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Evaluation of a cesium adsorbent grafted with ammonium 12-molybdophosphate

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HIGHLIGHTS

- A Cs adsorbent was developed using radiation-induced graft polymerization.
- Ammonium 12-molybdophosphate was uniformly loaded onto the Cs adsorbent.
- A high amount of Cs was adsorbed in a wide pH range.
- A high selectivity for Cs in the presence of coexistent ions was confirmed.
- An inspection for using the Cs adsorbent to drinking water was successfully passed.

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ABSTRACT

A fibrous cesium (Cs) adsorbent was developed using radiation-induced graft polymerization with a cross-linked structure containing a highly stable adsorption ligand. The ligand, ammonium 12-molybdophosphate (AMP), was successfully introduced onto the fibrous polyethylene trunk material. The resulting Cs adsorbent contained 36% nonwoven fabric polyethylene (NFPE), 1% AMP, 2% triallyl isocyanurate (TAIC) and 61% glycidyl methacrylate (GMA). The adsorbent's Cs adsorption capacity was evaluated using batch and column tests. It was determined that the adsorbent could be used in a wide pH range. The amount of desorbed molybdenum, which can be used as an estimate for AMP stability on the Cs adsorbent, was minimized at the standard drinking water pH range of 5.8–8.6. Based from the inspection on the adherence of these results to the requirements set forth by the Food Sanitation Act by a third party organization, it can be concluded that the developed Cs adsorbent can be safely utilized for drinking water.

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1. Introduction

In March 2011, a considerable amount of radioactive cesium (Cs) was released from the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Station operated by Tokyo Electric Power Co., Inc. due to the confluence of an earthquake, a tsunami and a hydrogen explosion. Decontamination activities have been implemented as part of the restoration process to decrease the air radiation dose rate. However, disaster areas that suffered extensive damage have recently issued evacuation orders three years after these incidents occurred. Another extremely vital task for the restoration process is securing safe water sources. The majority of

the disaster areas are located in mountainous regions, where the stream and/or the well water are the only main sources of sustenance. Accordingly, there is a strong possibility that the water sources from these natural environments are contaminated with Cs. In fact, an investigation of well waters in Fukushima by the Ministry of the Environment in Japan, indicated that radioactive Cs was present in 8 out of 1,200 samples (Ministry of the Environment in Japan, 2014). However, the amounts of detected radioactive Cs in all the samples were lower than 10 Bq/L, the standard for safe drinking water set by the Ministry of Health, Labour and Welfare in Japan (Ministry of Health, Labour and Welfare in Japan, 2012). It is essential that the people who are either residing or returning to the disaster areas can use their water sources with confidence despite the low amounts of Cs detected within the 8 samples.

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In the past, various types of Cs adsorbents with different chemical and physical properties have been investigated. Such adsorbents include zeolite (Mimura and Yamagishi, 2012; Hossein et al., 2013; Vereshchagina et al., 2013; Gu et al., 2000), ferrocyanide (Okamura et al., 2014), crown ether (Awual et al., 2014), vermiculite (Hang et al., 2014), hydrous manganese (Nilchi et al., 2012) and ammonium 12-molybdophosphate (AMP) (Mimura and Yamagishi, 2012; Todd et al., 2002; Iwanade et al., 2012). To satisfy the requirements for drinking water, the adsorbent has to abide by certain guidelines established by the Food Sanitation Act (Ministry of Health, Labour and Welfare in Japan 1959) or the Water Supply Act (Ministry of Health, Labour and Welfare in Japan, Water Supply Act, 2003) in Japan. One of the requirements states that the materials in the adsorbent cannot include and elute toxic materials such as heavy metals. Furthermore, for practical applications, these materials must have high selectivity toward Cs amongst other coexisting ions. Because AMP has a high affinity for Cs, the compound was introduced into silica gel (Mimura and Yamagishi, 2012), an inorganic ion-exchange composite (Mimura and Yamagishi, 2012) and polyacrylonitrile (Nilchi et al., 2012). Although the performance of these Cs adsorbents is acceptable, those investigations were not particularly aimed at the adsorbents' applications in drinking water. AMP was also introduced into an acrylonitrile-grafted chain (Iwanade et al., 2012). However, high quantity of AMP was eluted from the adsorbent during the Cs adsorption process.

In this study, a Cs adsorbent was particularly developed for its application to drinking water. The adsorbent was synthesized using radiation-induced graft polymerization of glycidyl methacrylate (GMA) with AMP onto a nonwoven fabric polyethylene (NFPE). The cross-linked structure was introduced into the adsorbent to attain a high stability of the loaded AMP. The Cs adsorption performances were evaluated using batch and column tests.

2. Experiment

2.1. Materials and chemicals

NFPE was purchased from Kurashiki Textile Manufacturing Co., Ltd., and it was used as the trunk material of the Cs adsorbent. AMP was purchased from Nippon Inorganic Colour and Chemical Co. Ltd.; GMA was purchased from Mitsubishi Gas Chemical Company, INC.; dimethyl sulfoxide (DMSO) was purchased from Kishida Chemical Co., Ltd.; polyoxethylene sorbitan monooleate (Tween 80), which was used as the surfactant, was purchased from Kanto Chemical Co., Ltd.; and triallyl isocyanurate (TAIC), which was used as the crosslinking agent, was purchased from Nippon Kasei Chemical Co., Ltd. for the synthesis of the grafted Cs adsorbent. Nitric acid was purchased from Kanto Chemical Co., Ltd. and methanol (MeOH) was purchased from Taiyo Chemical Industry Co., Ltd. for washing the grafted NFPE.

A standard solution (1000 mg/L) containing Cs, potassium (K), calcium (Ca), magnesium (Mg) and sodium (Na) ions was used for the adsorption tests and quantitative analyses. Furthermore, ultrapure nitric acid and lithium hydroxide purchased from Kanto Chemical Co., Ltd were used to adjust the pH. The distilled water that was used for washing after reactions and the ultrapure water that was used for diluting solutions were both obtained with the Milli-Q deionization system from Merck Ltd. Tap water in Takasaki, Gunma, Japan was used for diluting solutions in the adsorption tests.

2.2. Synthesis of Cs adsorbent

2.2.1. Preparation of monomer solution

The monomer solution was prepared by mixing Tween 80, GMA, TAIC and AMP in a DMSO solvent. The weight percentages of Tween 80, GMA, TAIC, AMP and DMSO were 0.8, 9.8, 0.9, 0.5 and 88.0 wt%, respectively. After mixing the reagents, the monomer mixture was homogenized for 5 min until the reagent was dissolved and/or dispersed. The monomer solution was then deoxygenated by bubbling nitrogen gas through the solution.

2.2.2. Radiation-induced graft polymerization

The NFPE was irradiated to 50 kGy with an electron beam in nitrogen atmosphere and at dry ice temperature. The irradiated NFPE was immediately immersed in the monomer mixture, and grafting was performed for 2 h at 313 K. The grafted NFPE was washed with distilled water, 0.01 mol/L nitric acid and MeOH in succession to remove the residual monomer solution at room temperature. The NFPE was then dried in a vacuum oven at 303 K and then further washed with distilled water at 313 K to reduce the amount of the desorbed component from the grafted NFPE. The degree of grafting (Dg) was calculated using the changes in the weight after the grafting process with the following equation:

$$Dg[\%] = 100 \times ((W_1 - W_0)/W_0) \quad (1)$$

where W_0 and W_1 are the weight of the NFPE before and after the grafting process, respectively. The resultant grafted NFPE comprising AMP, TAIC and GMA are referred to as the Cs adsorbent (Fig. 1).

2.3. AMP distribution on Cs adsorbent

High resolution images of the NFPE and Cs adsorbent fibers' surfaces were taken with a scanning electron microscope (SEM, Hitachi High-Technologies Co., Type N and SU3500). The Mo elemental mapping was performed using a SEM-energy dispersive X-ray spectrometer (EDX, Horiba Ltd., X-max N50) to evaluate the distribution of AMP on the grafted NFPE.

2.4. Composition of Cs adsorbent

The composition of the Cs adsorbent was investigated using several experiments. The amount of NFPE (R_N) in the Cs adsorbent was first calculated using the weight gain before and after the grafting process with the following equation:

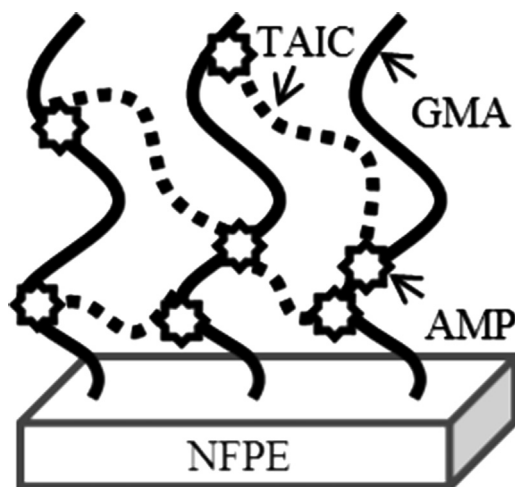


Fig. 1. Schematic of a grafted Cs adsorbent.

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