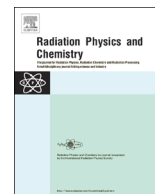




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## Cesium removal in freshwater using potassium cobalt hexacyanoferrate-impregnated fibers

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## HIGHLIGHTS

- Dimethylaminoethyl methacrylate was grafted onto a 6-nylon fiber.
- Potassium cobalt hexacyanoferrate compounds were precipitated onto the grafted fibers.
- With increasing KCl concentration, the rate of cesium removal from cesium chloride solution increased.
- To achieve the practical use, we established the manufacturing equipment on a scale of 100 kg.

## ARTICLE INFO

## Article history:

Received 16 November 2012

Accepted 2 April 2013

Available online 21 April 2013

## Keywords:

Potassium cobalt hexacyanoferrate

Impregnation

6-Nylon

Adsorbents

Cesium removal

Radiation-induced graft-polymerization

## ABSTRACT

Potassium cobalt hexacyanoferrate compounds (KCo-HCFs) were impregnated onto a 6-nylon fiber by radiation-induced graft polymerization and subsequent chemical modifications. First, dimethylaminoethyl methacrylate was graft-polymerized onto the nylon fiber. Second, hexacyanoferrate ions were bound to graft chains via an anion-exchange interaction. Third, KCo-HCFs were formed on the nylon fiber via the precipitation reaction of hexacyanoferrate ions with cobalt ions in the presence of potassium chloride. The resulting KCo-HCF-impregnated fiber had an impregnation percentage of the fiber for KCo-HCFs of 7%. The cesium concentration in 10 ppm cesium chloride solution with the immersion of this fiber decreased to 0.6 ppm within 60 min at a ratio of liquid volume (10 mL) to fiber mass (0.1 g). The fiber was fabricated into a braid with a length of 100 cm and a diameter of 8 cm for practical use at sites contaminated with cesium.

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## 1. Introduction

Among the radioisotopes released from the Fukushima Daiichi nuclear power plant, which was seriously damaged by a magnitude-9.0 earthquake and subsequent tsunami, cesium-137 has spread over a relatively extensive area. Radioactive cesium binds to soil particles or dissolves into surface waters. Cesium-137 with a long half-life of 30 years should be removed and safely stored.

Potassium metal hexacyanoferrates (Barton et al., 1958; Lehto et al., 1992) and zeolites have been reported to be selective inorganic adsorbents for cesium removal. Potassium hexacyanoferrate solution is mixed with a metal solution to form fine particles of potassium metal hexacyanoferrate with diameters of less than 100 nm, which are not suitable for packed beds. Watari and coworkers (1965, 1968), and Tanihara (1996, 1998) impregnated

potassium metal hexacyanoferrates into macroporous anion-exchange resins. Moreover, Mimura et al. (1999a, 1999b) impregnated potassium nickel hexacyanoferrate onto silica gel and zeolite. However, these adsorbents were designed for the treatment of radioactive waste. To improve the rate of adsorption of cesium ions at extremely low concentrations onto adsorbents, adsorbents in fiber form are preferable to the adsorbents in bead form (Fujiwara, 2007; Ikeda et al., 2011).

We succeeded in preparing a potassium cobalt hexacyanoferrate compound-impregnated fiber capable of removing cesium ions from seawater by radiation-induced graft polymerization and subsequent chemical modifications (Ishihara et al., 2011). The fiber is regarded as a hybrid adsorbent consisting of inorganic compounds (potassium cobalt hexacyanoferrate compounds) and an organic support (dimethylaminoethyl methacrylate-grafted nylon fiber). Therefore, the fiber can be molded into various assemblies depending on the conditions of the site contaminated with cesium. However, a high affinity for cesium ions of the fiber prepared in our previous study (Ishihara et al., 2011) was applicable only to cesium removal from seawater. To apply the hybrid

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fiber to cesium removal from surface waters with a much lower ionic strength than seawater, the adsorption capability of the hybrid fiber for cesium ions should be enhanced.

The objectives of this study were twofold: (1) to confer higher affinity for cesium ions in freshwater to the nylon fiber by improving the impregnation conditions of potassium cobalt hexacyanoferrate compounds and (2) to fabricate a braid of fibers for practical use in agricultural and industrial waters.

## 2. Materials and methods

### 2.1. Materials

A 6-nylon fiber with a diameter of 40  $\mu\text{m}$  was used as the trunk polymer for grafting. Dimethylaminoethyl methacrylate (DMAEMA,  $\text{CH}_2=\text{C}(\text{CH}_3)\text{COOC}_2\text{H}_5\text{N}(\text{CH}_3)_2$ ) was purchased from Tokyo Chemical Industry Co., Ltd., and used without further purification. Potassium hexacyanoferrate and cobalt chloride were purchased from Wako. The other chemicals used were of analytical grade or higher. Cesium chloride was dissolved in demineralized water to yield a concentration of 10 ppm or mg-Cs/L.

### 2.2. Impregnation of potassium cobalt hexacyanoferrate compounds

The impregnation of potassium cobalt hexacyanoferrate compounds (KCo-HCFe's) onto the nylon fiber consisted of the following four steps (Fig. 1): (1) electron-beam irradiation. The nylon fiber was irradiated with an electron beam under nitrogen atmosphere at a dose of 40 kGy. (2) Grafting of DMAEMA. The electron-beam-irradiated fiber was immersed in 10 (v/v)% DMAEMA aqueous solution at 313 K for 6 h. The resulting DMAEMA-grafted fiber was referred to as the DMAEMA fiber. The degree of grafting, defined below, was set at 40%.

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

where  $W_0$  and  $W_1$  are the masses of the nylon and DMAEMA fibers, respectively. (3) Binding of hexacyanoferrate ions. The DMAEMA fiber, which was previously conditioned by immersing in 0.1 M HCl for 30 min, was immersed in 0.015 M potassium hexacyanoferrate ( $\text{K}_4[\text{Fe}(\text{CN})_6]$ ) aqueous solution at 298 K for 30 min. (4) Precipitation of potassium cobalt hexacyanoferrate compounds. The hexacyanoferrate-bound DMAEMA fiber was immersed in 0.125 M  $\text{CoCl}_2$  aqueous solution at 298 K in the presence of KCl in the concentration range of up to 0.5 M. The resulting potassium cobalt hexacyanoferrate compound-impregnated fiber was referred to as a KCo( $x$ )-HCFe fiber, where  $x$  in parentheses indicates the KCl concentration in molar unit. For comparison, a KCo(0)-HCFe fiber was prepared in the absence of KCl. The impregnation percentage of the fiber was defined as

$$\text{Impregnation percentage (\%)} = 100 (W_2 - W_1) / W_2 \quad (2)$$

where  $W_2$  is the mass of the KCo( $x$ )-HCFe fiber.

After the fiber was vacuum-dried, the fiber surface was observed by scanning electron microscopy (SEM, JEOL, JSM-6510A). In addition, the X-ray diffraction (XRD) pattern of the

KCo( $x$ )-HCFe-impregnated fiber was measured with M18HXF (MacScience).

### 2.3. Cesium removal in batch mode

The adsorption rate of the KCo(0.5)- or KCo(0)-HCFe fiber was determined in batch mode. Approximately 0.1 g of the fiber was gently stirred in 10 mL of 10 mg/L cesium chloride solution at 298 K for a contact time range of 3–60 min. The cesium concentration in the liquid was determined by inductively-coupled plasma mass-spectrometry (ICP-MS). The percentage of cesium removal from the solution and the distribution coefficient were evaluated as

$$\text{Removal percentage (\%)} = 100 (C_i - C_f) / C_i \quad (3)$$

$$\text{Distribution coefficient (cm}^3/\text{g)} = [(C_i - C_f)V / W] / C_f \quad (4)$$

where  $C_i$  and  $C_f$  are the initial and final cesium concentrations, respectively.  $V$  and  $W$  are the volume of the solution and weight of the KCo( $x$ )-HCFe fiber, respectively.

## 3. Results and discussion

### 3.1. Properties of potassium cobalt hexacyanoferrate compound-impregnated fibers

The XRD patterns of the KCo(0.5)- and KCo(0)-HCFe fibers are shown in Fig. 2 along with that of the 6-nylon fiber. Compared with the KCo(0)-HCFe fiber, the KCo(0.5)-HCFe fiber exhibited

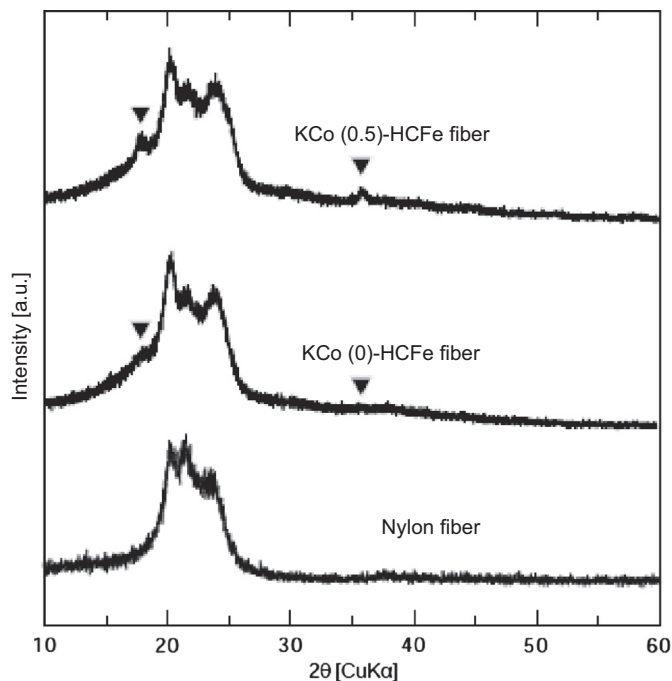


Fig. 2. XRD patterns of KCo(0.5)- and KCo(0)-HCFe fibers, and nylon fiber. The peaks indicated by ▼ correspond to  $\text{K}_2\text{Co}[\text{Fe}(\text{CN})_6]$ .

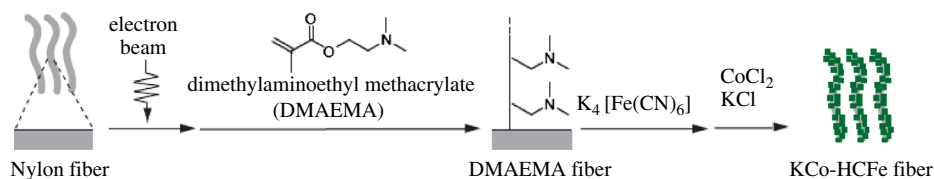


Fig. 1. Scheme of impregnation of potassium cobalt hexacyanoferrate onto nylon fiber by radiation-induced graft polymerization and subsequent chemical modifications.

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