

# Sorption of Radioactive $^{60}\text{Co}$ and $^{137}\text{Cs}$ in low level radioactive waste onto crosslinked crown ether polymeric composite



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## 1. Background and Goal of the present work

Both radioactive  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  have several hazardous effects on both human health and the environment. To mitigate these effects, proper handling, containment, and disposal of low level radioactive waste are crucial. Crown ethers are cyclic chemical compounds which can play a crucial role for treating radioactive waste due to its ability to selectively bind certain radionuclides, specifically cesium and cobalt. Treatment of radioactive waste is a complex processes in which crown ether cover the part of a broader waste management strategy.

## 2. General experimental, preparation process and characterization

### 2.1. Synthesis of Polymeric Composite based Crown Ether PVP(DMAEMA- 15crown-5)

The copolymerization of Polyvinylpyrrolidone (PVP) with Dimethylaminoethyl methacrylate (DMAEMA) was achieved in a flask with deoxygenated water as reaction medium and N,N'-Methylenebisacrylamide (NMBA) used as a crosslinking agent by gamma radiation-induced polymerization process. Typically, the initial co-monomer concentration mixture PVP 9wt%, DMAEMA 17wt% and NMBA 2wt% were dissolved together, and the mixture was purged with nitrogen, and then 5wt% of 15 crown 5 was added. The total mixture was exposed to ultrasonic homogenizer at 15 min using ice bath and controlled temperature, then exposed to gamma irradiation. The percent conversion was calculated gravimetrically and found to be 96%.

### 2.2. Characterisation of Polymeric composite PVP(DMAEMA- 15crown-5)

FT-IR spectrum of PVP(DMAEMA- 15crown-5) polymeric composite, and SEM micrograph were illustrated in Fig. 1. The characteristic function groups of the prepared material were observed at 3181, 1701, 1443, 1164, and 2950  $\text{cm}^{-1}$  for intermolecular hydrogen bonding between maleamic acid and amide group, C=O, C-C aromatic, dibenzo-18-crown-6, and OH stretching of carboxylic acid. The characteristic morphology of capped crown ether is illustrated in Fig. 1(b).

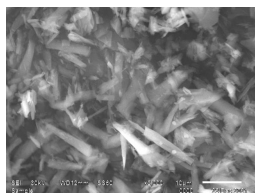
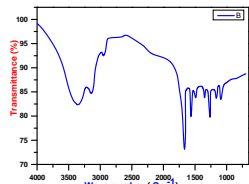


Fig. 1. (a) FTIR spectrum and (b) SEM of the Polymeric composite PVP(DMAEMA- 15crown-5)

### 2.3. Radiological and physicochemical characterisation of the radioactive wastewater sample

The radioactive wastewater sample used in this study is classified as a very high salinity class due to very high electrical conductivity (CF) and total dissolved solids (TDS), which plays important role in decreasing the sorption process (Table 1). The pH of the investigated real radioactive waste sample was 4.5, which is still within the acceptable range for the examined optimal conditions, as shown in the subsections below. The wastewater contains two radionuclides  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ . The obtained gamma-spectrum for the initial concentration of radionuclides in 10 mL waste sample is displayed in Fig.2. The radioactivity level of each component is  $64.84 \pm 8.1$  Bq for  $^{137}\text{Cs}$  and  $444.38 \pm 21.1$  Bq for  $^{60}\text{Co}$ . The activity and efficiency were calculated by the following equation:

$$A(\text{Bq/L}) = \frac{N_{\text{Tot}} - N_{\text{B.G.}}}{I_{\gamma} \times \epsilon \times V \times t} \dots \dots \dots (1)$$

Table 1. International standards for (CF) and (TDS) in distilled, drinking water and liquid radioactive waste

Parameter	Drinking water	Distilled water	Radioactive liquid Waste
TDS(mg/L)	350	182	4305
EC (us/cm)	500	260	6150
TSS(mg/L)	25.67	13.5	311.4
CF	50.0	26.0	615.0

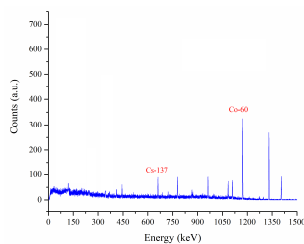


Fig. 2. Gamma spectrum for the radioactive wastewater sample by hyper-pure germanium (HPGe) detector.

## 3. Influencing factors in the treatment of radioactive waste

### 3.1. Effect of contact time

The effect of contact time is essential parameter because it might describe the sorption kinetics of the adsorbent for a specific adsorbate concentration. The effect of contact time was examined for sorption of real radioactive  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  at various time (1–24 h), V/m ratio 0.5 L/g, 24°C and at pH 5.0 for  $^{60}\text{Co}$  and pH 9.0 for  $^{137}\text{Cs}$ . Figure 3 demonstrates a rapid initial rate for sorption of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  (about 85 and 62%, respectively) within the first 2 hours, followed by a slower sorption rate that gradually approached equilibrium after 180 minutes.

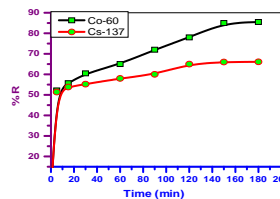


Fig. 3. Effect of contact time on sorption of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  onto PVP(DMAEMA-15crown-5)

### 3.2. Effect of pH

The pH of the aqueous solution is a key parameter for radioactive sorption on the synthesized polymeric composite, as it influences radionuclide speciation and functional groups throughout the sorption process. The sorbent surface charges are varied when the pH of the solution is changed, and the chemical speciation in the solution is dependent on this variable. The effect of pH on the sorption of radionuclides  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  using PVP(DMAEMA- 15crown-5) composite was studied at different pH values as displayed in Fig. 4. The maximum sorption percentage (%R) is obtained at pH 5.0 for  $^{60}\text{Co}$  and pH 9.0 for  $^{137}\text{Cs}$ , therefore, they are selected as the optimum pHs.

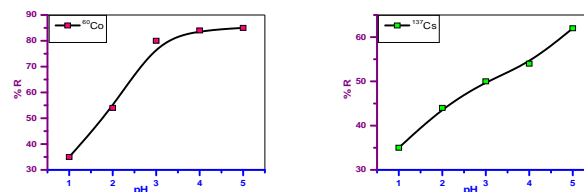


Fig. 4. Effect of pH on the sorption of 100 mg/L of each of  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$  onto 0.02 g Polymeric composite, V=10 ml and time= 2 hours

### 3.3. Effect of metal ions concentration

The mechanism of metal ion sorption is highly dependent on the concentrations of metal ions. The effectiveness of an initial concentration of 100–2000 mg/L on the sorption of a polymeric composite PVP(DMAEMA- 15crown-5) for Cs(I), and Co(II) ions was studied as displayed in Fig. 5. The obtained results indicated that maximum sorption capacities are 44 and 99 mgg-1 for cesium and, cobalt, respectively.

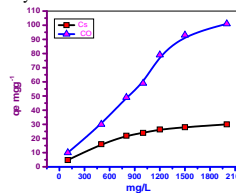


Fig. 5. Effect of metal ion concentration on sorption of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  on PVP(DMAEMA- 15crown-5) composite

### 3.4. Desorption process

The desorption process of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  radionuclides from the PVP(DMAEMA-15crown-5) composite was carried out efficiently using 1.0 M  $\text{HNO}_3$  aqueous solution. Figure 6 demonstrated that within 10 minutes, almost 85% of the studied metal ions was desorbed. It is obvious that desorption of the ions reached a maximum of 97% after 15 minutes. Within 20 minutes, the desorption of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  reached 100%. Depending on the previous results, it could be stated that the PVP(DMAEMA- 15crown-5) composite can be effectively and easily regenerated and re-used for the sorption of radionuclides from an aqueous solution.

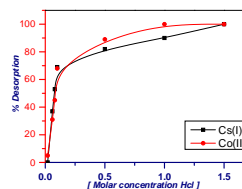


Fig. 6. Desorption process of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  using Nitric acid

## 4. Conclusion and Acknowledgement

In this study PVP(DMAEMA-15crown-5) polymeric composite was successfully prepared using ultrasonic homogenizer for deagglomeration and dispersion then using gamma radiation for free radical polymerization. The formation of polymeric composite was confirmed by various techniques, the various functional groups of the prepared composite were identified by FTIR. The prepared composite was investigated for the removal of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  radionuclides from aqueous solution, and revealed that the PVP(DMAEMA-15crown-5) polymeric composite demonstrated high sorption performance reached to 44 and 99 mgg-1 for cesium and, cobalt, respectively. The desorption process of  $^{60}\text{Co}$ , and  $^{137}\text{Cs}$  radionuclides from the PVP(DMAEMA-15crown-5) composite was carried out efficiently using 1.0 M  $\text{HNO}_3$  aqueous solution.