Removal of radiocesium using polyacrylonitrile nanofibers incorporated with Prussian blue

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1. Introduction

After the Fukushima Daiichi Nuclear Power Plant accident in Japan in 2011, the demand drastically increased for efficient technology to remove radiocesium from wastewater due to its hazardous effect on human and environment. Many methods and materials have been introduced to remove Cs from radioactive waste solutions [1-3]. Recently, nanostructured inorganic materials including titanate, vanadate, hexacyanoferrate have been attracted as adsorbents for Cs removal [4]. Many kinds of support materials such as zeolite, graphene and magnetites can be investigated to improve adsorption efficiency and overcome the obstacles of adsorbent application. Polyacrylonitrile nanofibers (PAN) have been used in filtration due to its high chemical resistance and wettability with water. Prussian blue (PB) is easily available in the market and has shown excellent adsorption ability for Cs.

In this study, we incorporated PB into PAN for removal of Cs. The physicochemical properties and adsorption performances of PB/PAN were investigated.

2. Methods and result

2.1 Preparation of PB/PAN

A 10 wt% PAN solution was prepared by dissolving PAN (Sigma-Aldrich) in N,N-dimethyformamide (Sigma-Aldrich). It was heated at 85oC while stirring for 4h, followed by cooling at room temperature, and stirring for 12 h. 5 or 10 wt% of PB powder (Sigma-Aldrich) was added to the PAN solution and stirred for 12 h. The PB/PAN was prepared by electrospinning, with the PB containing the PAN solution being fed at 20 ul/min and 20 kV of the electric field.

2.2 Characterization of PB/PAN

Fig. 1 shows the morphology of PAN, PB(5)/PAN, PB(10)/PAN using a field emission scanning electron microscope (Hitachi S48020, Japan). The PAN nanofiber exhibited an ultrafine and uniform structure, whereas the PB/PAN exhibited a partly nonhomogeneous fibrous structure. The PB nanoparticles with diameters of 20-60 nm are distributed well rather than aggregated in the PAN nanofiber.

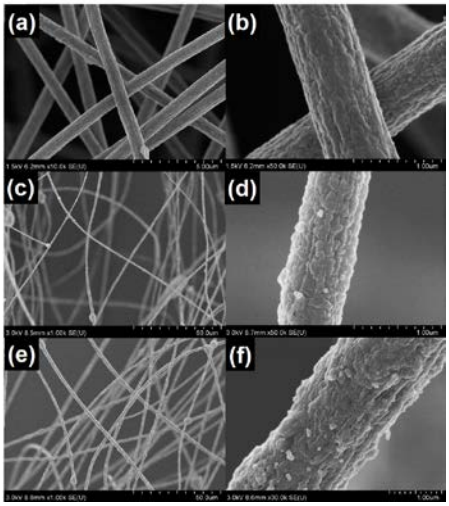

Fig. 1. SEM images of PAN (a,b), PB(5)/PAN (c,d), and PB(10)/PAN (e,f)

In Fig 2, The PAN nanofibers exhibited two peaks, a strong diffraction peak at 16.8° and a weak diffusion diffraction peak at 27.9°. In regarding PB(5)/PAN, PB(10)/PAN, sharp diffraction peaks were observed at 17.4, 24.7, 35.3 and 39.5°. These can be assigned to the planes of PB particles. The intensity of them depends on the amount of PB addition.

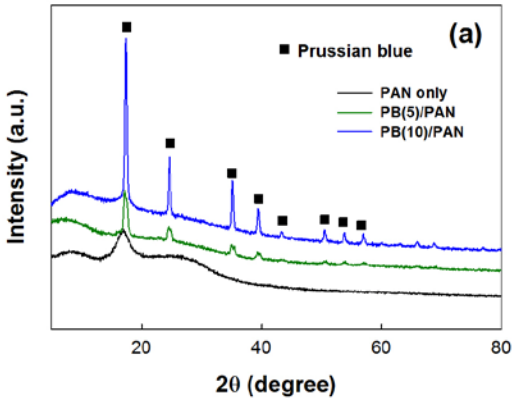

Fig. 2. XRD patterns of PAN, PB(5)/PAN, PB(10)/PAN

Fig. 3 shows N_2 adsorption-desorption isotherms of PAN, PB(5)/PAN and PB(10)/PAN. The PB/PAN nanofibers showed type IV isotherms with type H3 hysteresis, which is a characteristic of a mesoporous structure. PB(10)/PAN has a higher volume adsorbed compared with PB(5)/PAN, which was attributed to an increase in the mesopore content of the fibers by increasing the amount of PB particles.

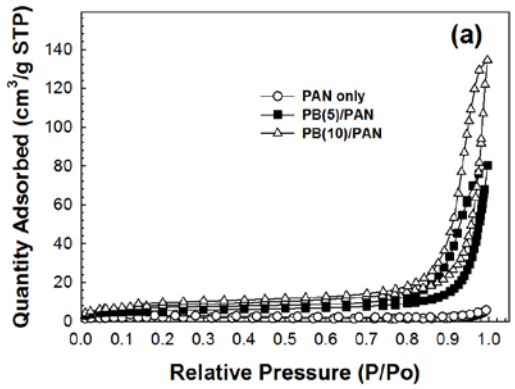

Fig. 3. N_2 adsorption-desorption isotherms of PAN, PB(5)/PAN, PB(10)/PAN

2.3 Cs adsorption on PB/PAN

In Fig. 4, the adsorption capacity increased with increasing equilibrium concentration of Cs^+. The maximum adsoprtion capacity was 0.14 mmol g^{-1}.

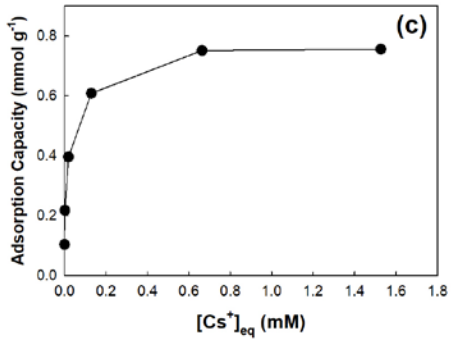

Fig. 4. Adsorption isotherm of Cs ion on PB/PAN

2.4 *Removal of ^{137}Cs by PB/PAN*

The removal efficiency of ^{137}Cs by PB(10)/PAN was tested by varying the contact time. A 10 mL of 1000 Bq L^{-1} of ^{137}Cs was passed through the syringe filter containing 40 mg of PB(10)/PAN, which takes about 10 s. The removal efficiency was 87±3%, despite a short reaction time (Fig. 5). PAN only (40 mg) adsorbed 5±3% of ^{137}Cs. An aliquot of the filtered solution was taken and ^{137}Cs was analyzed by LSC.

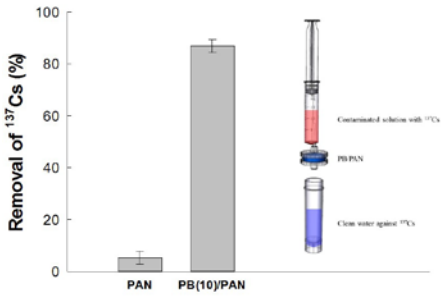

Fig. 5. Removal of ^{137}Cs using PAN and PB(10)PAN for short reaction time (10 s)

The time-dependent removal of ^{137}Cs was tested with 40 mg of PB(5)/PAN and PB(10)/PAN which were soaked in 10 mL of 1000 Bq L^{-1} of ^{137}Cs. The removal of ^{137}Cs increases with increasing reaction time and reaches equilibrium after 1 h (Fig. 6).

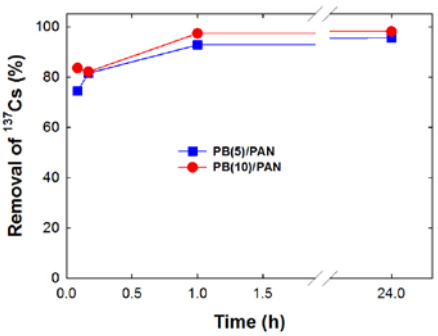

Fig. 6. Removal of ^{137}Cs using PB(5)/PAN and PB(10)/PAN with reaction time

3. Conclusions

The morphology, physicochemical properties and adsorption performances of PB/PAN were investigated. It is confirmed that the PB nanoparticles were successfully incorporated into the PAN without changing their intrinsic structure. In regarding the removal of Cs by PB/PAN, PB(10)/PAN showed high removal efficiency (87 %) within 10 s by simple filtration. PB/PAN can be applied for Cs removal from radioactive waste water due to its high removal efficiency and rapid adsorption.

REFERENCES

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