

Gamma radiation-induced synthesis of polyaniline/CuWO₄ nanocomposite for potential sorption of Cobalt-60 and Cesium-137 from aqueous solutions

Mohamed Mohamady Ghobashy^{1*}, M.I.A. Abdel Maksoud², M.I. Aly³, R.S. Hassan³, A. H. Ashour²

¹Radiation Research of Polymer Department, National Centre for Radiation Research and Technology (NCRRT), Egyptian Atomic Energy Authority (EAEA), Cairo, Egypt, ²Radiation Physics Department, National Center for Radiation Research and Technology (NCRRT), Egyptian Atomic Energy Authority (EAEA), Cairo, Egypt, ³Hot Laboratories Centre, Egyptian Atomic Energy Authority (EAEA), Post Code 13759, Cairo, Egypt. Mohamed.ghobashy@eaea.org.eg ID 50#

1. Background and Goal of the present work

Radioactive waste is emitted into the environment as nuclear power stations and laboratories operate more frequently. This has become a major concern for public health, safety, and the environment due to the potential dangers of even tiny levels of radioisotopes. Cs-137, a radionuclide with a half-life of 30.28 years, is a dangerous fission product found in low-level radioactive liquid waste. It is mobile in the final repository and can have a long-term negative impact. Cobalt-60 is not directly produced from the fallout of nuclear fuels, but rather indirectly through the presence of impurities in the stainless steel used to construct nuclear reactors. The release of these radionuclides can occur due to nuclear power plant accidents, nuclear testing sites, and waste from other nuclear facilities. The present work focuses on the synthesis of a nanocomposite of polyaniline/CuWO₄ (PANI/CWO) using gamma radiation. This nanocomposite is capable of adsorbing cobalt-60 and cesium-137 from aqueous solutions.

2. General experimental

2.1. Preparation of CWO NPs: Copper (II) chloride and sodium tungstate dihydrate were obtained from Sigma-Aldrich and used as sources of Cu and W. The co-precipitation technique was used to produce pure CWO NPs. CuCl₂ and Na₂WO₄·2H₂O were diluted in separate beakers with deionized water, mixed together vigorously for 30 minutes, and then centrifuged and rinsed with deionized water. The washed CWO sample was roasted and crushed into powder to obtain pure CWO NPs.

2.2. Preparation of PANI/CWO nanocomposite: The process involves distributing the produced CWO NPs in DMF, homogenizing the mixture using ultrasonic for 30 minutes, and then sonication for 1 hour with the addition of Tween 80. After stirring at room temperature for 1 hour, aniline monomer is added to the reaction mixture and stirred with ultrasonication for half an hour. The resulting mixture is then irradiated at a dose of 50 kGy using Co-60 gamma-cell sources at a dosage rate of 1.1 kGy/h. After filtering and washing with an ethanol/water mix, the final product is dried at 60°C to yield PANI/CWO nanocomposite.

2.3. Reagents and Analysis: The compounds used in the study are of analytical reagent grade (AR) and were obtained from British Drug Houses (BDH) in England. The concentration of hydrogen ions was measured using a digital pH-meter, and the pH reading was adjusted using dilute concentrations of NaOH and HCl. The gamma counter, consisting of a 3-inch NaI (Tl) well-type crystal attached to a Nuclear enterprise model SR5 Scaler-rate meter, was used to analyze the ¹³⁷Cs in the batch study. The solution samples were counted using test tubes. The removal efficiency for metal ions during equilibrium (R%) and the quantity of metal ions adsorbed onto the adsorption sites during equilibrium (q_e in mg/g) were computed using specific relations

$$R\% = \frac{A_0 - AC_e}{A_0} \times 100, \quad q_e = \frac{\sum \%R}{100} \times C_0 \times \frac{V}{m}$$

3. Results and Discussion

3.1. Structural Analysis of PANI/CWO Nanocomposite

XRD analysis in **Figure 1** was conducted to study the structure of the PANI/CWO nanocomposite. The XRD peaks of pure CWO NPs matched well with its anorthic (triclinic) phase, as shown in **Figure 2**. The detected 2θ values and corresponding planes of CWO NPs include 15.16° (010), 19.22° (100), 23.16° (110), 23.75° (0-11), 24.32° (011), 26.13° (101), 28.88° (-1-11), 29.61° (111), and 31.84° (020). The intensity of the diffraction peaks decreased after decorating the CWO NPs with PANI, indicating an interaction between PANI and CWO NPs. The crystallite size of the PANI/CWO nanocomposite was estimated to be 62.67 nm, which is larger than the 52.56 nm size of pure CWO NPs. This increase in size can be attributed to the decoration of PANI on the surface of CWO NPs.

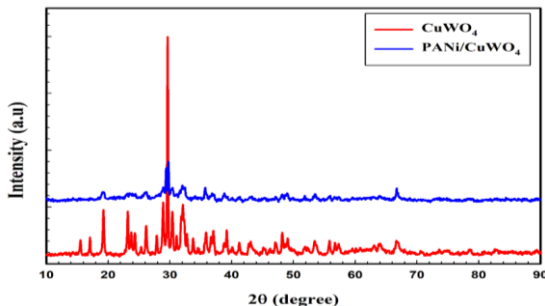


Fig. 1 XRD patterns of PANI/CWO nanocomposite

3.2. Surface Morphology of PANI/CWO Nanocomposite

Fig. 2 shows SEM images of the PANI/CWO nanocomposite, revealing that it consists of CWO NPs decorated by PANI. **Fig. 3** exhibits the elemental mapping images of the PANI/CWO nanocomposite. The mapping images demonstrate that the elements (C, O, N, Cu, and W) have successfully diffused across the nanocomposite without any exogenous elements, indicating the purity of the nanocomposite.

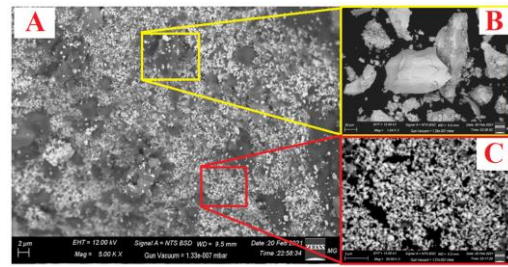


Fig. 2 SEM images of (A) PANI/CWO nanocomposite, (B) CWO NPs, and (C) PANI.

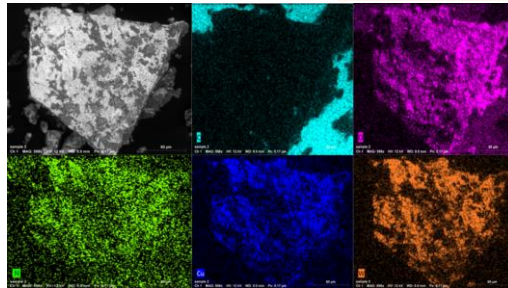


Fig. 3 The elemental mapping images of PANI/CWO nanocomposite.

3.4. Effect of pH and contact time on removal of cobalt-60 and cesium-137

Fig. 4. show the separation factor, S_(Co/Cs), increases with pH for the PANI/CWO nanocomposite adsorbent until pH9. **Fig 5.** represents the removal of cobalt and cesium radioisotope is enhanced gradually as the contact time increases until it remains unchanged after 30 min.

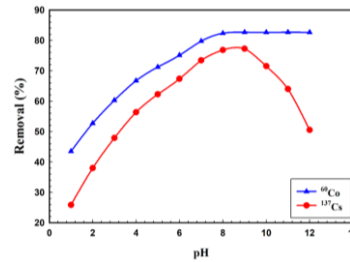


Fig. 4. Impact of pH on removal of cobalt-60 and cesium-137

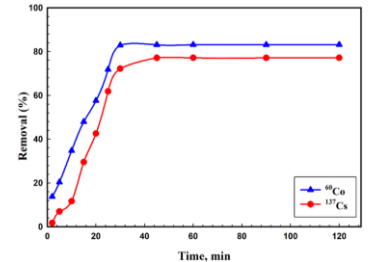


Fig. 5. Effect of contact time on removal of cobalt-60 and cesium-137

3.3. Equilibrium Isotherm models

The non-linear fitting of all models in **Fig. 6** also confirms that the Hills model is more fitted with the experimental values compared to other models. The Hills model suggests that adsorption is a cooperative phenomenon and involves the binding of various species onto homogeneous substrates.

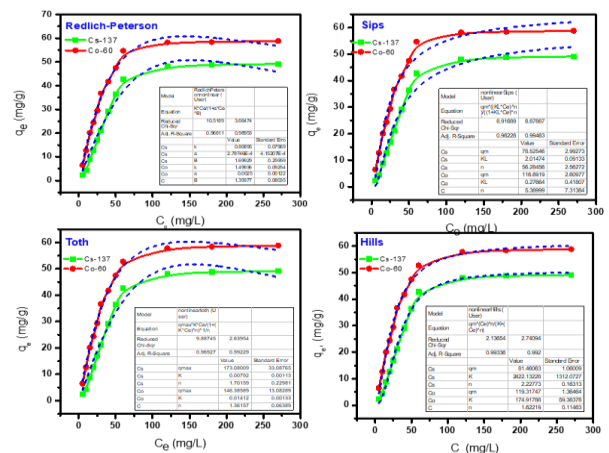


Fig. 6. Isotherm non-linear plots: Redlich-Peterson, Sips, Toth, and Hills for adsorption of ⁶⁰Co and ¹³⁷Cs using PANI/CWO nanocomposite.

4. Conclusions and Acknowledgements

The study aimed to investigate the potential of polyaniline/copper tungstate (PANI/CWO) nanocomposite for removing cobalt-60 and cesium-137 radionuclides from wastewater. Radionuclides like these can be dangerous pollutants released from nuclear power plants and laboratories. Gamma irradiation was used to synthesize the PANI/CWO nanocomposite. Various characterization techniques confirmed the successful preparation of the nanocomposite. The nanocomposite showed good adsorption capacity for both cobalt-60 and cesium-137. Batch adsorption experiments were conducted to determine the optimal conditions and adsorption behavior. Factors like pH, contact time, initial concentration, and temperature were studied. The maximum adsorption capacities were 120 mg/g for cobalt-60 and 80 mg/g for cesium-137. The adsorption process was found to follow the Langmuir isotherm model and second-order kinetic model. Thermodynamic parameters indicated that the adsorption of both radionuclides was exothermic and spontaneous. In summary, the study demonstrated that the PANI/CWO nanocomposite has good potential for removing cobalt-60 and cesium-137 radionuclides from wastewater. The nanocomposite could be useful in the treatment and disposal of radioactive waste.