Removal of Radioactive Cesium Using Prussian Blue-Graphene Oxide Hydrogel Beads in a Fixed-Bed Column System

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1. Introduction

In this study, Prussian blue-graphene oxide (PB-GO) hydrogel beads were synthesized to remove cesium from aqueous solutions. The PB-GO composites were prepared as PB nanoparticles attached to the surface of multi-layered GO sheets. The PVA-alginate hydrogel beads with PB-GO on their cross-linked structure were prepared and packed in a fixed-bed column system. The effect of various parameters, such as initial cesium concentration, flow rate, bed height, and adsorbent size, on the removal of cesium in the fixed-bed column system was investigated. Furthermore, the Thomas, Adams–Bohart, and Yoon–Nelson models were used to analyze the breakthrough curves using non-linear regression methods.

2. Results and discussion

2.1 Characterization of PB-GO composites

The representative TEM images of the vacuum dried PB-GO composite are shown in Fig. 1 (a) and (b). Fig. 1 (a) indicates a uniform dispersion of PB particles embedded in the graphene sheets.

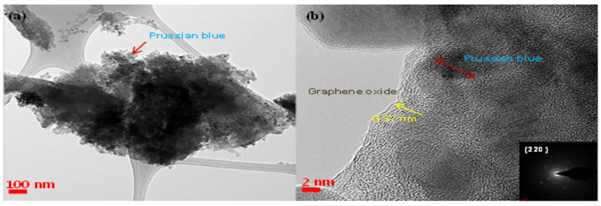

Fig. 1. TEM images of PB-GO hydrogel beads: (a) low resolution, (b) high resolution (insert in (b) is the corresponding SAED pattern of the PB-GO hydrogel beads).

The high resolution TEM image in Fig. 1 (b) clearly shows that the uniform PB nanoparticles were encapsulated in multi-layered GO sheets (3-8 layers) with an interlayer distance of 0.37 nm, which agrees with the lattice spacing between the (220) planes of the PB nanocrystal [1].

2.2 Kinetic studies

2.2.1 Effect of initial cesium concentration

The effect of initial cesium concentration (1, 3, or 5 mM) on the adsorption in the fixed-bed column was studied. As can be seen in Fig. 2 (a), the breakthrough time decreased with increasing initial cesium concentration. At lower initial cesium concentrations, the breakthrough curves were dispersed and the breakthrough rate was slower. Sharper breakthrough curves were obtained as the initial cesium concentration increased. These results demonstrated that the change of the concentration gradient affected the saturation rate and breakthrough time [2].

2.2.2 Effect of flow rate

The breakthrough curves for different flow rates (0.83, 1.67, and 2.49 mL/min) in Fig. 2 (b). The breakthrough curves were steeper at the higher flow rates because cesium ions did not have enough time to make contact with the PB-GO hydrogel beads. Both the breakthrough time and saturation time decreased with an increased flow rate. An increase in flow rate might increase in the movement of the mass transfer zone, which resulted in a decrease in the required time to reach the breakthrough concentration.

2.2.3 Effect of bed height

The effect of bed height (5, 10, or 20 cm) on the breakthrough curve in Fig. 2 (c). Both breakthrough time and saturation time increased with an increase of the bed height. Since an increase in the total amount of the adsorbent provided more adsorption binding sites, a longer time was required to reach the breakthrough and saturation points. Fig. 2 (c) shows that the slope of the breakthrough curve decreased with increasing bed height due to a broadened mass transfer zone, suggesting a slow attainment of the saturation point after achieving breakthrough at a higher bed height.

2.2.4 Effect of adsorbent size

The influence of particle size on the adsorption capacity of the PB-GO hydrogel beads was studied with respect to average particle size (in the range of 2-5 mm). Increasing the adsorbent particle size increased the breakthrough time (Fig. 2 (d)). The adsorption capacity decreased from 161.1 to 130.96 mg/g as the average particle size increased from 2 to 5 mm. Smaller particles have a shorter diffusion path, thus allowing cesium ions to penetrate deeper and more quickly into the adsorbent.

2.3 Dynamic modeling of the breakthrough curves

The breakthrough curves for the adsorption of the PB-GO hydrogel beads were analyzed using the Adams–Bohart, Thomas, and Yoon–Nelson models to determine the dynamic behaviors in the column [3-5].

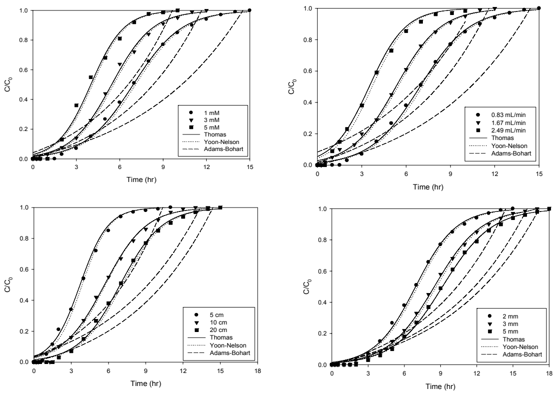

Fig. 2. The Influence of operational parameters on the cesium adsorption breakthrough curves using PB-GO hydrogel beads: (a) influent cesium concentrations, (b) flow rate, (c) bed height, and (d) adsorbent size.

The experimental results were fitted to the models through non-linear regression using Matlab 7.8. To evaluate the fitness of the breakthrough model equations, Chi-square (χ^2) and average percentage errors (APE) were calculated [6,7].

The Thomas model's correlation coefficients are high (R^2=0.991-0998) and its corresponding χ^2 and APE values are low, which indicates that the model is able to describe the dynamic behaviors in the fixed-bed column. Adams–Bohart model is not as appropriate a predictor for the breakthrough curve as the Thomas model.

3. Conclusions

The PB-GO hydrogel beads have a strong potential as an efficient adsorbent for the treatment of wastewater polluted with cesium in continuous operation mode.

4. Acknowledgements

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5. References

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