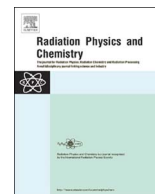




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Development of a water purifier for radioactive cesium removal from contaminated natural water by radiation-induced graft polymerization

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ABSTRACT

Six years after the Fukushima-nuclear accident, the dissolved radioactive cesium (Cs) is now hardly detected in environmental natural waters. These natural waters are directly used as source of drinking and domestic waters in disaster-stricken areas in Fukushima. However, the possibility that some radioactive Cs adsorbed on soil or leaves will contaminate these natural waters during heavy rains or typhoon is always present. In order for the returning residents to live with peace of mind, it is important to demonstrate the safety of the domestic waters that they will use for their daily life. For this purpose, we have synthesized a material for selective removal of radioactive Cs by introducing ammonium 12-molybdophosphate (AMP) onto polyethylene nonwoven fabric through radiation-induced emulsion graft polymerization technique. Water purifiers filled with the grafted Cs adsorbent were installed in selected houses in Fukushima. The capability of the grafted adsorbent to remove Cs from domestic waters was evaluated for a whole year. The results showed that the tap water filtered through the developed water purifier contained no radioactive Cs, signifying the very effective adsorption performance of the developed grafted adsorbent. From several demonstrations, we have commercialized the water purifier named “KranCsair®”.

Furthermore, we have also developed a method for the mass production of the grafted nonwoven fabric. Using a 30 L grafting reactor, it was possible to produce the grafted nonwoven fabric with a suitable range of degree of grafting. When an irradiated roll of nonwoven trunk fabric with a length of 10 m and a width of 30 cm was set in the reactor filled with glycidyl methacrylate (GMA), AMP, Tween 80 monomer emulsion solution at 40 °C for 1 h, the difference of Dgs in the length and the width on roll of fabrics was negligible.

1. Introduction

The big tsunami caused by the severe earthquake in East Japan damaged the electricity supply and stopped the circulation of the cooling water of the nuclear reactor in the Fukushima Daiichi Nuclear Power Station. As a result of the melt-down of the nuclear reactor, radioactive materials were dispersed in the surroundings, including numerous water bodies. In order to remove the radioactive materials, it is important to focus on reducing volume of the radioactive wastes. Various decontamination methods have been proposed so far, and many types of adsorbents have been developed such as zeolite (Rajec and Domianova, 2008), Prussian blue (Fekry et al., 2003), potassium ferrocyanide (Haas, 1993), and ammonium 12-molybdophosphate (Todd et al., 2002; Iwanade et al., 2012). However, most of the synthesized

adsorbents were for decontaminating insoluble radioactive materials, i.e. none of these adsorbent materials is applicable in treatment of drinking water. In mountainous regions affected by the nuclear disaster, there are many areas that do not utilize water purification plants. Almost 90% of the residents in these areas used well and streaming waters for their daily life.

Currently, it is known that most of radioactive cesium (Cs) are immobilized in clay minerals, etc. Mukai et al. (2014, 2016). In the disaster-stricken areas in Fukushima, however, water sources were drawn from highlands where decontamination has not been conducted, and radioactive Cs were likely to be mixed into the water source during heavy rain and snowfall. Therefore, it is an important task to develop an adsorbent for Cs removal to ensure the safety of drinking water in any case. In order to apply the developed materials in treatment of drinking

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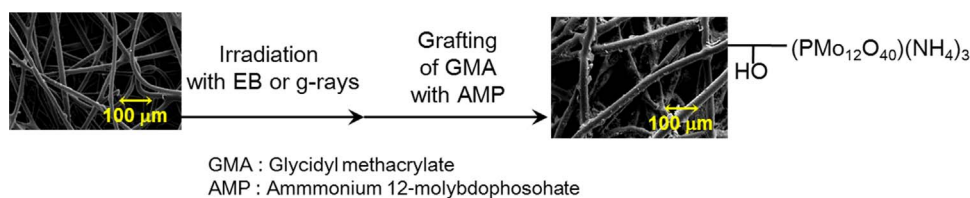


Fig. 1. Preparation scheme of a fibrous grafted adsorbent for Cs removal.

water, it was required that adsorption ligands do not release from the material. Almost all of the materials stated above have adsorption ligands that were desorbed during water treatment. Also, majority of these adsorbents are of granular resin type that was prepared by the copolymerization of styrene and divinylbenzene (Wachinski and Etzel, 1997). Such adsorbents were typically used at a flow rate less than 10 h^{-1} of space velocity which was normalized by dividing the flow rate of the solution by the volume of adsorbent packed in the column. If the synthesized adsorbent is to be used as a filter in the developed water purifier, it is necessary that it can treat the water at a flow rate similar to the normal operation of a faucet.

To address the issues of ligand stability and water flow rate limitation, a fibrous adsorbent was synthesized by radiation-induced emulsion graft polymerization which could introduce a functional group having superior affinity for Cs ions. In our previous report, we have shown that the grafted Cs-adsorbent could work effectively for the dissolve Cs ions without any desorption of ligands (Iwanade et al., 2012; Shibata et al., 2016) and that the fibrous nature of the grafted materials could remove various metal ions even at high flow rates, e.g. passing with SV of 500 h^{-1} or more in the column treatment (Seko et al., 2004; Takeda et al., 2010; Sekine et al., 2010). An additional advantage of this polymeric grafted adsorbent, besides its capability to remove radioactive Cs, is the simplicity of its application because it directly adsorbed the toxic components. That is, sludge, which required a secondary treatment process, was not produced compared with the coagulation method using zeolite and polymeric flocculants.

In the present study, a fibrous polymer adsorbent for the selective removal of radioactive Cs from contaminated waters have been developed by introducing ammonium 12-molybdophosphate (AMP) onto polyethylene nonwoven fabric through radiation-induced emulsion graft polymerization technique. To evaluate the efficiency of the AMP grafted fibrous adsorbent in removing Cs from contaminated waters, we conducted conventional batch and column experiments using real well water samples. Also, the performance of the water purifier containing the developed Cs-adsorbent was evaluated and monitored for one whole year at selected Fukushima households.

2. Experiment

2.1. Chemicals and materials

Polyethylene based nonwoven fabric (NF), with core-in-sheath shape and was composed of polyethylene coated polypropylene, was kindly provided by Kurashiki Seni Kako Co. Okayama Japan and was used for as a trunk material of the radioactive cesium adsorbent (Cs-adsorbent). Glycidyl methacrylate (GMA) from Mitsubishi Gas Chemical Company, Inc., polyoxethylene sorbitan monooleate (Tween 80) and dimethyl sulfoxide (DMSO) from Kanto Chemical Co., Ltd. were used in the radiation-induced graft polymerization and chemical modification steps. Distilled water, for washing after the grafting and modification reactions, and ultrapure water, for diluting solutions, were obtained from a Millie-Q system from Merck Ltd. Methanol (MeOH) purchased from Taiyo Chemical Industry Co., Ltd. was used as washing solvent for the grafted NF. Ammonium 12-molybdophosphate (AMP) was purchased from Nippon Inorganic Color and Chemical Co., Ltd. In order to uniformly impart AMP on the grafted polymer chain, triallyl isocyanurate (TAIC, Nippon Kasei Chemical Co., Ltd.) was used as a

crosslinking agent.

2.2. Emulsified monomer solution

In order to uniformly disperse AMP, an inorganic substance, in DMSO solvent, a combination of emulsion graft polymerization, using a surfactant, and an immobilization method, using a crosslinking agent, was applied. The grafting approach used to synthesize the Cs-adsorbent is shown in Fig. 1. The radiation-induced emulsion graft polymerization method was implemented as previously reported (Seko et al., 2007, 2010). The emulsified monomer solution was prepared with the following composition: 9.8 (w/v)% of GMA, 0.5 (w/v)% of AMP, 0.9 (w/v)% of TAIC, 0.8 (w/v)% of Tween80 in 88 (w/v)% of DMSO as a grafting solvent. All of these substances were mixed and then stirred for 10 min with a homogenizer (Yamato Scientific Co., Ltd.) to make them homogeneous.

2.3. Mass production by pre-irradiation grafting with γ -irradiation

Because a large amount of Cs-adsorbent was necessary for field works to evaluate its performance to remove the Cs spread over the environment, we had to develop a mass production method for Cs-adsorbent synthesis. The prepared monomer solution was transferred to a 30 L reaction tank for graft polymerization. The monomer was deoxygenated using nitrogen gas bubbling to avoid contact between the created radicals and oxygen. The trunk nonwoven fabric (NF) of the Cs-adsorbent, which was cut into a roll shape with a width of 30 cm and a length of 10 m, was irradiated to 160 kGy with γ ray at a dose rate of 10 kGy/h. The irradiation was carried out under dry ice temperature. In order to reduce unevenness in absorbed dose, the NF was rotated by 180° after receiving half of the target absorbed dose, i.e. 80 kGy. Furthermore, a mesh-like net made of polyethylene was alternately wound up in a rolled NF in order to increase its contact efficiency with monomer solution during graft polymerization. To ascertain the distribution of degree of grafting in the total roll length, a small piece of NF was placed at equal intervals in the roll (Fig. 2). It was used as an index of the degree of grafting for the rolled NF. The degree of grafting (DG) of small pieces was calculated from the weight increment before and after grafting as follows:

$$\text{Degree of Grafting (DG) (\%)} = 100(W_1 - W_0)/W_0 \quad (1)$$

where, W_0 and W_1 are the weights of trunk NF and grafted NF, respectively. Grafting was performed at 40°C for 1 h. The resulting grafted NF was washed with DMSO and water to remove non-reacted chemicals, and then dried at 40°C .

2.4. Removal tests of radioactive Cs

The well water samples, collected from areas where radioactive Cs was detected, were used to evaluate the developed Cs-adsorbent for drinking water. First, the insoluble Cs in well water was removed by filtration through $0.45 \mu\text{m}$ and $0.1 \mu\text{m}$ membrane filters. The filtrate was used in the adsorption tests. The soluble radioactive Cs concentration, which is the total number of Cs-134 and Cs-137, was measured by a germanium semiconductor detector (SEICO EG & G CO., LTD., Japan). Cs concentration was 56 Bq/L after filtration. In the batch removal test, a 0.18 g Cs adsorbent, with 35 mm diameter and about

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