



Roll-to-roll production of a cellulose filter with immobilized Prussian blue for ^{137}Cs adsorption



Hyowon Kim^a, Jaeyoung Seon^a, Sunho Yoon^b, Sungjun Bae^b, Sungwook Choung^c,
Yuhoon Hwang^{a,*}

^a Department of Environmental Engineering, Seoul National University of Science and Technology, Seoul 01811, Republic of Korea

^b Department of Civil and Environmental Engineering, Konkuk University, Seoul 05029, Republic of Korea

^c Korea Basic Science Institute, Cheongju 28119, Republic of Korea

ARTICLE INFO

Editor: Yunho Lee

Keywords:

Prussian blue
Cellulose filter
Radioactive cesium
Roll-to-roll

ABSTRACT

Nuclear power plant accidents can release radioactive cesium in the water and, thus, proper removal technologies are required. Since Prussian blue can selectively adsorb Cs, studies to attach it onto supporting materials such as activated carbon and filters have been conducted. Here, we propose a roll-to-roll method for fabricating cellulose filters with immobilized Prussian blue easily and in large quantities; the physical properties and Cs adsorption performance of these filters were similar to their counterparts prepared in a conventional batch reactor. We performed batch and continuous filtration experiments to confirm their decontamination performance for radioactive cesium with 300 Bq/L of initial concentration. In the batch test, an adsorbent dosage of 0.05 g/L enabled a > 95 % decontamination in 24 h; the continuous filtration experiment revealed that a 95 % decontamination was kept for up to 10 h. The proposed roll-to-roll method could allow the production of several filters for the effective removal of cesium from the water.

1. Introduction

Nuclear energy is released when the nuclear mass of a radioactive element changes through radiation decay. Although nuclear power stations are considerably expensive to build, their longevity and low running cost make them particularly cost-effective. Most of the CO₂ emissions associated with nuclear power plants occur during construction and fuel processing and not during electricity generation [1,2]. However, nuclear power generation produces radioactive waste and there is a concern about the possibility of major disasters when accidents occur; for example, in 2011, the Fukushima Daiichi accident caused severe damages and the remediation was very difficult and time-consuming. A nuclear accident can spread radioactive contaminants worldwide through water, air, and soil [3]. In particular, radioactive materials discharged in water systems, such as the seas and rivers, have extremely high diffusion rates, causing problems to the drinking water supply.

^{137}Cs is a radioisotope commonly released during a nuclear accident and accounts for 6.3 % of fission products [4]. It has a long half-life (30 years) compared to other radioactive contaminants and is difficult to remove from the water because it has a small hydration radius, a large diffusion coefficient, and is easily dissolved in water [5–8]. When ^{137}Cs

enters the human body, it can induce genetic diseases and disorders, mutations, and cancers [9,10].

Prussian blue (PB) is a metal ferrocyanide compound (Fe₄[Fe(CN₆)₃]) used as a blue dye. It is an effective Cs adsorbent and, thus, was given to the people exposed to radiocesium during the Chernobyl nuclear accident [11]; PB has high selectivity toward cesium because the size of its cubic lattice structure well matches the size of hydrated cesium ions [12–16]. Takahashi et al. (2018) recently attributed this high selectivity to the following cationic exchange process. The lattice structure of metal ferrocyanides is formed by a porous network of metal cations and anions of iron. If this structure presents a vacant region, hydrated Cs⁺ can enter through it and get adsorbed via cation exchange; in the case of Prussian blue, Cs⁺ is exchanged with the K⁺ inside the Prussian blue lattice [17].

However, PB has a very small size (5–200 nm) and is highly dispersible; hence, it is difficult to separate and recover from water systems [18–20]. Therefore, immobilizing Prussian blue on supporting materials is one of the leading research topics related to the improvement of its practical applicability. Previous studies have focused on the production and use of filter-type adsorbents containing PB [21], magnetic materials/PB composites for the removal by magnetic forces [22,23], carbon nanomaterials such as carbon nanotubes and graphene

* Corresponding author.

E-mail address: yhhwang@seoultech.ac.kr (Y. Hwang).

<https://doi.org/10.1016/j.jece.2020.104273>

Received 20 May 2020; Received in revised form 6 July 2020; Accepted 12 July 2020

Available online 15 July 2020

2213-3437/ © 2020 Elsevier Ltd. All rights reserved.

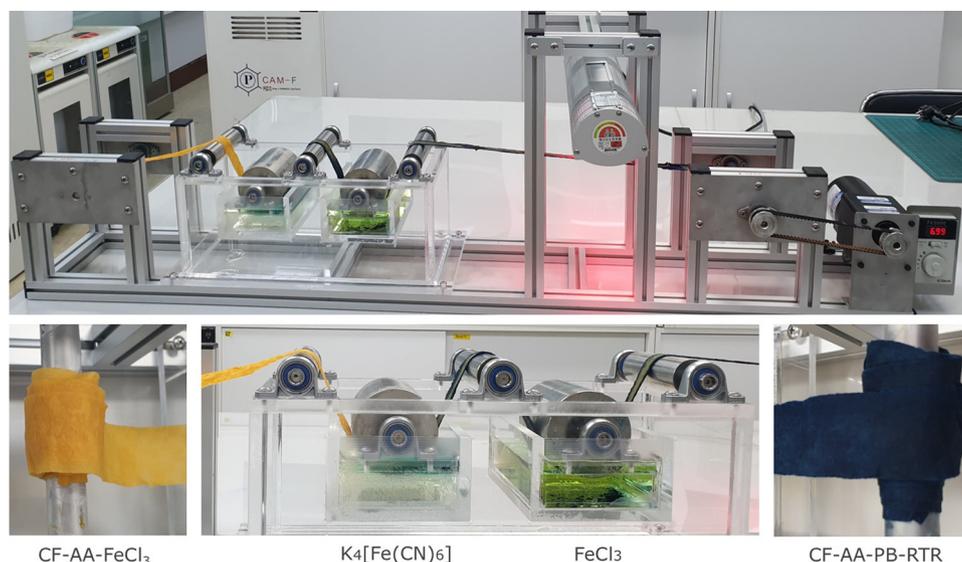


Fig. 1. A roll-to-roll machine for mass production of acrylic acid-modified cellulose filters with immobilized Prussian blue (CF-AA-PB-RTR).

oxide [24–26], PB-containing polymer beads [27,28], PB immobilized on porous materials [29], and natural minerals as a PB support [30]. These various PB composites have exhibited a superior cesium adsorption capacity with high selectivity, which makes them suitable for Cs remediation. However, the fabrication complexity should be considered for practical application.

In our previous work, we successfully developed a method for preparing a filter-type Cs adsorbent containing PB [31]. The approach includes the use of commercial cellulose filters (CFs) as the supporting matrix, their functionalization via the simple chemical reaction to obtain carboxylic groups, and PB synthesis through layer-by-layer (LbL) assembly. This procedure provided higher PB loading and stronger stability against PB leaching in aqueous solutions, resulting in better Cs adsorption performance compared to the unmodified CFs.

To assure the availability of filter-type Cs adsorbent to the wide population in a short time, their manufacture should be adaptable to the processes currently available in the textile industry. Most studies on producing Cs adsorbent with PB are based on batch processing and fall short of demonstrating that the materials can be produced and processed with large scale equipment [31–33]. Pad-dyeing technology is a process of continuous dyeing in which the fabric in open width is padded with dyestuff. Compared to the conventional batch system, it has several advantages such as simplicity of the dyeing process, reduction in water, energy, chemical consumption; therefore, it has been widely applied in the industrial scale dyeing process [34,35]. This type of production was also called the roll-to-roll production, and it has been applied in nanoparticle fabrication [36] as well as electronic application [37,38].

In this study, we propose an improvement for the rapid production of cellulose filters with immobilized Prussian blue in large quantities. Prussian blue was synthesized via the roll-to-roll method, which allowed its automatic operation. The characteristics and cesium adsorption performance of the new adsorbent were compared with those of the adsorbents fabricated through the conventional method in a batch reactor; moreover, the practical applicability of the prepared adsorbent for radioactive ^{137}Cs removal was evaluated via batch and continuous filtration tests.

2. Materials and methods

2.1. Materials

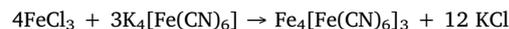
The cellulose filter was purchased from Taebong (Republic of

Korea); its tensile strength and permeability coefficient were 1.22 N/cm^2 and 0.079 cm/s , respectively. The CF surface modification was performed with acrylic acid (AA, 99 %), potassium persulfate (KPS, 99 %), sodium chloride (93 %), and ethanol (99 %), obtained from Samchun Chemical Reagent Corporation Ltd. (Republic of Korea). Iron (III) chloride hexahydrate (97 %) and potassium ferrocyanide trihydrate (99 %), by Duksan Chemical Reagent Corporation Ltd. (Korea), were the PB precursors. A standard ^{133}Cs (1000 mg/L; Kanto Chemical Corporation Inc., Japan) and a standard ^{137}Cs source (certified by the Korea Research Institute of Standards and Science) solutions were used. All the solutions were prepared with deionized water.

2.2. Preparation of CF with immobilized PB via the batch method

Our previous study [31] described in detail the preparation of CF with immobilized PB (CF-AA-PB). In brief, the synthesis process consists of modifying the CF surface with (AA) and synthesizing PB via the LbL approach in batch mode. First, the surface modification by AA was performed via radical polymerization with KPS.

Then, PB was immobilized on the AA-modified CF (CF-AA) via LbL assembly, which is basically similar to the in situ methodology. PB was synthesized in situ by sequentially injecting its precursors in the CF-AA. The major chemical reaction involved was as follows [39].



The CF-AA (250 mg) was added to a solution (50 mL) of 20 mM FeCl_3 for 24 h. Then, CF-AA, with the immobilized iron(III), was separated from the solution and it was reacted with 50 mL of a 20 mM $\text{K}_4\text{Fe}(\text{CN})_6$ solution for 15 min, followed by the addition of a 5 mM FeCl_3 solution (50 mL) for 15 min, finally giving CF-AA-PB.

2.3. Preparation of CF with immobilized PB via the roll-to-roll method

A roll-to-roll machine (Fig. 1) was designed to produce, similarly to the batch method described in Subsection 2.2, large quantities of CF-AA-PB. A CF-AA roll with a 1 m length and a 3 cm width was used. As in the batch method, the duration of the first FeCl_3 , the $\text{K}_4\text{Fe}(\text{CN})_6$, and the additional FeCl_3 immersion were 24 h, 15 min, and 15 min, respectively. Since the first FeCl_3 impregnation step was very long compared to the others, we decided to preliminarily immerse the CF-AA roll into a 20 mM FeCl_3 solution for 24 h and to perform the roll-to-roll fabrication only for the last two steps.

The iron(III)-impregnated CF-AA roll (CF-AA- FeCl_3) was inserted at

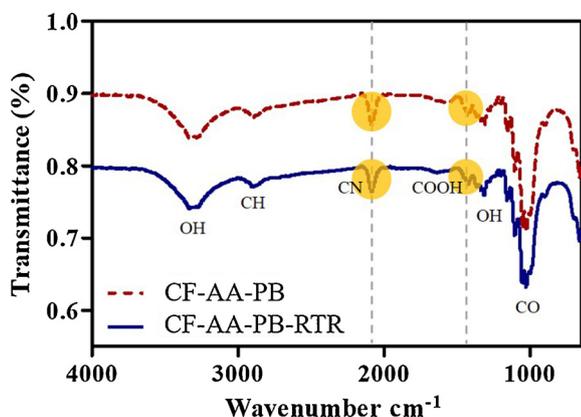


Fig. 2. Fourier-transform infrared spectra of acrylic acid-modified cellulose filters with immobilized Prussian blue, fabricated via the batch method (CF-AA-PB) and the roll-to-roll technique (CF-AA-PB-RTR).

one end of the roll-to-roll machine and slowly moved through two reaction chambers, respectively containing a 20 mM potassium ferrocyanide and a 5 mM iron chloride solution, allowing the same LbL synthetic process as in Subsection 2.2. to ensure a reaction time of 15 min in each chamber, we calculated the suitable diameter of the cylinders in the chambers and the motor speed. The diameter of the cylinders and rolling speeds were 8 cm and 0.84 cm/min, respectively, allowing 15 min of reaction time at each bath. After the PB synthesis, the roll was dried by passing through a heating lamp. The resulting product was denoted as CF-AA-PB-RTR in order to be distinguished from that prepared via the batch method.

2.4. Filter characterization

The characteristics of CF-AA-PB and CF-AA-PB-RTR were compared to confirm the physical and chemical similarity between them. The effect of functionalization was evaluated by monitoring the changes in the functional groups on the surface by using a Fourier-transform infrared spectroscopy (FTIR) analyzer (TENSOR27, Bruker, Germany) operated in the 400–4000 cm^{-1} range. A scanning electron microscopy/energy dispersive spectroscopy (SEM/EDS) system (SU8010, Hitachi High Technologies Corporation, Japan) was used to observe their surface modifications and qualitatively analyze their chemical compositions.

2.5. ^{137}Cs adsorption

2.5.1. Adsorption performances of CF-AA-PB and CF-AA-PB-RTR

The similarity between CF-AA-PB and CF-AA-PB-RTR was evaluated also based on their cesium adsorption performance. Solutions with 0.1 and 1 mg/L concentrations of a non-radioactive isotope, ^{133}Cs , were prepared; thereafter, we immersed, separately, 0.1 g of each filter into each of them (50 mL), which were contained in sealed conical polyethylene tubes. The pH of the aqueous cesium solutions was not adjusted (initial pH = 6.2), and they were continuously shaken at 303 K for 24 h in an agitator. The adsorption batch test was performed in duplicate. The cesium ion concentration in the resulting supernatant was measured with an inductively coupled plasma mass spectrometry (ICP-MS) system (NexION 350D, Perkin-Elmer, USA). All sample was measured three times, and the average was obtained for analytical value.

2.5.2. Continuous filtration experiment with CF-AA-PB-RTR

This test was performed to determine the flow rate required for an effective treatment. A polypropylene filter holder for 25-mm membranes (Advantec, USA) was utilized. The CF-AA-PB-RTR was cut into a

25 mm diameter circle and placed in the holder. Then, the 1 mg/L ^{133}Cs solution was passed through the filter in the upward direction. The filtration velocity was varied (1, 0.5, and 0.2 cm/min, resulting in contact times of 6, 12, and 30 s, respectively) so as to evaluate its effect on the cesium removal. The operation was continued for 10 h.

2.6. ^{137}Cs removal by CF-AA-PB-RTR

2.6.1. Batch adsorption experiment

The radioactive ^{137}Cs , with a physical half-life of 30.1 years, was used for evaluating the practical applicability of CF-AA-PB-RTR. The ^{137}Cs concentration was set to 300 Bq/L. We selected 300 Bq/kg of ^{137}Cs water, which loaded 30 times more radioactivity than the upper limit value of 10 Bq/kg for drinking water set by the WHO. Various CF-AA-PB-RTR dosages (0.01, 0.05, 0.1, 0.5, and 1 g/L) were added to 50 mL of a 300 Bq/L ^{137}Cs solution, and the resulting mixture was stirred for 24 h with a vertical rotary stirrer. Furthermore, the ^{137}Cs concentration in the solutions was measured for 3600 s by using an HPGe gamma-ray spectroscopy (CANBERRA, RAD Small Anode Germanium Well Detector, USA). Then, the removal efficiency was calculated based on the initial and the final ^{137}Cs concentration. The adsorption batch test was performed in duplicate.

2.6.2. Continuous filtration experiment

This test was performed similarly to that described in Subsection 2.5.2. The experiment was conducted at a 0.2 cm/min linear filtration velocity, which was the optimal condition determined in the analogous ^{133}Cs test. The ^{137}Cs concentration was set to 300 Bq/L and the experiment was performed for 70 h. The sample was periodically taken with a fraction collector and the ^{137}Cs concentration in the filtrate was determined with the HPGe gamma-ray spectroscopy.

3. Results and discussion

3.1. Comparison of CF-AA-PB and CF-AA-PB-RTR performance

3.1.1. FTIR analysis

FTIR analysis was performed to confirm the chemical similarity between CF-AA-PB and CF-AA-PB-RTR, and the results are illustrated in Fig. 2. Overall, the FTIR spectra of CF-AA-PB and CF-AA-PB-RTR were very similar, without significant differences.

In accordance with the chemical composition of cellulose, hydroxyl, methyl, and carbonyl peaks were observed at, respectively, 3640–3200, 3000–2850, and 1730–1700 cm^{-1} [40]. Since the surface of both filters was modified with AA, also the peak of carboxylic acid (1760–1690 cm^{-1}) was clearly detected [41]. Moreover, the two samples exhibited a sharp peak at 2070 cm^{-1} attributed to the cyanide group, which is one of the building blocks of PB and, thus, indicates its successful formation [19]. These similar peaks suggest that the two adsorbents have also similar properties.

3.1.2. Morphological analysis

The microscopic morphology of CF-AA-PB and CF-AA-PB-RTR was investigated via high-resolution SEM (Fig. 3). The structure of the CFs mainly consisted of entangled thin fibers (diameter of around 10–20 μm) and many internal spaces. We could hardly observe a noticeable difference between the two materials, even at different magnifications; however, CF-AA-PB-RTR showed a more uniform PB distribution than CF-AA-PB, which exhibited some cracks on the PB layer.

The amount of PB immobilized on the two filters was measured through EDS analysis (Table 1). Due to the chemical composition of cellulose (i.e., $(\text{C}_6\text{H}_{10}\text{O}_5)_n$), the carbon and oxygen contents were large. Note that nitrogen and iron were detected in both samples, directly demonstrating the successful preparation of $\text{Fe}_4[\text{Fe}(\text{CN})_6]_3$. The N and Fe contents were, respectively, 8.19 % and 14.67 % in CF-AA-PB and 7.05 % and 11.50 % in CF-AA-PB-RTR. This means that CF-AA-PB

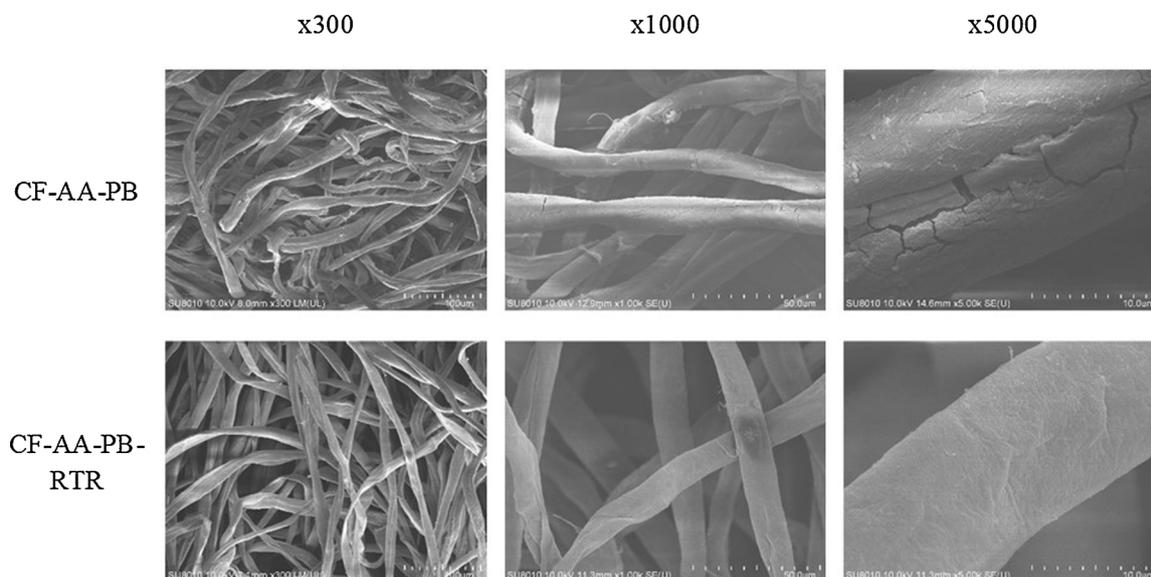


Fig. 3. High-resolution scanning electron microscopy images at different magnifications of acrylic acid-modified cellulose filters with immobilized Prussian blue, fabricated via the batch method (CF-AA-PB) and the roll-to-roll technique (CF-AA-PB-RTR).

Table 1

Energy dispersive spectroscopy results for acrylic acid-modified cellulose filters with immobilized Prussian blue, fabricated via the batch method (CF-AA-PB) and the roll-to-roll technique (CF-AA-PB-RTR).

	Carbon (%)	Oxygen (%)	Nitrogen (%)	Iron (%)
CF-AA-PB	28.42	48.72	8.19	14.67
CF-AA-PB-RTR	29.49	51.96	7.05	11.50

contained a slightly higher PB (13.9 % for N and 21.6 % for Fe). Since the amount of immobilized PB directly correlates with the cesium adsorption performance, we successively conducted an adsorption experiment (Subsection 3.1.3).

3.1.3. ^{133}Cs adsorption performance

Fig. 4. ^{133}Cs removal rate of acrylic acid-modified cellulose filters with immobilized Prussian blue, fabricated via the batch method (CF-AA-PB) and the roll-to-roll technique (CF-AA-PB-RTR). The adsorbent dosage was 2 g/L.

Adsorption experiments using isotopes (^{133}Cs) were conducted to evaluate the cesium adsorption performance of CF-AA-PB and CF-AA-PB-RTR. **Fig. 4** compares the cesium removal efficiencies obtained when adding 2 g/L CF-AA-PB and CF-AA-PB-RTR to 0.1 and 1 mg/L cesium solutions. When the initial cesium concentration was 0.1 mg/L, the removal efficiency of CF-AA-PB and CF-AA-PB-RTR was 99.9 % and

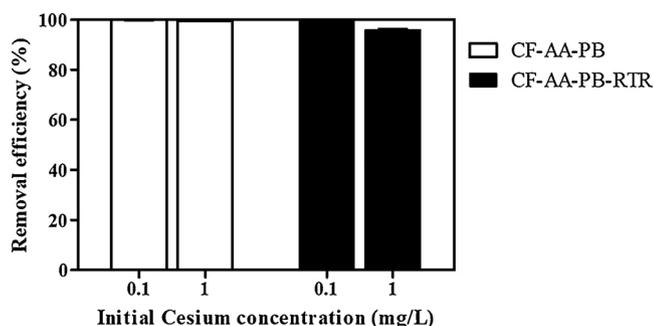


Fig. 4. ^{133}Cs removal rate of acrylic acid-modified cellulose filters with immobilized Prussian blue, fabricated via the batch method (CF-AA-PB) and the roll-to-roll technique (CF-AA-PB-RTR). The adsorbent dosage was 2 g/L.

99.5 %, respectively; at 1 mg/L, the corresponding efficiency was 99.6 % and 96.1 %. This slightly higher cesium removal efficiency of CF-AA-PB might be due to its high PB loading, as revealed by the SEM/EDS analysis.

The material characterization and cesium adsorption performance evaluation confirmed the similarities between CF-AA-PB-RTR and CF-AA-PB in terms of functional groups, PB loading, and cesium adsorption capacity. In this regard, we successively evaluated the radiocesium decontamination capacity of CF-AA-PB-RTR.

3.2. ^{137}Cs decontamination by CF-AA-PB-RTR

3.2.1. Batch adsorption experiment

Different CF-AA-PB-RTR dosages (0.01, 0.05, 0.1, 0.5, and 1 g/L) were injected in 50 mL of a 300 Bq/L ^{137}Cs solution, which was successively stirred and reacted for 24 h. Then, the removal efficiency was calculated based on the initial and the final ^{137}Cs concentration, which was measured as the radiation dose by the HPGe gamma-ray spectroscope (**Fig. 5**).

An adsorbent dosage of 0.01 g/L removed 92.6 % of ^{137}Cs from the solution; the removal efficiency of CF-AA-PB-RTR increased along with this dosage (97.6 %, 99.5 %, 99.5 %, and 99.9 % at, respectively, 0.05, 0.1, 0.5, and 1 g/L). According to the World Health Organization recommendations for radionuclides in drinking water, the guidance level for ^{137}Cs is 10 Bq/L. Thus, for an initial ^{137}Cs concentration of 300 Bq/L, a minimum removal efficiency of 96.7 % (indicated by the dotted line in **Fig. 5**) is required. When we used a CF-AA-PB-RTR dosage of 0.05 g/L, we reached a higher removal efficiency, i.e., 97.6 %, confirming the applicability of CF-AA-PB-RTR for the radiocesium decontamination of

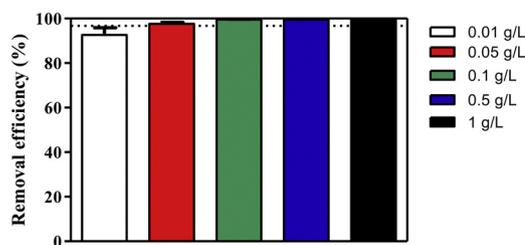


Fig. 5. ^{137}Cs removal efficiency of different dosages of the acrylic acid-modified cellulose filter with immobilized Prussian blue, fabricated via the roll-to-roll technique. The initial ^{137}Cs was 300 Bq/L.

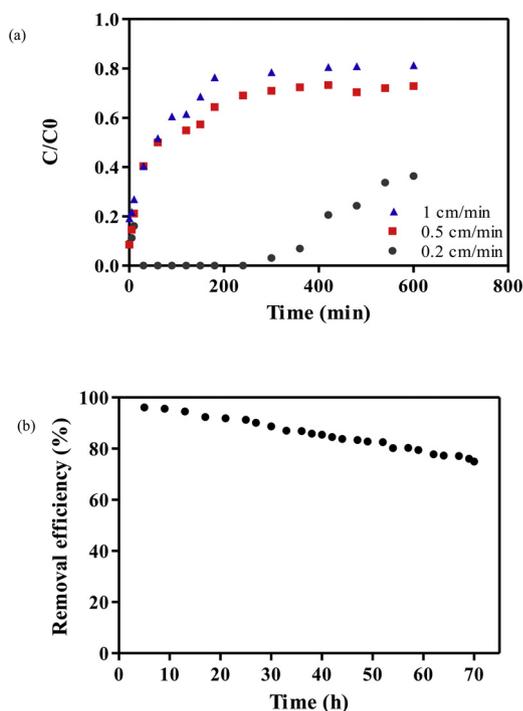


Fig. 6. (a) ^{133}Cs removal efficiency at different filtration velocities for an initial ^{133}Cs concentration of 1 mg/L. (b) ^{137}Cs removal efficiency at a filtration velocity of 0.2 cm/min for an initial ^{137}Cs concentration of 300 Bq/L.

water.

3.2.2. Continuous filtration experiments

Subsection 3.2.1 has illustrated the superior decontamination performance of CF-AA-PB-RTR, but the conducted batch experiment is different from its actual application scenario. Therefore, we successively evaluated the CF-AA-PB-RTR performance also in a continuous filtration process, that is, where contaminated water passes through it.

To determine the appropriate conditions of the filtration process, we preliminarily carried out an experiment with ^{133}Cs ; we varied the filtration velocity (1.0, 0.5, and 0.2 cm/min, resulting in contact times of 6, 12, and 30 s, respectively) while measuring the ^{133}Cs concentration in the filtrate (Fig. 6(a)). The initial ^{133}Cs concentration was 1 mg/L. At filtration rates of 1 and 0.5 cm/min, the ^{133}Cs concentration in the effluent increased immediately after the beginning of the filtration operation, exceeding 0.5 mg/L after about 60 min and, then, stabilizing gradually. However, the ^{133}Cs concentration was slightly lower at 0.5 cm/min than at 1.0 cm/min; after 10 h of filtration, it was 0.73 and 0.81 mg/L at, respectively, 0.5 and 1.0 cm/min.

When the filtration velocity was set to 0.2 cm/min, a significantly different trend was observed. We could detect the outflow of ^{133}Cs for about 10 min immediately after the process started, but no ^{133}Cs was present in the effluent during the next 4 h. After 5 h, the ^{133}Cs concentration in the effluent began to increase slowly and became 0.36 mg/L after 10 h. Therefore, we decided to use the filtration velocity of 0.2 cm/min in the next experiment with radiocesium.

We performed the ^{137}Cs adsorption experiment in continuous filtration mode to mimic an actual situation of radiocesium removal (Fig. 6(b)). The initial ^{137}Cs concentration was 300 Bq/L and the experiment lasted 70 h. CF-AA-PB-RTR maintained a ^{137}Cs removal efficiency of 95 % or more during the first 10 h. Then, the efficiency continuously decreased, reaching 90 % after 27 h and 80 % after 57 h; at the end of the experiment, the removal efficiency was 75 %. For the results observed during the first 10 h indicated that, in an actual radioactive accident, CF-AA-PB-RTR could be effectively used as an initial response to radiocesium release.

4. Conclusions

This study aimed to develop a filter material for protecting the water sources in the case of radioactive cesium leakage. In a previous work, we synthesized a cellulose filter with immobilized Prussian blue via the batch method; here, for an easy mass production method, we adopted the roll-to-roll fabrication technique. To confirm the similarity between the filters prepared via the two methods, we evaluated their physical properties through FTIR and SEM analyses. Their ^{137}Cs adsorption performance was also compared. The results confirmed that they had similar properties and performance. Then, we performed batch adsorption and continuous filtration experiments to investigate the removal efficiency of radioactive cesium by CF-AA-PB-RTR. In the batch adsorption experiment, we observed an efficiency higher than 95 % when using a 0.05 g/L dosage of the adsorbent. In the continuous filtration experiment, a similarly higher efficiency was maintained for more than 10 h; moreover, an efficiency higher than 90 % was still exhibited after 24 h. Based on such a high performance, CF-AA-PB-RTR is expected to remove radioactive cesium in contaminated waters effectively.

CRedit authorship contribution statement

Hyowon Kim: Investigation, Writing - original draft. **Jaeyoung Seon:** Investigation. **Sunho Yoon:** Methodology. **Sungjun Bae:** Investigation, Methodology. **Sungwook Chung:** Methodology. **Yuhoon Hwang:** Conceptualization, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This study was supported by the Creative Convergence Research Project (CAP-15-07-KICT) of the National Research Council of Science and Technology (NST). This research was also supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2020R1A6A1A0304274211).

References

- [1] M.S. Dresselhaus, I.L. Thomas, *Alternative energy technologies*, *Nature* 414 (2001) 332–337.
- [2] C.W. Forsberg, *Sustainability by combining nuclear, fossil, and renewable energy sources*, *Prog. Nucl. Energy* 51 (2009) 192–200.
- [3] A. Mathieu, M. Kajino, I. Korsakissok, R. Périllat, D. Quélo, A. Quérel, O. Saunier, T.T. Sekiyama, Y. Igarashi, D. Didier, Fukushima Daiichi-derived radionuclides in the atmosphere, transport and deposition in Japan: a review, *Appl. Geochem.* 91 (2018) 122–139.
- [4] D. Yang, S. Sarina, H. Zhu, H. Liu, Z. Zheng, M. Xie, S.V. Smith, S. Komarneni, Capture of radioactive cesium and iodide ions from water by using titanate nanofibers and nanotubes, *Angew. Chem. Int. Ed.* 50 (2011) 10594–10598.
- [5] M.R. Awual, T. Yaita, T. Taguchi, H. Shiwaku, S. Suzuki, Y. Okamoto, Selective cesium removal from radioactive liquid waste by crown ether immobilized new class conjugate adsorbent, *J. Hazard. Mater.* 278 (2014) 227–235.
- [6] A. El-Kamash, Evaluation of zeolite A for the sorptive removal of Cs⁺ and Sr²⁺ ions from aqueous solutions using batch and fixed bed column operations, *J. Hazard. Mater.* 151 (2008) 432–445.
- [7] F. Jia, J. Wang, Separation of cesium ions from aqueous solution by vacuum membrane distillation process, *Prog. Nuclear Energy* 98 (2017) 293–300.
- [8] F. Ma, Z. Li, H. Zhao, Y. Geng, W. Zhou, Q. Li, L. Zhang, Potential application of graphene oxide membranes for removal of Cs(I) and Sr(II) from high level-liquid waste, *Sep. Purif. Technol.* 188 (2017) 523–529.
- [9] M.A. Olatunji, M.U. Khandaker, H.N.M.E. Mahmud, Y.M. Amin, Influence of adsorption parameters on cesium uptake from aqueous solutions- a brief review, *RSC Adv.* 5 (2015) 71658–71683.
- [10] T. Sangvanich, V. Sukwarotwat, R.J. Wiacek, R.M. Grudzien, G.E. Fryxell,

- R.S. Addleman, C. Timchalk, W. Yantasee, Selective capture of cesium and thallium from natural waters and simulated wastes with copper ferrocyanide functionalized mesoporous silica, *J. Hazard. Mater.* 182 (2010) 225–231.
- [11] P.J. Faustino, Y. Yang, J.J. Progar, C.R. Brownell, N. Sadrieh, J.C. May, E. Leutzinger, D.A. Place, E.P. Duffy, F. Houn, S.A. Loewke, V.J. Mecozzi, C.D. Ellison, M.A. Khan, A.S. Hussain, R.C. Lyon, Quantitative determination of cesium binding to ferric hexacyanoferrate: Prussian blue, *J. Pharm. Biomed. Anal.* 47 (2008) 114–125.
- [12] C. Delchet, A. Tokarev, X. Dumail, G. Toquer, Y. Barré, Y. Guari, C. Guerin, J. Larionova, A. Grandjean, Extraction of radioactive cesium using innovative functionalized porous materials, *RSC Adv.* 2 (2012) 5707–5716.
- [13] P.A. Haas, A review of information on ferrocyanide solids for removal of cesium from solutions, *Sep. Sci. Technol.* 28 (1993) 2479–2506.
- [14] B. Kong, J. Tang, Z. Wu, J. Wei, H. Wu, Y. Wang, G. Zheng, D. Zhao, Ultralight mesoporous magnetic frameworks by interfacial assembly of Prussian blue nanocubes, *Angew. Chem. Int. Ed.* 53 (2014) 2888–2892.
- [15] C. Loos-Neskovic, S. Ayrault, V. Badillo, B. Jimenez, E. Garnier, M. Fedoroff, D.J. Jones, B. Merinov, Structure of copper-potassium hexacyanoferrate (II) and sorption mechanisms of cesium, *J. Solid State Chem.* 177 (2004) 1817–1828.
- [16] P.C.F. Pau, J.O. Berg, W.G. McMillan, Application of Stokes' law to ions in aqueous solution, *J. Phys. Chem.* 94 (1990) 2671–2679.
- [17] A. Takahashi, H. Tanaka, K. Minami, K. Noda, M. Ishizaki, M. Kurihara, H. Ogawa, T. Kawamoto, Unveiling Cs-adsorption mechanism of Prussian blue analogs: Cs⁺-percolation via vacancies to complete dehydrated state, *RSC Adv.* 8 (2018) 34808–34816.
- [18] Y. Kim, I. Kim, T.S. Lee, E. Lee, K.J. Lee, Porous hydrogel containing Prussian blue nanoparticles for effective cesium ion adsorption in aqueous media, *J. Ind. Eng. Chem.* 60 (2018) 465–474.
- [19] H. Wi, H. Kim, D. Oh, S. Bae, Y. Hwang, Surface modification of poly(vinyl alcohol) sponge by acrylic acid to immobilize Prussian blue for selective adsorption of aqueous cesium, *Chemosphere.* 226 (2019) 173–182.
- [20] S.C. Jang, S.B. Hong, H.M. Yang, K.W. Lee, J.K. Moon, B.K. Seo, Y.S. Huh, C. Roh, Removal of radioactive cesium using Prussian blue magnetic nanoparticles, *Nanomaterials Basel (Basel)* 4 (2014) 894–901.
- [21] H. Kim, M. Kim, W. Lee, S. Kim, Rapid removal of radioactive cesium by polyacrylonitrile nanofibers containing Prussian blue, *J. Hazard. Mater.* 347 (2018) 106–113.
- [22] H. Yang, S. Jang, S.B. Hong, K. Lee, C. Roh, Y.S. Huh, B. Seo, Prussian blue-functionalized magnetic nanoclusters for the removal of radioactive cesium from water, *J. Alloys. Compd.* 657 (2016) 387–393.
- [23] H. Yang, J.R. Hwang, D.Y. Lee, K.B. Kim, C.W. Park, H.R. Kim, K. Lee, Eco-friendly one-pot synthesis of Prussian blue-embedded magnetic hydrogel beads for the removal of cesium from water, *Sci. Rep.* 8 (2018) 11476.
- [24] H. Basu, S. Saha, M.V. Pimple, R.K. Singhal, Graphene-Prussian blue nanocomposite impregnated in alginate for efficient removal of cesium from aquatic environment, *J. Environ. Chem. Eng.* 6 (2018) 4399–4407.
- [25] F. Chen, G. Jin, S. Peng, X. Liu, J. Tian, Recovery of cesium from residual salt lake brine in Qarham playa of Qaidam Basin with Prussian blue functionalized graphene/carbon fibers composite, *Colloids Surf. A Physicochem. Eng. Asp.* 509 (2016) 359–366.
- [26] A.A. Kadam, J. Jang, D.S. Lee, Facile synthesis of pectin-stabilized magnetic graphene oxide Prussian blue nanocomposites for selective cesium removal from aqueous solution, *Bioresour. Technol.* 216 (2016) 391–398.
- [27] A.K. Vipin, B. Hu, B. Fugetsu, Prussian blue caged in alginate/calcium beads as adsorbents for removal of cesium ions from contaminated water, *J. Hazard. Mater.* 258–259 (2013) 93–101.
- [28] D. Oh, B. Kim, S. Kang, Y. Kim, S. Yoo, S. Kim, Y. Chung, S. Choung, J. Han, S. Jung, H. Kim, Y. Hwang, Enhanced immobilization of Prussian blue through hydrogel formation by polymerization of acrylic acid for radioactive cesium adsorption, *Sci. Rep.* 9 (2019) 16334.
- [29] R. Turgis, G. Arrachart, C. Delchet, C. Rey, Y. Barré, S. Pellet-Rostaing, Y. Guari, J. Larionova, A. Grandjean, An original “Click and bind” approach for immobilizing copper hexacyanoferrate nanoparticles on mesoporous silica, *Chem. Mater.* 25 (2013) 4447–4453.
- [30] H.A. Alamudy, K. Cho, Selective adsorption of cesium from an aqueous solution by a montmorillonite-Prussian blue hybrid, *Chem. Eng. J.* 349 (2018) 595–602.
- [31] H. Kim, H. Wi, S. Kang, S. Yoon, S. Bae, Y. Hwang, Prussian blue immobilized cellulosic filter for the removal of aqueous cesium, *Sci. Total Environ.* 670 (2019) 779–788.
- [32] S. Jang, Y. Haldorai, G. Lee, S. Hwang, Y. Han, C. Roh, Y.S. Huh, Porous three-dimensional graphene foam/Prussian blue composite for efficient removal of radioactive ¹³⁷Cs, *Sci. Rep.* 5 (2015) 17510.
- [33] H. Bang, K. Watanabe, R. Nakashima, W. Kai, K. Song, J.S. Lee, M. Gopiraman, I. Kim, A highly hydrophilic water-insoluble nanofiber composite as an efficient and easily-handleable adsorbent for the rapid adsorption of cesium from radioactive wastewater, *RSC Adv.* 4 (2014) 59571–59578.
- [34] Z. Yuan, Y. Zhu, J. Shi, X. Liu, L. Huang, Life-cycle assessment of continuous dyeing technology for cotton fabrics, *Int. J. Life Cycle Assess.* 18 (2013) 659–672.
- [35] A.D. Broadbent, J. Bissou-Billong, M. Lhachimi, Y. Mir, S. Capistran, Continuous dyeing of cotton with reactive dyes using infrared heat, *Ind. Eng. Chem. Res.* 44 (2005) 3954–3958.
- [36] M. Jiao, Y. Yao, G. Pastel, T. Li, Z. Liang, H. Xie, W. Kong, B. Liu, J. Song, L. Hu, Fly-through synthesis of nanoparticles on textile and paper substrates, *Nanoscale.* 11 (2019) 6174–6181.
- [37] S. Bae, H. Kim, Y. Lee, X. Xu, J. Park, Y. Zheng, J. Balakrishnan, T. Lei, H. Ri Kim, Y.I. Song, Y. Kim, K.S. Kim, B. Özyilmaz, J. Ahn, B.H. Hong, S. Iijima, Roll-to-roll production of 30-inch graphene films for transparent electrodes, *Nat. Nanotechnol.* 5 (2010) 574–578.
- [38] A. Lund, S. Darabi, S. Hultmark, J.D. Ryan, B. Andersson, A. Ström, C. Müller, Roll-to-roll dyed conducting silk yarns: a versatile material for E-Textile devices, *Int. J. Adv. Mater. Technol.* 3 (2018) 1800251.
- [39] L. Samain, F. Grandjean, G.J. Long, P. Martinetto, P. Bordet, D. Strivay, Relationship between the synthesis of Prussian blue pigments, their color, physical properties, and their behavior in paint layers, *J. Phys. Chem. C.* 117 (2013) 9693–9712.
- [40] D. Ciolacu, F. Ciolacu, V.I. Popa, Amorphous cellulose - Structure and characterization, *Cell. Chem. Technol.* 45 (2011) 13–21.
- [41] Y. Shi, Z. Xue, X. Wang, L. Wang, A. Wang, Removal of methylene blue from aqueous solution by sorption on lignocellulose-g-poly(acrylic acid)/montmorillonite three-dimensional cross-linked polymeric network hydrogels, *Polym. Bull.* 70 (2013) 1163–1179.