NiHCF-sericite

Fig. 1. SEM photograph and EDX spectrum.

A comparison study on adsorption capacity of natural sericite and NiHCF-sericite for cesium ions was performed and the result is shown in Fig. 2. The adsorption capacity of NiHCF-sericite (14.8 mg/g-dry mass) was much higher than that of natural sericite (6.7 mg/g-dry mass) at initial pH 5.0 of waste solution. The increase of adsorption capacity can be explained by the existence of NiHCF incorporated onto the surface of sericite. The nickel hexacyanoferrate (NiHCF) has an excellent affinity to cesium ions, as mentioned in introduction.

Fig. 3 shows the effect of pH on the cesium adsorption. A study beyond pH 8.0 was not done because of insoluble cesium-hydroxide-precipitate in the waste solution (data not shown). The cesium adsorption increased as pH value increased from 2 to 5; however, further increase in pH did not improve the adsorption capacity for cesium. Low adsorption capacity of NiHCF-sericite for cesium ions at lower pH was related to the competition of excessive protons for the available sorption sites [24]. In acid conditions, most silanol and aluminol groups are protonated and, hence, they are less available to retain cesium ions. Since maximum adsorption capacity for cesium ions was obtained at initial pH 5.0 of waste solution, therefore, all further sorption experiments were performed at pH 5.0.

Adsorption isotherm is the main requirement to investigate the adsorption mechanism for its ability in describing the interactive behavior between the adsorbate and adsorbent. Langmuir isotherm is based on the assumption of a homogeneous surface and constant sorption potential. Furthermore, it assumes that the adsorption occurs at specific homogeneous adsorption sites of the adsorbent [25]. The Freundlich isotherm is also widely used and it is an empirical equation expressing adsorption at multilayer and energetically heterogeneous surface [26]. Fig. 4 and 5 show the application of nonlinear Langmuir, Freundlich isotherms to the cesium adsorption on NiHCF-sericite at initial pH 5.0 of waste solution. Lang-

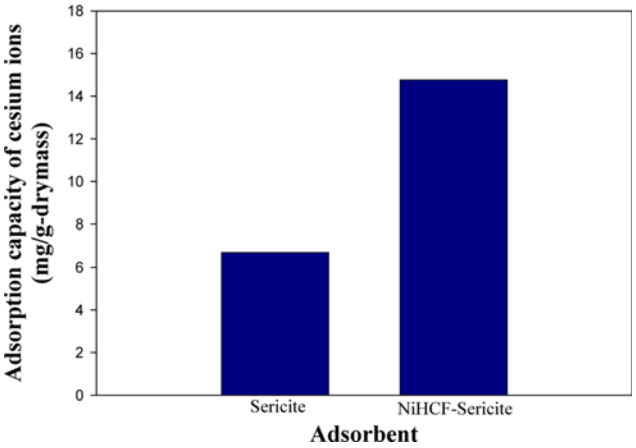


Fig. 2. Copmarison of adsorption capacity for cesium ions of sericite and NiHCF-sericite from aqueous solution (Initial concentration: 20 mg/L, adsorbent concentration: 1.0 g/L, initial pH of cesium solution: 5.0).

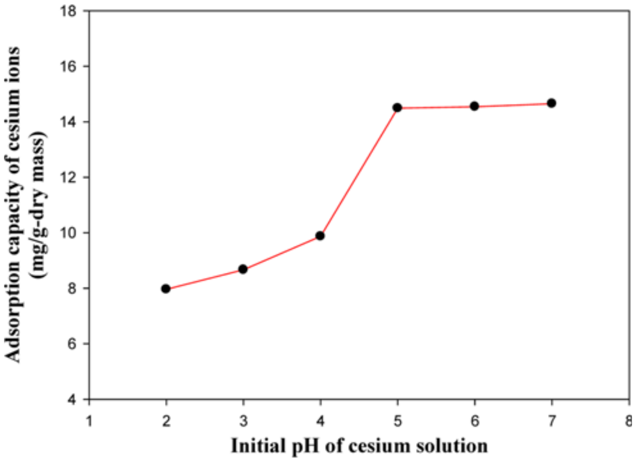


Fig. 3. Effect of initial pH of cesium solution on adsorption capacity of cesium ions (Initial concentration of cesium ions: 20 mg/L, NiHCF-sericite concentration: 1.0 g/L, working volume: 100 mL).

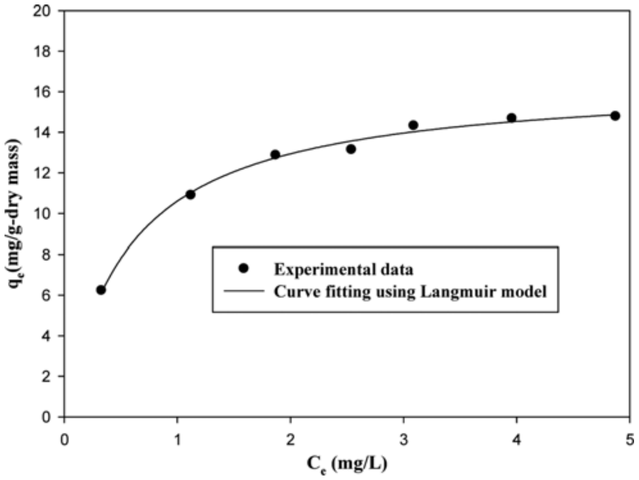


Fig. 4. Isothermal adsorption curve for cesium ions of NiHCF-sericite using iangmuir model (Initial pH of cesium solution: 5.0, working volume: 100 mL).

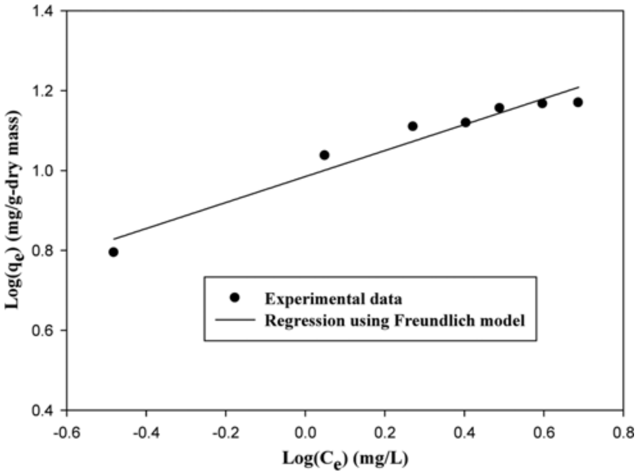


Fig. 5. Isothermal adsorption for sesium ions of NiHCF-sericite using Freundlich model (Initial pH of cesium solution 5.0, working volume: 100 mL).

Table 2. Equilibrium isotherm model parameters of NiHCF-sericite for cesium ions

Langmuir isotherm model			Freundlich isotherm model		
q_m (mg/g)	K_L (L/mg)	r^2	1/n	K_f (L/g)	r^2
16.583	1.774	0.994	0.325	9.658	0.947

muir and Freundlich model equations are generally expressed as follows, respectively:

$$1/q_e=1/q_m+(1/q_mK_L)1/C_e \quad (2)$$

$$\text{Log } q_e=\log K_f+(1/n)\log C_e \quad (3)$$

where q_e, q_m, K_L, K_f and 1/n are adsorption capacity (mg/g) corresponding to metal ion concentration (C_e), maximum adsorption capacity of metal ions (mg/g), Langmuir adsorption constant (L/mg), K_f, is a constant related to the adsorption capacity, and 1/n is an empirical parameter related to the adsorption intensity which varies with the heterogeneity of material [27], respectively. The obtained model parameters and correlation coefficient (r^2) of different isotherms for cesium ions are summarized in Table 2. The higher correlation coefficients show that Langmuir isotherm fits the adsorption data better than Freundlich model. It means that the adsorption of cesium ions on NiHCF-sericite is probably a monolayer adsorption process. The maximum adsorption capacity of NiHCF-sericite for cesium ions was evaluated as 16.583 mg/g. Table 3 shows the recent works for cesium adsorbents considering maximum adsorption capacity. Although a direct comparison for the adsorption capacity between NiHCF-sericite and other adsorbents, as presented in Table 2, was difficult due to the varying experimental condition applied in those studies, NiHCF-sericite is a promising adsorbent for adsorption of cesium ions in waste solution.

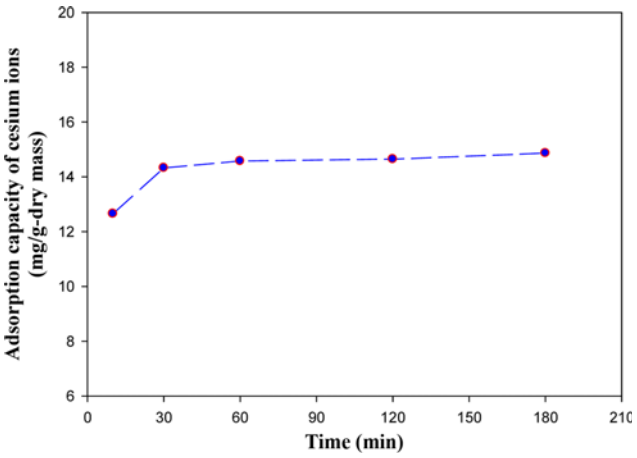


Fig. 6. Adsorption capacity for cesium ions of NiHCF-sericite respect to time (Initial concentration of cesium ion: 20 mg/L, sericite concentration: 1.0 g/L, initial pH of cesium solution: 5.0).

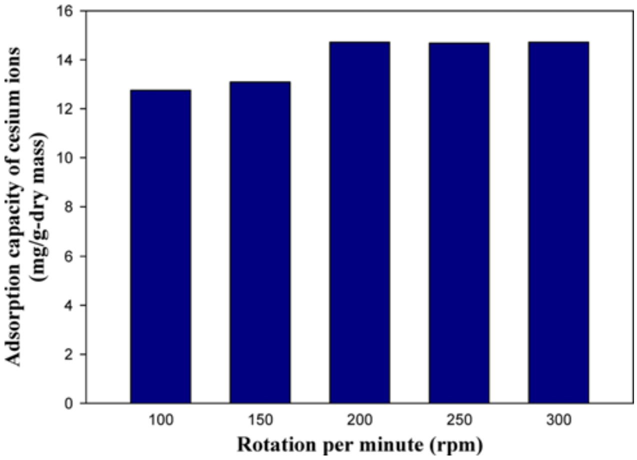


Fig. 7. Effect of rotation per minute (rpm) on adsorption capacity of cesium ions using NiHCF (Initial concentration of cesium ions: 20 mg/L, adsorbent concentration: 1.0 g/L, initial pH of cesium solution: 5.0, agitation time: 1 hr).

Adsorption kinetics, which demonstrates the adsorption rate of adsorbate, is one of the most important characteristics representing the adsorption efficiency of adsorbents, especially, in batch process. Effect of agitation time and rate on adsorption capacity of cesium ions are shown in Fig. 6 and 7, respectively. Most of the adsorption of cesium ions was completed within 30 min, as shown in Fig. 6. It is probably due to the abundant active sites on the clay surface, and the direct electrostatic and complexation reaction on the outside surface are also important factors for the rapid adsorption [32]. Tsi et al. and Bayulken et al. reported similar behaviors about cesium adsorption [12,33]. The adsorption capacity of cesium ions increased with agitation rate and obtained maximum value at 200 rpm, as shown in Fig. 7. The increase certainly highlights the limitation of the mass transfer through the solution and in the boundary layer during the adsorption process when an agitation rate of 200 rpm or lower was selected. However, further increase does not improve the adsorption capacity for cesium ions. Therefore, the optimum agitation rate for adsorption of cesium ions was chosen

Table 3. Comparison of maximum adsorption capacity of cesium ion from waste solutions using various adsorbents

Adsorbent	Maximum adsorption capacity (mg/g)	Reference
NiHCF-sericite	16.583	This study
Nickel hexacyanoferrate-walnut shell	4.94	[9]
Oxidized multiwall carbon nanotubes	12.75	[28]
Magnetite PVA-alginate beads	28.32	[29]
Inorganic mesoporous silica	27.40	[30]
Crown ether immobilized new class conjugate	77.70	[31]

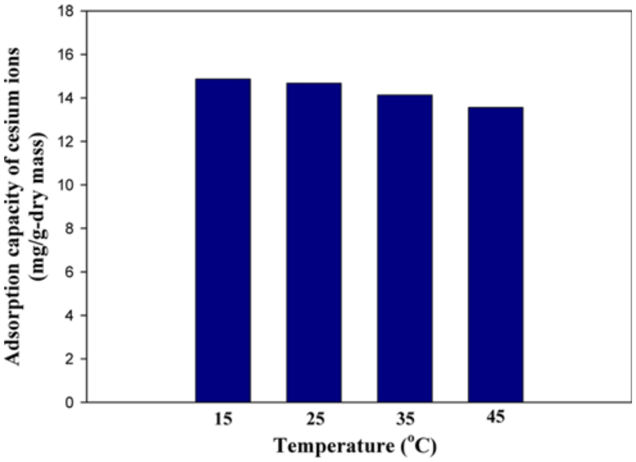


Fig. 8. Effect of temperature on adsorption capacity of cesium ions using NiHCF (Initial concentration of cesium ions: 20 mg/L, adsorbent concentration: 1.0 g/L, initial pH of cesium solution: 5.0).

as 200 rpm. A similar result was reported by Cuevas et al [35].

Fig. 8 shows the effect of temperature on adsorption capacity of cesium ions. The adsorption capacity decreased as temperature increased. It means that cesium adsorption by means of NiHCF-sericite was determined as the exothermic process and the result is similar to Zhang's report [34]. Therefore, 25 °C was chosen as the optimal temperature because of ease of operation although the highest adsorption capacity was obtained at 15 °C.

The adsorption process was also affected by ionic strength in the waste solution. The effect of ionic strength (i.e., the background electrolyte concentration) on adsorption of metal ions can be explained in two ways: (a) by affecting the interfacial potential and therefore the activity of electrolyte ions and adsorption; (b) by affecting the competition of the electrolyte ions and adsorbing anions for available sorption sites. That is, at high ionic strength, adsorption sites are surrounded by counter ions in which they partially lose their charge, and this weakens the binding force by an electrostatic interaction. Thus, this effect may give inactivation of ion exchange mechanism between metal ions and adsorbent [36]. The adsorption capacity of cesium ions decreased as NaCl concentration increased, as shown in Fig. 9. However, the ionic strength only had a slight effect on adsorption capacity for cesium ions below 0.1 M of NaCl concentration, while adsorption capacity greatly decreased above 0.5 M of NaCl concentration. It is probably due to the adsorption mechanism between NiHCF-sericite and cesium ions, which is thought to be forming of inner sphere complexes by covalent and ionic binding which are well known that ionic strength effect is negligible [37]. It means that the NiHCF-sericite can be sufficiently applied to remove cesium ions even though sodium ions with around 0.1 M concentration are coexisted with cesium ions.

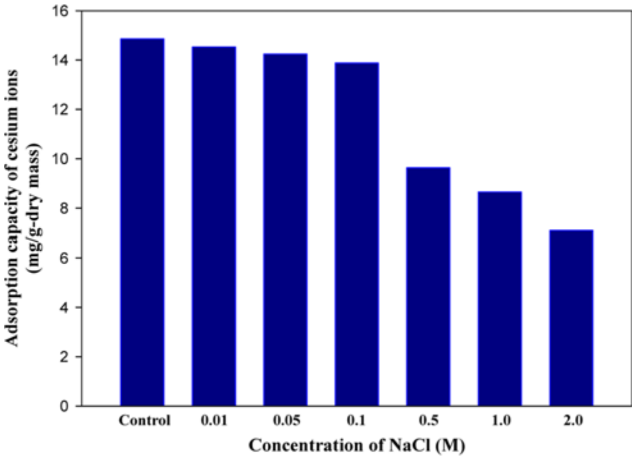


Fig. 9. Effect of ionic strength on the cesium adsorption capacity of NiHCF (Initial concentration of cesium ions: 20 mg/L, initial pH of cesium solution: 5.0, NiHCF concentration: 1.0 g/L).

Most water and wastewater are mixed with various metals such as lead, copper, cadmium, nickel, zinc, potassium, calcium and magnesium, and it is very important to selectively remove target metal only. Thus, a study on the selectivity to various metals using NiHCF-sericite was performed and the results are shown in Table 4. In the case of cesium ions, the adsorption capacity was the 0.107 mmol/g and the value was almost the same with that of cesium only. It shows adsorption capacity of NiHCF-sericite for cesium ions was not greatly affected by other metals. It can be explained as a function of NiHCF which has excellent selectivity to cesium ions, as mentioned above. The selectivity order of various metals for NiHCF-sericite is as follows:

Cesium>>Lead>Cadmium>Nickel>Zinc
>Potassium, Calcium, Magnesium

It is essential to describe regeneration aspects of the process in

Table 4. Adsorption capacities of various metal ions in mixed metal solution by NiHCF-sericite (Initial metal concentration: 0.15 mM, Initial pH of mixed solution: 5.0, NiHCF concentration: 1.0 g/L, Adsorption capacity of cesium only: 0.111 mmol/g)

Metalic ion species	Adsorption capacity (mmol/g)
Cesium	0.107
Lead	0.013
Cadmium	0.008
Zinc	0.004
Nickel	0.002
Potassium	0.001
Calcium	0.001
Magnesium	0.001

Table 5. Regeneration ability of NiHCF-sericite for cesium ions (Initial concentration of cesium ions: 20 mg/L, Influent pH of cesium solution: 5.0, Desorbing agent: 1.0 M of HNO_3)

Cycle number	Adsorption capacity (mg/g)
1	13.9
2	13.2
3	10.6
4	7.3

order to improve its cost-effectiveness by recycling the adsorbent for reuse in multiple cycles. As shown in Table 5, adsorption capacity of cesium ions can be maintained as 13.2 mg/g until two cycles; however, the values quickly decreased due to incomplete desorption of NiHCF-sericite for cesium ions at the 3 cycle. The result shows that NiHCF-sericite has regeneration ability for cesium ions, to some extent. Therefore, it could be concluded that NiHCF-sericite with excellent selectivity to cesium ions and good regeneration capacity can be sufficiently applied to the removal of cesium ion from waste solution.

CONCLUSION

NiHCF-sericite as a novel adsorbent was developed and used to remove cesium ions from waste solution. The highest adsorption capacity for cesium ion was achieved at initial pH 5.0 of waste solution, and the maximum adsorption capacity of NiHCF-sericite was estimated as the 16.583 mg/g by Langmuir isotherm model. The higher correlation coefficients show that Langmuir isotherm fits the adsorption data better than Freundlich model. The adsorption process of NiHCF-sericite for cesium was exothermic reaction and was almost completed within 30 min. Also, the adsorption capacity of cesium ions was not greatly affected by ionic strength below 0.1 M NaCl concentration and other metals in mixed waste solution. In addition, sericite incorporated NiHCF overcame the difficulty of separation of NiHCF nano-particles from solution. Therefore, it was concluded that adsorption process using NiHCF-sericite can be sufficiently applied to the removal system of cesium ions from waste solution and the conventional treatment process using commercial ion-exchange resin can be replaced with novel process using NiHCF-sericite with high selectivity and mechanical strength.

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