

Fibrous zeolite-polymer composites for decontamination of radioactive waste water extracted from radio-Cs fly ash

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Abstract— Fly ash polluted in Fukushima was treated by washing water and then the radioactive waste water was decontaminated with zeolite polymer composite fiber. The character was compared in zeolite, prussian blue and the composite fiber. The strong alkali condition of the radioactive waste water damaged in PB, while zeolite and the composite fiber successfully treated the radio-Cs by sorption process of batch and column experiments. The ability of the composite fiber was superior performance in the time course of the sorption process. The radioactive Cs binding to the composite fiber and zeolite was followed in Freundlich mechanism, showing multibanding to their adsorbents in the extra-diluted condition, but, each adsorbent obeyed saturated mechanism for non-radioactive Cs

Index Terms—Fly ash, zeolite, sorption process.

I. INTRODUCTION

A large quantity of radioactive material radiated in Fukushima environment by an accident of the Fukushima first Nuclear Power Plant started from East Japan great earthquake disaster of March 11, 2011 [1-4]. The decontamination work to radiocesium (Cs) has been pushed forward and implemented for removal of radioactive materials zealously at the present after particularly times have passed for six years [5-7]. A method of decontaminating includes there in various ways, but, as a radioactive Cs adsorption material, zeolite attracts attention to be really used. However, problems of the zeolite are pointed out in the real decontamination that is necessarily to recovery the waste zeolite after the decontamination [8-13]. Therefore, there are various kinds of improvement and demands of the advancement utilization. Among them, for the efficient removal of radioactive Cs from low-level radioactive liquid wastes, natural zeolites, because of their low cost and selectivity, are important alumino-silicates used in sorption processes [14, 15]. However, it is noteworthy that powder zeolites cannot be used continuously for long-term removal because of the difficulty of powder recovery and the apparent limitations of the adsorption capacity at the powder surface [16]. Therefore, efforts have been undertaken to develop new materials that are suitable for use as radioactive Cs adsorbents.

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On the other hands, consumption of fossil fuels in power plants for energy production is increasing in Fukushima, leaving fly ash waste for radioactive landfills with low level contaminants [17,18]. The action to utilize destruction by fly ash having radioactive contamination effectively becomes the situations such as interruption and the review for recycling to become raw materials of cement and the metal refinement. By the policy of the Japanese country [18], all cesium contaminant levels decide for the thing more than 8000Bq/kg to process undergrounding in the last disposal place established as designated waste in the national land. It is a policy of Ministry of the Environment for less than 8000 Bq/kg that such fly ash has been in the management type disposal ground that after appropriate measures giving leaking, a gross quantity has kept in large quantities by processing facilities without being handled. But the wastes destruction by fired fly ash (with more than 8,000 Bq/kg in radioactivity concentration) as the designated waste as of December 31, 2013 approximately 97,000t (Ministry of the Environment) [19].

Amount of the designated waste, designated waste disposal treatment information site) exist. A technique to separate radioactive cesium from fly ash where the Cs attached includes a washing method. The washing method shows the solution that cesium is high in radioactive fly ash and summarizing a technique in about the removal of the radioactive cesium by the water washing of radioactive fly ash in National Institute for Environmental Studies (NIES) including recovery of radioactive cesium by such zeolite adsorbent [20]. Aqueous wastes that contain long-lived radionuclides require treatment using chemical precipitation, evaporation, filtration, and solvent extraction. Such decontamination process has been recommended by the International Atomic Energy Agency [7].

A designated disposal system is necessary for such wastes. In addition to zeolite, it has been known that extensive research has been conducted related to the adsorption of Cs on various materials with Prussian blue (PB) [21-24]. Therefore, we paid attention to a characteristic of the zeolite and developed the fibrous adsorbent, which was made by composite of zeolite and polymer for decontamination materials in Fukushima [16], for the character comparison in the waste water treated with radioactive fly ash. This report introduced the decontamination behavior of strong alkali waster water contaminating radioactive Cs. The decontamination function to such fly ash waster water contaminating radioactive Cs was shown as a first report by using zeolite polymer composite fibers.

II. MATERIALS AND METHODS

2.1. Materials.

All chemicals and reagents used for this study were of analytical grade purity. They were used without further purification. Natural mordenite zeolite was acquired from Japan Nitto Funka Trading Co. Ltd. (Miyagi, Japan). Poly(ethersulfone) (PES) was used as received (PES, MW = 50 000; BASF Japan Ltd., Ludwigshafen, Germany). Also, *N* methyl- 2-pyrrolidone (NMP; Nacalai Tesque Inc., Kyoto, Japan) was used without purification. Preparation of zeolite composite fibers was conducted using a modified wet spinning process [16, 17]. The zeolite polymer composite fiber composed 30wt% of zeolite in porous polymer fibers. Therefore when the decontamination process of the polluted washing water was carried out, the amounts of zeolite was 6 g relative to the 2g of the composite fibers. PB was product of Kanto Kagaku (Japan). Scanning electron microscope (SEM) of the composite fiber was measured with Hitachi TM3030Plus Tabletop SEM, which was equipped with highly sensitive low-vacuum secondary electron detector with reflective electron images. The secondary electron images were determined for C, S, O, Si and Al components in the cross section and fiber surface.

2.2 Methods

Batch binding experiments were conducted in aqueous solution aliquots (50 mL), each with diluted non-radiative Cs (Nacalai Tesque Inc.) with 20–500 mg/L concentration in the presence of the composite fibers (0.1 g). The original aqueous solution of non-radiative Cs solution was dispersed with the composite fibers and was shaken at 25 ° C in a 50 mL glass container. The incubation time for a saturated binding process was specified as 48 h. The Cs concentration after batch binding experiments was determined using an atomic adsorption spectrophotometer (AA-6300; Shimadzu Corp., Japan). Here, the 852.11 nm emission band for the characterized Cs was detected using an emission lamp (L233–55NB; Hamamatsu Photonics K.K., Japan). The gamma radioactivity of ¹³⁴Cs radionuclide in the tested samples was quantified using a typical nondestructive γ -ray spectroscopic technique with an automated gamma analyzer (WallacWIZARD-1480) with a well type NaI (TI) detector and Ge semi-conductive detector (Segemsmca7600; Seiko easy and G company). The circulation of the radioactive waste water was taken place with Iwaki metering pump (model EHN-B31VC4R, Iwaki co. Ltd, Japan).

III. RESULTS AND DISCUSSION

3.1 RADIOACTIVE CESIUM FLY ASH IN FUKUSHIMA

As followed with the Tohoku earthquake and tsunami of March 11, 2011, the explosions at the Fukushima Daiichi Nuclear Power Plant on March 15 released massive quantities of radionuclides into the atmosphere. It was reported that the total amount of ¹³⁷Cs released into the atmosphere was estimated to be approximately 8.6×10^{15} Bq [25, 26]. After 6 years have passed, still the contamination is seriously influenced to people life in Fukushima area. The radioactive leveled fly ash has been stored now without treatments in the anywhere fields in Fukushima near people living house

(Figure 1). Especially the treatments of radioactive fly ash are problems even though the radioactive amounts are less than 8,000 Bq/kg, which NIES in Japanese government set up guide line for the fly ash contaminated by radioactive cesium [20], because the majority of radioactivity fell on fly ash after burning. However, very little was reported in the decontamination of radioactive waste water used for the radioactive extraction in the fly ash. In the guide line, it has been recommended that the radioactive fly ash should be washed with water [20] to remove the radioactive Cs from the ash and used zeolite. However, little is known about the reports for the radioactive waste water used for the water washing of the fly ash.



Figure 1 Pictures of fly ash and the storage state in Fukushima.

In the present work, fly ash was sampled in Kohriyama city on April 10 2014 and the radioactive Cs measured was 6843 Bq/kg and comparison was made for decontamination of the radioactive waste water by using each PB, zeolite and zeolite polymer composite fiber. Here, the washing process with water was carried out as followed extraction process. The fly ash (200g) was suspended in 2 L water according to the guide line for 6 hours. Then, the suspension water polluted with radioactive Cs was filtrated with paper filter and then was decontaminated with adsorbents. The pH was at 13 after washing and then 500 mL was used to soak with 2g of each adsorbent, PB, zeolite and fibrous zeolite polymer composite. The radioactive waste water polluted with radioactive ¹³⁷Cs was used for following decontamination processes in batch treatment and also column one in following sections. Here, the composite fiber composed of PES and zeolite was prepared by wet spinning process [16, 17]. The fiber diameter was 300 μ m and about 1,000 mm length in each fiber.

Figure 2 shows both reflective electron images and secondary electron images for the composite fiber containing carbon (C) and sulfur (S) for the polymer section and Si, Al and oxygen (O) for zeolite. The pictures of (a) and (h) are reflective electron images of the cross section and surface of the composite fibers and the color images of (b)-(g) are secondary electron images. The latter images suggested that the cross section contained their color components mainly for yellow and pink of C and S, respectively. The Si and Al components (orange colors) were resulted from the loading 30 wt% zeolite powders. On the side surface portion of the fiber, it was also apparent that the zeolite section was found. In our previous report [16] for practical field experiments in Fukushima, the characteristics of zeolite composite fibers was seen as Cs adsorbents. The prepared composite fibers were opaque and

were satisfactorily strong fibers composing of macroporous structures.

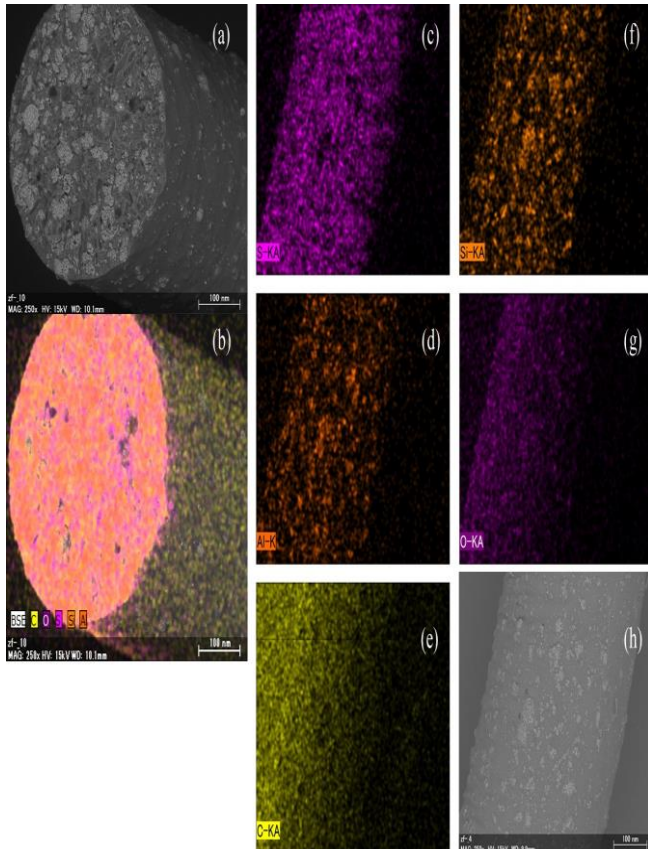


Figure 2 Reflective electron images and secondary electron images of composite fibers.

3.2 REMOVAL OF THE RADIOACTIVE CESIUM FROM RADIOCESIUM CONTAMINATED WASHING WATER FOR RADIO FLY ASH.

Figure 3 shows pictures of the radioactive waste water and PB powders which were used for the decontamination process of the waste water. The color of the radioactive waste water was transparent with pH 13 and the radioactive Cs level was in the range of 2,500-3,000Bq/kg for the radioactive waste water. After the extraction and the contaminated amounts of ^{137}Cs for 120 min, the waste water was changed to yellowish color in the case of PB treatment. As seen in the picture for the PB, the dark black blue powders were changed to be brown color in the waste water, meaning that the PB powders were dissoluble into the aqueous layer during the process and the iron ligand changed to oxidized species having brown color. It was noted that this color leakage to the waste water began, when the decontamination process was started, although the natural water at neutral pH had no such color change for the PB. The comparison indicated that PB was unfavorable to use for the decontamination to the strong alkali solution. On the other hand, there was no color change like such PB experiment in zeolite and the composite fibers after 720 min treatment in the alkali waste water was not changed in the observation of the cross section and the surface of the composite fiber in SEM view.

Before washing After washing for 20 720 min



Figure 3 Pictures of the radioactive waste water and PB powders.

Figure 4 shows time change of the radioactive Cs (Bq/kg) at different decontamination times for PB, zeolite and the composite fiber. As well seen in the PB system, the reduction of the radioactive Cs was reached to 2,000 Bq/kg after 720 min passed, while the zeolite powder system was to 500 Bq/kg. Contrary, the value of the composite fiber system was reached to 500 Bq/kg when the time was 120 min and then gradually decreased to 260 Bq/kg at 720 min, meaning that most of the radioactive Cs was adsorbed to the composite fibers within 120 min. At 720 min, about 90% of the Cs was decontaminated by the composite fibers. Relative to the composite fibers, the value of the PB was 34% in the reduction to the initial Cs concentration and remained brown color solution. This noted that batch experiment as the composite fiber was used effectively decontaminated the radioactive Cs in the strong alkali waste water.

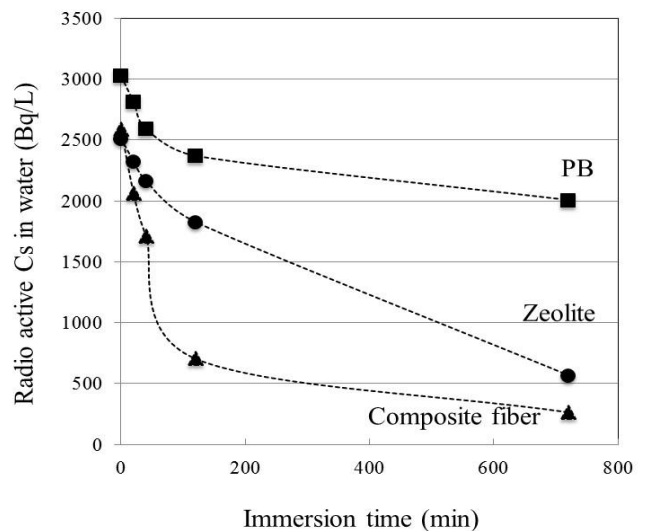


Figure 4 The radioactive Cs concentration change different time for PB, zeolite and the composite fiber.

For the adsorption isotherm, comparison was made in both of zeolite and the composite fiber in the alkali waste water. Here, radioactive Cs fly ash, which was sampled in Naraha town on October 8, 2015 and the radioactive Cs concentration was 3,141 Bq/kg. The extraction of the Cs to the water phase was carried out as mentioned above and the prepared Cs concentration and pH for the radioactive waste water was 823 Bq/L and 12.5, respectively. Different amounts of both adsorbents of zeolite and the composite fiber were used, as the amounts of Zeolite powders or the composite fibers were changed in 0.05g, and 10 g for the alkali waste water. In their conditions, the ratio of the amount of adsorbent to the constant radioactive Cs, 823 Bq/L, was varied in 200 ml at 30 °C. The saturation adsorption was confirmed at 24 h by starting at 80 rpm in the solution and then the resident amounts of radioactive Cs was measured to evaluate the adsorbed amount to the fiber and zeolite. These isotherm results for the zeolite powder and the composite fiber are presented in Figure 5. To elucidate the mechanisms of adsorption and adsorption capacity, adsorption isotherm data were subjected to two sorption isotherms of Langmuir and Freundlich. As seen in (a), the relationship between the adsorbed amounts vs. equilibrium concentration in the range of 50-600 Bq/L showed non-linear in the composite fiber, while the zeolite was linearly in the relation. So the relation was not obeyed in Langmuir. Figure 5 (b) shows Freundlich plots for both cases. The Freundlich equation is an experimental model equation based on a heterogeneous and multilayer adsorption system. Its empirical equation can be used to describe non-ideal adsorption on heterogeneous surfaces and multilayer adsorption, as $q_e = K_F C_e^{(1/n)}$ where q_e stands for the equilibrium adsorption amount [Bq/L], K_F and n , respectively, denote Freundlich constant [-], and C_e signifies equilibrium concentration [Bq/L]. Therein, the Freundlich constant K_F is obtainable by plotting $\ln(q_e)$ against $\ln(C_e)$ (Fig. 5 (b)).

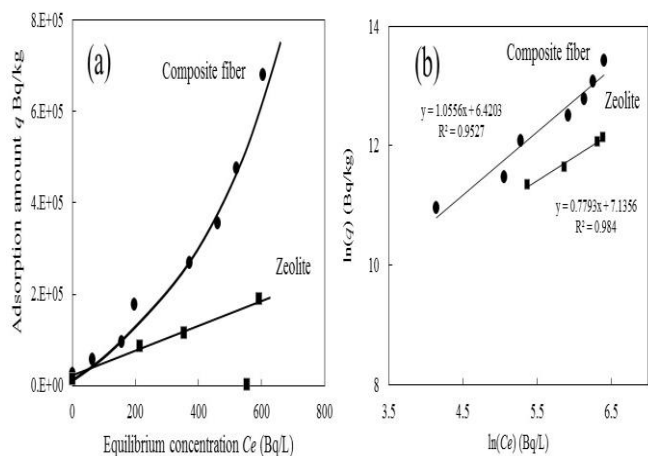


Figure 5 (a) Adsorption isotherm, (b) Freundlich plot of radioactive Cs for composite fiber and zeolite.

Actually, the equilibrium isotherm curves could be compared with non-radioactive Cs in the equilibrium concentration for mmol/L range (Figure 6) for the both adsorbents. The curve well obeyed Langmuir relationship, meaning saturation binding of non-radioactive Cs. Because of the saturation behavior of Cs in both cases, the Cs binding obeyed monolayer binding on the composite fibers and zeolite powders. For the Cs-substrate, the saturation binding amounts

were confirmed respectively as 97 mg/g and 145 mg/g for the zeolite composite fiber and zeolite powder as seen in Figure 6. The comparison results indicated that the composite fibers had somewhat lower saturation on the Cs binding relative to the zeolite powders. This result derived from the PES scaffold coverage by the zeolite powders. Langmuir analysis showed that, for the composite fibers, the adsorbent was mostly able to capture Cs, with maximum adsorption capacity of 139 [mg/g] and respective Langmuir constants of 0.12 L/mg, 149 mg/g, and 0.15 L/mg for the zeolite powders. The zeolite powders exhibited somewhat higher performance than that of the composite fibers.

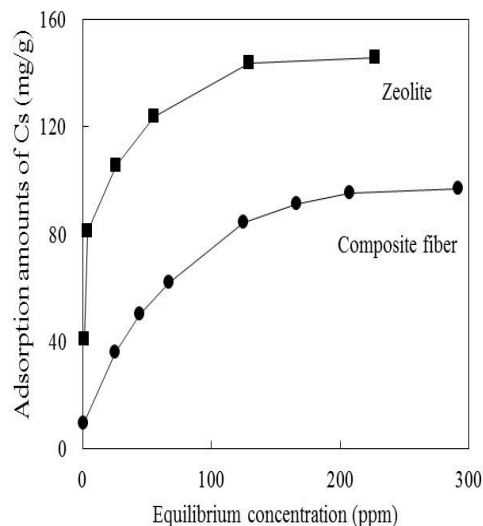


Figure 6 Adsorption isotherm of non-radioactive Cs for composite fiber and zeolite.

The comparison implied that the radioactively contaminated Cs solution containing so much salts of $Ca^{2+}=4,000$ ppm, $Na^+=2,000$ ppm and $K^+=2,000$ ppm, which were interfered the ion exchange mechanism of the zeolite site with radioactive Cs. Here, the value of the radioactive Cs concentration at 600 Bq/L was corresponded to 1.8×10^{-9} mg/L, resulting from very high sensitive detection against the radioactive Cs in extra diluted condition. Therefore, the concentration was very low relative to the present other ions of Ca^{2+} , Na^+ and K^+ in the waste water. This meant that it was very difficult to explain the ion exchange mechanism for the binding of the radioactive Cs in the strong alkali condition in the presence of higher concentration of Ca^{2+} , Na^+ and K^+ . The result of the Freundlich mechanism in the present work suggested that the radioactive Cs in the radioactively contaminated Cs solution prepared from the fly ash suspension was experienced by multilayer binding to the adsorbent sites of zeolite.

3,3 CIRCULATION DECONTAMINATION PROCESS USING COLUMN BINDING TO CONTAMINATED RADIO AQUEOUS SOLUTION

Decontamination of radioactive Cs for fly ash aqueous solution containing radioactive Cs with 823 Bq/L concentration and pH 12.5 was conducted. Figure 7 involves the circulation scheme for the experiment. The composite fibers were filled in a 40cm length column with a 4 cm inner

diameter with different loading % of 10, 38 and 78% for 10 g, 50 g and 135 g in the column volume, respectively. For the composite fibers-column, then the radioactive waste water with 823 Bq/L was passed to flowing by the circulation pump at 200 ml/min. Figure 7 shows radioactive Cs concentration measured at several circulated time for the waste water in the water bath container with 10L volume. After circulation started, the value was decreased with the circulate time within 120 min and then gradually decreased to be 40 Bq/L at 480 min for the case of the 78% loading. This meant that the radioactive Cs was adsorbed to the composite fibers during the circulation of the radioactive waster water. When the comparison was made at different amounts of composite fibers, significant decrease of the radioactivity in the water bath was observed, meaning that lower loading of the composite fibers easily reached to the saturated condition of the zeolite components in the column, while the higher loading column could almost 95 % decontamination by the column treatment at 480 min.

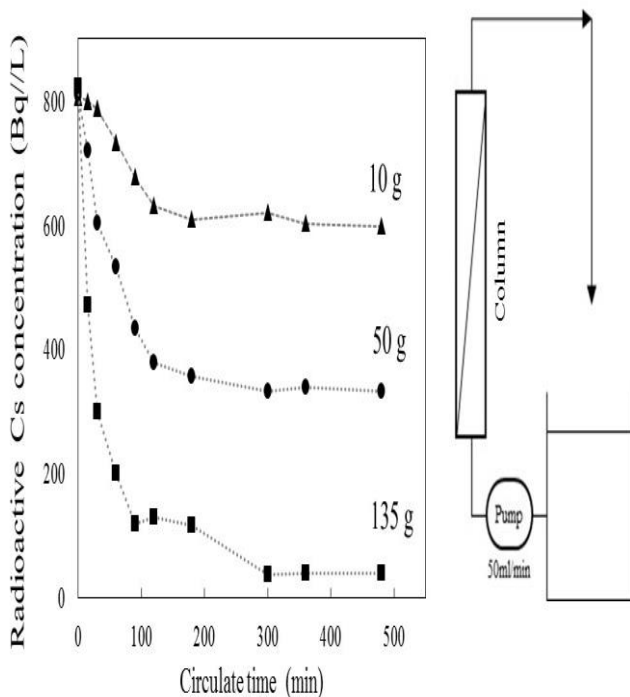


Figure 7 schematic representation of the circulation experiment, and the results.

In Figure 8, similar circulation experiments were carried out at different circulation speeds of the radioactive Cs waste water. Here, the radioactive fly ash was obtained in Iwaki city on 2012, May 2nd. The concentration of radio Cs was 6,560Bq/kg and the resultant radio waste water contained 488 Bq/L. For the decontamination of the Cs by the composite fiber, the loading amounts was 78% in the column. Water flow was changing with 242 mL/min and 121 ml/min for the column permeation. Both flow experiments show reduction of the radioactive Cs in the water bath container, meaning that the composite fiber successfully adsorbed the radioactive Cs in the waste water by column permeation. Relative to 121 mL/min permeation, the 242 mL/min one had efficient reduction of the Cs concentration.

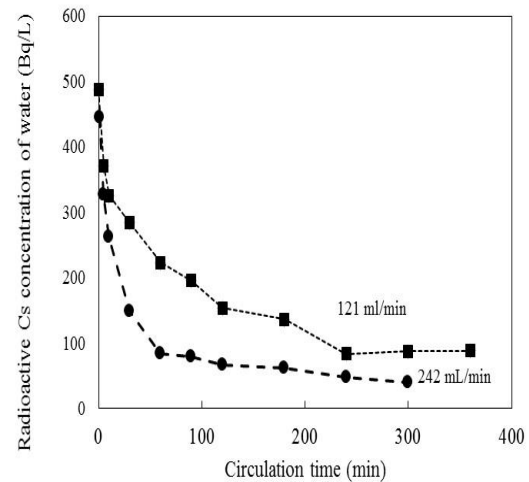


Figure 8 Radioactive Cs removal performance of different flow ratio.

IV. CONCLUSION

Radioactive Cs water wastes obtained for radioactive fly ash decontamination were used in removal tests for the sorption processes of zeolite, zeolite composite fiber and PB. While the strong alkali pH was damaged to PB, zeolite and zeolite composite fiber were behaved reduction of radioactive Cs by the batch and column sorption processes. Relative to zeolite, the composite fiber having 30wt% loading zeolite had excellent character in the decontamination of radioactive Cs. In extra diluted radioactive Cs relative to the batch sorption test for 823 Bq/kg concentration of radioactive Cs solution, the binding was obeyed in Freundlich mechanism with multilayer binding to the adsorbent sites of zeolite.

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