

## Adsorption of radioactive cesium and applications

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### 1. Introduction

Environmental pollution is one of the greatest problems which causes irreparable damage to the earth [1]. In recent years, water pollution caused by heavy metal ions, organic dyes and radionuclides has become a serious environmental hazard. In order to address global warming and to satisfy the increased demand for energy, nuclear power is being considered as a primary source of power due to its carbon-free energy footprint. However, despite its advantages, the uncontrolled release of radionuclides from nuclear power plants is perceived as a major threat to the environment. Of these radionuclides, cesium ( $^{137}\text{Cs}$ ) poses threat to humans and ecosystem due to its long half-life (30.2 years) and high water solubility [2,3]

### 2. Methods and Results

In this section a Prussian blue (PB)/graphene oxide (GO)/chitosan (CS) organic-inorganic composite was successfully synthesized and utilized as an adsorbent for the selective removal of cesium ( $\text{Cs}^+$ ) ions.

#### 2.1 Characterization of composite

XRD (X-ray diffraction) patterns were obtained using a Bruker D2 PHASER (Germany) diffractometer and  $\text{Cu K}\alpha$  radiation. XPS (X-ray photoelectron spectroscopy) was performed using a Thermo Scientific, K-Alpha electron spectrometer equipped with an Al X-ray source (XPS peaks were fitted with CASA XPS software and binding energies were obtained with respect to C 1s at 284.6 eV). FTIR spectra were obtained using a Jasco FT/IR-6600 unit using KBr pellets from 4000 to 400  $\text{cm}^{-1}$ . Raman spectroscopy was conducted using a FEX Raman spectrometer (FEX, NOST) using a 785 nm laser source. X-ray diffraction patterns of GO, GO/CS and PB/GO/CS are shown in Fig. 1a. The XRD spectrum of GO showed a sharp peak at  $10.8^\circ$ , whereas that of GO/CS showed broad diffraction peaks at  $2\theta$  values of  $8.52^\circ$  and  $18.49^\circ$ . The spectrum of GO/CS did not contain the sharp peak at  $10.8^\circ$  in the spectrum of GO, indicating the exfoliation of GO into the CS matrix. Also, in the GO/CS composite, the  $8.52^\circ$  characteristic peak of GO was shifted from  $10.8^\circ$  due to a d-space increase of GO from 8.18 Å to 10.36 Å, indicating that CS chains were inserted into the structure of GO. Upon adding chitosan,  $11.2^\circ$  peak in the spectrum of GO/CS shifted to  $8.32^\circ$ .

Interestingly, after adding PB to GO/CS, new strong diffraction peaks were observed at  $17.35^\circ$ ,  $24.53^\circ$ ,  $34.97^\circ$  and  $39.99^\circ$  which were putatively attributed to the (200), (220), (222) and (400) planes of PB. The absence of the characteristic  $10.8^\circ$  peak of GO and reduction in the intensities of characteristic GO/CS peaks in PB/GO/CS confirmed the formation of PB/GO/CS.

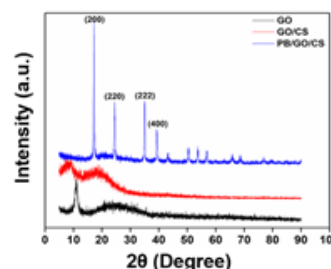


Fig. 1. XRD data of GO, GO/CS and PB/GO/CS.

The FT-IR spectra of GO, GO/CS and PB/GO/CS are shown in Fig. 1b. GO showed characteristic peaks at 3393, 1738, 1626, 1262 and 1408  $\text{cm}^{-1}$  corresponding to stretching vibrations of O–H, C=O, C=C, C–O and COO–, respectively. The spectrum of GO/CS showed N–H stretching vibration at 1552  $\text{cm}^{-1}$ . In PB/GO/CS, the adsorption at 2091  $\text{cm}^{-1}$  showed the presence of the  $-\text{C}\equiv\text{N}-$  group and that at 500  $\text{cm}^{-1}$  was attributed to the formation of  $\text{Fe}^{2+}-\text{CN}-\text{Fe}^{3+}$ , indicating the formation of PB. The peak at 596  $\text{cm}^{-1}$  was putatively attributed to Fe–O stretch.

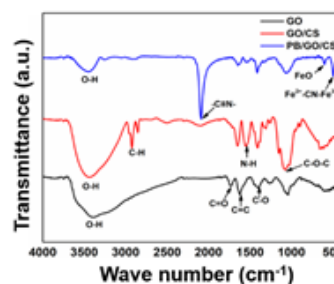


Fig. 2. FT-IR data of GO, GO/CS and PB/GO/CS.

The Raman spectra of GO/CS and PB/GO/CS are shown in Fig. 1c. Both GO/CS and PB/GO/CS displayed the characteristic D and G bands of graphene-based materials at 1336 and 1592  $\text{cm}^{-1}$ , respectively. The ID/G ratio of GO/CS and PB/GO/CS were 1.04 and 1.07, respectively. The incorporation of PB resulted

in a slight increase in ID/IG ratio, which we attributed to perturbation of the GO/CS matrix by PB nanoparticles. Also, the Raman spectrum of PB/GO/CS showed a strong vibrational band at 2149  $\text{cm}^{-1}$ , which was attributed to stretch vibration of the carbon nitrogen triple bond in Prussian blue.

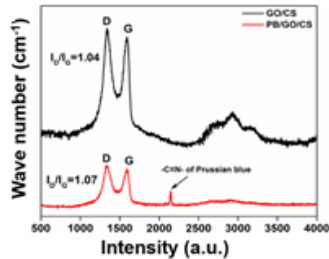


Fig. 3. Raman data of GO, GO/CS and PB/GO/CS.

## 2.2 Adsorption isotherm analysis

Experimental data were fitted to the Langmuir and Freundlich isotherm models. According to the experimental results from  $\text{Cs}^+$  adsorptions by PB/GO/CS and GO/CS, the Freundlich model was a better fit to the adsorption data than the Langmuir model for both systems with a regression correlation coefficient ( $R^2$ ) of 0.99.

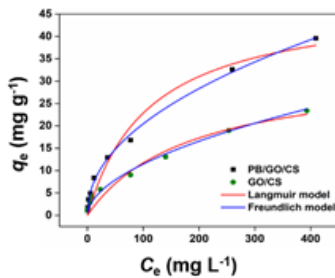


Fig. 4. Langmuir and Freundlich isotherm models of  $\text{Cs}^+$  adsorption by GO/CS and PB/GO/CS.

## 3. Conclusions

In this study, we demonstrated that the design and manufacture of functional Prussian blue (PB) for removal of cesium which means high selectivity of PB towards  $\text{Cs}^+$  ions which forms crystals with a cage size similar to the hydrated radius of  $\text{Cs}^+$  ions. Due to its low-cost, high adsorption capacity, and superior  $\text{Cs}^+$  ions selectivity, Functional PB based adsorbents are a promising material for the selective removal of the  $\text{Cs}^+$  ions from the environment and for protecting ecosystems from the radiation hazards.

## REFERENCES

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