Sorption of $^{60}$Co(II) from aqueous solutions onto biosynthesized zinc oxide nanocomposites

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Abstract
Nano ZnO is biosynthesized using Lactobacillus sp. Poly Acrylic acid-co-Acrylonitrile/ZnO, PAACAN/ZnO, and poly Acrylic acid-co-Maleic acid/ZnO, PAACMA/ZnO, nanocomposites were synthesized using $^{60}$Co $\gamma$-rays. PAACAN/ZnO and PAACMA/ZnO nanocomposites were characterized and used as sorbents for $^{60}$Co(II) radionuclide. The monolayer adsorption capacities of $^{60}$Co(II) onto PAACMA/ZnO and PAACAN/ZnO are 18.235, 7.105 mgg$^{-1}$, respectively at pH 4.5 and 20 °C. The sorption reaction has a pseudo 2nd order mechanism. 0.1 M EDTA could be used as an eluent for desorption. The desorption efficiencies of $^{60}$Co(II) from PAACAN/ZnO and PAACMA/ZnO were 67.51 and 84.85%, respectively. The sorption process is endothermic and spontaneous.

Keywords Nano ZnO composites · $^{60}$Co radioactive waste · Sorption

Introduction
Radioactive waste is considered as an obstacle for scientists all over the world. It needs special management in order to prevent unnecessary effects on the society and the environment [1]. $^{60}$Co, in concert consider as one of the radioactive wastes generated from nuclear reactors, may be present in trace amounts in the environment [2, 3]. It decays by gamma radiation and has a relatively long half-life ($t_{1/2} = 5.27$ years). External and internal exposure to $^{60}$Co can increase cancer risk. Hence, there is a need for a suitable and economically method to eliminate that exposure.

Adsorption [4] is the simplest, most efficient, and most economical method that could be used for radioactive waste treatment. Selecting a suitable adsorbent for such treatment is the main step in the treatment process [5]. Several adsorbents such as activated and impregnated carbon [6], clay [7], aluminosilicates [8], organic and inorganic gels [9] and others have been reported as adsorbents for radioactive waste. Organic composites have much greater interest than inorganic composites especially in industrial applications because they have a higher capacity than inorganic resin.

PolyAcrylic acid (PAA) is an important water sorption and low cost polymer having ionizable hydrophilic properties. The active functional group COOH is responsible for its sorption properties [10].

Even so, due to the poor mechanical characteristics and thus insufficient physical, chemical, or mechanical bonding of polymer chains, the use of PAA is still restricted, especially in the field of tissue engineering [11]. The implementation of artificial cross linkers such as $N,N'$-methylenbisacrylamide (DAM) for the formation of a polymeric network through the formation of several covalent cross linking points is a common trend for modifying the mechanical properties of PAA. Furthermore, the ease of fabrication, reinforcing techniques based on the incorporation of functionalized nonmaterials as additives or filler materials, such as clay [12], silica nanoparticles [13], carbon nanotubes [14], ferritin particles [15], and graphene oxide [16], the interest of researchers have piqued to prepare nano-composite materials.

Nano-materials have huge applications for wastewater treatment based totally on the adsorption principle. In comparison to bulk particles, nanoparticles have a higher surface area, more adsorption sites, higher adsorption specificity, high permeability, high chemical stability, lower toxicity
and a lower price [17]. The usage of polymer grafted nano metal oxides for the elimination of toxic heavy metals from wastes has continued to attract extensive interest in recent years [18].

Nano ZnO (ZnONPs) is regarded as a good sorbent [19] due to its low cost and non-toxicity. Sol gel techniques [20], wet chemical method [21], green chemistry [22], and microwave-assisted method [23] have all been used to prepare ZnONPs. Chemical and physical methods are quite expensive and require a significant amount of labour and time. The biosynthesis of nano zinc oxide (ZnO NPs) from plants, fungi, and bacteria such as E.coli and Lactobacillus [24] are still a research topic.

Hydrophilic nature as well as good electro-chemical characteristics adequate for adsorption can be influenced through grafting water soluble monomers onto hydrophobic polymers. For grafting modifications, γ-ray and electron beam radiations are widely used as high-energy radiation, and gamma rays are more dominant [25].

In Jiao et al. [26], Poly (acrylic acid-co-maleic acid) was used for the recovery of copper from synthetic aqueous with a yield of 99.84%. Water-soluble transition metal salts were used as catalysts for the polymerization of maleic acid with a water-soluble initiator [27].

This work aims to biosynthesize of ZnO NPs by bacterial culture Lactobacillus sp. for the preparation of two novel nanocomposites; poly (Acrylic acid-co-Acrylonitrile)/ZnO (PAACAN/ZnO) and poly (Acrylic acid-co-Maleic acid)/ZnO (PAACMA/ZnO) by using gamma irradiation of 60Co with a dose of 25 KGy. The prepared nanocomposites used as sorbents for 60Co(II) radionuclide from radioactive waste. PAACAN/ZnO and PAACMA/ZnO nanocomposites characterized by FT-IR, DTA-TGA, SEM, and porosity measurements. pH, contact time, metal ion concentration, ionic strength, and sorbent weight were all studied as influencing factors in the sorption process. The mechanism of the sorption reaction is suggested by evaluating the fit of various kinetic and adsorption isotherm models.

**Experimental**

**Materials and procedure**

**Bacterial strain**

Bacterial cultivation Lactobacillus sp. was obtained from the Microbiology Laboratory, Soil and Water Research Department, Nuclear Research Centre, Atomic Energy Authority, Inshas, Egypt, for the biosynthesis of ZnO NPs. The bacteria were grown on de Man, Rogosa, and Sharp (MRS) agar medium (g L⁻¹) containing meat peptone 10, meat extract 10, yeast extract 5, glucose 20, dipotassium hydrogen phosphate 2, diammonium hydrogen citrate 2, sodium acetate trihydrate 5, magnesium sulphate heptahydrate 0.2, manganese sulphate tetrahydrate 0.05 agar 15, and 1L of dist. water.

**Synthesized of nano ZnO**

*Lactobacillus sp.* was grown on de Man, Rogosa, and Sharp (MRS) broth medium, which holds the immediately preceding mixture medium with no agar. The pH was adjusted to 5.4 before pure culturing of Lactobacillus sp. and incubated aerobically for 24 h at 100 rpm on a rotary shaker at 37 °C. The culture solution was diluted four times with sterile distilled water and allowed to grow for another 24 h. Figure 1a–c. After the incubation stage, the pH was adjusted to 6.0 using 0.4 M NaOH to delay the process of transformation. The mixture was then treated with 0.1 M ZnSO₄·H₂O. The temperature is raised to 80 °C by heating the prepared mixture on water bath. A white precipitate appears at the bottom of flask indicates the hydrolysis of ZnSO₄·H₂O to hydrous zinc oxide [28] according to chemical Eq. (1)

$$\text{Zn}^{2+} + \text{H}_2\text{O} + \text{OH}^- \rightarrow \text{ZnOOH}^- + 2\text{H}^+$$

(1)

The flask was removed from water bath and incubated at 37 °C for 12 h then the particles would get deposited at the bottom of the flask (Fig. 1d, e). The product was accumulated by centrifugation and washed with deionised water followed by drying at 40 °C in a hot air oven for 4 h and calcined at 300 °C/6 h [28]. Hydrolysis of ZnSO₄ to hydrous zinc oxide NPs was demonstrated by UV–Visible spectrum in the range 300–600 (Fig. 1f). It exhibits a strong absorption band at about 355 nm [29] and a monodispersed nature of the nanoparticle distribution.

**Synthesized of PAACAN/ZnO and PAACMA/ZnO nanocomposites**

In Group A, various molar ratios of acrylic acid (AA), acrylonitrile (AN), and calcined nano ZnO were copolymerized in the presence of methylene bis acrylamide (DAM) as a cross-linker to synthesise (PAACAN/ZnO) nanocomposites. In 80 ml deoxygenated water, AA, AN, and calcined ZnO Nps were mixed in the presence of DAM. At room temperature, the mixture is stirred for 4 h. The mixture was subjected to 25 KGY of gamma irradiation at a dose rate of 689.538 Gy/h using a cobalt-60 gamma cell 220 at the EAEA’s cyclotron facility. The preceding steps were repeated to create PAACMA/ZnO nanocomposites, while different molar ratios of AA and maleic acid (MA), as shown in Table 1 (Group B), were copolymerized with calcined nano ZnO and DAM. The materials were cut into small
pieces and soaked in acetone for 2 h to eradicate water and un-reacted monomers before being dried in a vacuum oven at 333 K for 24 h and sieved to size (< 250 µm).

**Instruments**

Investigation of the surface morphology of the nanocomposites carried out by scanning electron microscope (SEM) combined with energy-dispersive X-ray spectroscopy, backscatter diffraction (SEM, Philips XL 30 ESEM (25–30 keV accelerating voltage, 1–2 mm beam diameter and 60–120 s counting time) and The transmission electron micrograph image, TEM, for the particle size was recorded on a TEM, JEM2100, Jeol.s.b, and (Japan). The effective functional groups recognized by Fourier transform infrared spectra (FT-IR) (KBr pellet technique on a Perkin Elmer 1600 FTIR

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**Table 1** Composition of the co-monomers used to prepare PAACAN/ZnO and PAACMA/ZnO nanocomposites

| Group | Sample     | AA (mol%) | AN (mol%) | MA (mol%) | ZnO (mol%) | DAM (mol%) |
|-------|------------|-----------|-----------|-----------|------------|------------|
| A     | P₁AACMA/ZnO| 87.8      | –         | 9.76      | 0          | 2.44       |
|       | P₂AACMA/ZnO| 85.71     | –         | 9.53      | 2.38       | 2.38       |
|       | P₃AACMA/ZnO| 83.72     | –         | 9.30      | 4.65       | 2.33       |
|       | P₄AACMA/ZnO| 81.81     | –         | 9.09      | 6.82       | 2.28       |
|       | P₅AACMA/ZnO| 80        | –         | 8.89      | 8.888      | 2.222      |
| B     | P₁AACAN/ZnO| 58.53     | 39.03     | –         | 0          | 2.44       |
|       | P₂AACAN/ZnO| 57.14     | 38.1      | –         | 2.38       | 2.38       |
|       | P₃AACAN/ZnO| 55.81     | 37.21     | –         | 4.65       | 2.33       |
|       | P₄AACAN/ZnO| 54.54     | 36.36     | –         | 6.82       | 2.28       |
|       | P₅AACAN/ZnO| 53.35     | 35.54     | –         | 8.888      | 2.222      |
Spectrophotometer in wave number range 400–4000 cm⁻¹). The phase structures were determined by (XRD) (Philips X’PERT multipurpose X-ray diffractometer with copper emission lines). Porosity measurements are determined using pore size analyzer 9320. The activity is measured radiometrically by high resolution (7.5%) NaI (TI) detector and the point of zero charge pH (ZPC) is the point at which the surface charge of the sorbent is equal zero. At pH lower than this point, the surface charge is positive and after this point, the surface charge is negative. The efficient sorbents for cations have low value of point of Zero charge because the surface of charge is negative at a wide range of pH. The point of zero charge determined by batch technique using 0.1 g of the nanocomposites with 10 mL of 0.1 M NaCl and the solution shaked for 24 h at 20 °C at different pH range from 1 to 11. Finally, the pH is measured after shaking and the point of Zero charge determined by plotting pH against Δ pH (pHf−pHi). The point intersect the x-axis (pHi) is the point of zero charge [30].

Sorption studies

A stock solution of (Stable Co(II) were prepared by dissolving of amount of cobalt chloride with initial concentration \(C_0 = 100 \text{ mg L}^{-1}\) in double-distilled water and spiked with the \(^{60}\)Co radionuclides \((A_0 = 100 \text{ KBq/L})\). These solutions were kept as stock radioactive solutions for all experiments. The sorption behaviour of \(^{60}\)Co(II) onto (P5AACAN/ZnO) and P5AACAM/ZnO nanocomposites were studied. Variation of the sorption parameters performed to maintain the optimum sorption conditions. i.e. pH (2–8), contacts time (5–4320 min), initial concentration (50–300 mg L⁻¹), Temperature (20–60 °C) and sorbent dose (0.02–0.2 g). Samples were filtered and thus separated from the solution.

The Removal percentage of \(^{60}\)Co(II) radionuclide at each interval of time is given by Eq. (2):

\[
\text{Sorption efficiency } \% = \left( \frac{A_0 - A_e}{A_0} \right) \times 100
\]  

Sorbed amount \(q (\text{mgg}^{-1})\) is calculated using Eq. (3):

\[
q = \left( \frac{A_0 - A_e}{A_0} \right) C_0 \frac{V}{m}
\]

where \(A_0\) and \(A_e\) are the initial and equilibrium concentrations of the \(^{60}\)Co(II) radionuclide, \(m\) is the mass of the P5AACAN/ZnO or P5AACAM/ZnO nanocomposites (g) and \(V\) volume of solution (L). The distribution coefficient \(K_d\) (mLg⁻¹) given by Eq. (4).

\[
K_d = \left( \frac{A_0 - A_e}{A_e} \right) \frac{V}{m}
\]

Kinetic modeling

Three kinetic modelling fitting investigated to suggest a possible mechanism for the sorption reaction; Pseudo 1st order [31], pseudo 2nd order [32] and Elovich model [33, 34].

The pseudo 1st order equation is given by Eq. (5):

\[
\log \left( q_e - q_t \right) = \log q_m - \left( \frac{k_1}{2.303} \right)t
\]

where \(q_e\) and \(q_t\) are the sorbed amounts of \(^{60}\)Co(II) ions; (mgg⁻¹) at equilibrium time and at any time \(t\), respectively; \(k_1\) (min⁻¹) is the pseudo 1st order rate constant.

The pseudo 2nd order is described by the Eq. (6) [32]:

\[
\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e}
\]

where \(k_2\) (gm⁻¹ min⁻¹) is the pseudo 2nd order rate constant.

The surface coverage and activation energy are indicated in Elovich Eq. (7)

\[
q_t = \frac{1}{\beta} \ln \alpha + \frac{1}{\beta} \ln t
\]

where \(\alpha, \beta\) are the Elovich constants. \(\alpha (\text{mgg}^{-1} \text{ min}^{-1})\) is the rate of chemisorptions at zero coverage, whereas \(\beta (\text{gm g}^{-1})\) is the extent of surface coverage and chemisorption activation energy.

Isotherm modeling

Langmuir [35], Freundlich [36] and Temkin isotherm [37] are utilized during this work Langmuir isotherme model applied by Eq. (8)

\[
\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{bQ c_e}
\]

where \(q_{max}\) is the monolayer sorption capacity (mg g⁻¹), \(b\) sorption free energy constant and \(C_e\) is the metal ion concentration at equilibrium. The separation factor calculated from Langmuir constant

\[
R_L = \frac{1}{1 + bC_o}
\]

As the value of \(R_L\) gives an indication that, the isotherm type is linear \((R_L = 1)\), irreversible \((R_L = 0)\), unfavourable \((R_L > 1)\) or favourable \((0 < R_L < 1)\).

The linear Eq. (10) represents Freundlich model

\[
\ln q_e = \ln k + \frac{1}{n} \ln C_e
\]
\[
\log q_e = \log K_f + \frac{1}{n} \log C_e
\]

(10)

where \(K_f\) denotes for Freundlich constants, and \(n\) denotes for sorption capacity and intensity.

Temkin considers that the sorption heat varies linearly with the degree of overlap. The following is the model:

\[
q_e = \frac{RT}{b_T} \ln A_T + \frac{RT}{b_T} \ln C_e
\]

(11)

where \(A_T\) is the maximal binding energy (Lmol\(^{-1}\)), \(b_T\) is the sorption heat, \(R\) is the universal gas constant (8.314 JK\(^{-1}\) mol\(^{-1}\)), and \(T\) is the absolute temperature (K).

**Thermodynamic parameters**

Thermodynamic parameters can be calculated from the temperature dependent adsorption isotherms based on following equations [38] by applying Eqs. (12–15)

\[
K^o_e = K_e \times 1000 \times \text{M.wt of adsorbate} \times \frac{[\text{Adsorbate}]}{\gamma}
\]

(12)

where \(\gamma\) is the coefficient of activity (dimensionless), \([\text{Adsorbate}]^o\) is the standard concentration of the adsorbate (1 mol L\(^{-1}\)) and \(K^o_e\) is the thermodynamic equilibrium constant that is dimensionless, \(K_e\) is the best isotherm model fitted.

\[
\Delta G = -RT \ln K^o_e
\]

(13)

\[
\Delta G = \Delta H - T \Delta S
\]

(14)

The values of other thermodynamic parameters such as \(\Delta H^o\) (kJ mol\(^{-1}\)) and \(\Delta S^o\) (J mol\(^{-1}\) K\(^{-1}\)) have been calculated using the following Eq. (15):

\[
\ln K^o_e = \frac{\Delta S^o}{R} - \frac{\Delta H^o}{RT}
\]

(15)

where \(R\) is universal gas constant 8.314 J mol\(^{-1}\) K\(^{-1}\), and \(T\) is absolute temperature.

**Results and discussions**

**Characterization of the prepared nanocomposite**

**FT-IR analysis**

Figure 2a–c represents the peaks corresponding to the existence functions groups of ZnO nanoparticles. Figure 2a for the spectra and functional group involved in ZnO NPs biosynthesis illustrated many peaks in the range of 500–3000 cm\(^{-1}\). Broad peak obtained at 3377 corresponded to OH stretching vibrations of water molecules. Weak peaks obtained at 2961, 2927, 2858 attributed to stretching vibration of the C–H bond. The sharp peaks obtained at 1649 corresponded to C=C stretch alkene group. Three weak peaks at 1547, 1446, and 1392 were correlated to the carboxylate group. The strong peaks at 945 cm\(^{-1}\) resulted from C–O stretching vibrations.). The band at 673 cm\(^{-1}\) could be related to the binding between sugar monomers. Figure 2a reveals the peak observed at 528 corresponds to the stretching vibrations of ZnO NPs [39].

The FTIR peaks related to P\(_5\)AACAN/ZnO and P\(_5\)AACMA/ZnO nanocomposites (Fig. 2b, c), respectively. The peak assigned at 3444 cm\(^{-1}\) belongs to the absorption O–H group [40] in both nanocomposites but the intensity at P\(_5\)AACMA/ZnO nanocomposites is higher than its counterpart. The peaks at 1636 cm\(^{-1}\) related to C=C stretching vibrations of water molecules. Weak peaks obtained at 2961, 2927, 2858 attributed to stretching vibration of the C–H bond. The sharp peaks obtained at 1649 corresponded to C=C stretch alkene group. Three weak peaks at 1547, 1446, and 1392 were correlated to the carboxylate group. The strong peaks at 945 cm\(^{-1}\) resulted from C–O stretching vibrations.). The band at 673 cm\(^{-1}\) could be related to the binding between sugar monomers. Figure 2a reveals the peak observed at 528 corresponds to the stretching vibrations of ZnO NPs [39].

**Particles morphology**

Figure 3 depicts the particles morphology TEM of nano ZnO and SEM of P\(_5\)AACAN/ZnO and P\(_5\)AACMA/ZnO nanocomposites.
nanocomposites. TEM of Nano ZnO clarified the spherical agglomerated particles of nano ZnO [42], the average particle size is 16.04 nm. The small crystal size of the prepared ZnO is due to preferential erosion of ZnO crystal lattice [43]. The cracking layer surface clarifies the porous structure of P5AACAN/ZnO nanocomposite. The homogenous distribution ZnO is identified [44]. The micrographs taken at 10 µm clearly showed the ZnO incorporation in P5AACAN/ZnO. However, the micrograph of P5AACMA/ZnO nanocomposite shows well dispersed of nano ZnO. Aggregation of nano ZnO particles is noticeable.

**X-ray diffraction (XRD)**

Figure 4 includes the XRD pattern of calcined ZnO nanoparticles, P5AACAN/ZnO and P5AACMA/ZnO nanocomposites. X-ray diffraction peaks of calcined ZnO confirmed that the existing phase is Wurtzite phase and all the crystalline diffraction peaks with hexagonal phase consistent with the reported JCPDS data. The synthesized ZnO nanoparticle diameter was calculated using Debye–Scherrer formula using the diffraction peak intensity of (101):

\[
D = \frac{k\lambda}{\beta \cos \theta}
\]  

where 0.9 is Scherrer’s constant, \(k\) is the wavelength of X-rays (CuK\(_\alpha\)-0.154 nm), \(\theta\) is the Bragg diffraction angle, and \(\beta\) is the full width at half-maximum (FWHM) of the diffraction peak. The parameter calculation for average particle size of ZnO nanoparticle is shown in Table 2. The average particle size of the sample was found to be 25.16 nm. Nevertheless, it is concluded that ZnO is prepared at the nanoscale.

**Porosity measurements**

Table 3 includes the porosity measurements of P5AACAN/ZnO and P5AACMA/ZnO nanocomposites. The results indicate P5AACMA/ZnO nanocomposites had total pore area, porosity more than P5AACAN/ZnO nanocomposite. The average pore diameter (> 50 µm) for the two nanocomposite accounts for the presence of macropores. This
means the cracking of the surface and a predication to higher removal percentage.

**Sorption studies**

The removal percentage (%) and sorbed amounts of nZnO, PAACAN/ZnO, and PAACMA/ZnO nanocomposites towards $^{60}\text{Co(II)}$ are given in Table 4. It is observed that sorption efficiencies (%) and sorption amount of PAACAN/ZnO is less than that of PAACMA/ZnO nanocomposites this is due to the strong electron-withdrawing effect of the nitrile groups may have lowered the relative reactivity of PAACAN/ZnO [45]. However, the ionic carboxylate groups forms hydrogen bond with sorbed $^{60}\text{Co(II)}$ [46] in PAACMA/ZnO nanocomposites result in high removal percentage.

The removal percentage of nZnO toward $^{60}\text{Co(II)}$ is 26.35%. So, addition of different percentage of nZnO to PAACAN and PAACMA increase the removal percentage of the nano composites. Also, by increase mol % of nano ZnO in the two types of prepared composites, the removal percentage increases. This is due to adsorption of ionic polymer over the surface of the nano ZnO oxide particle imparting a high surface charge density and consequently an increase in the electrostatic attraction between nanocomposite and adsorbed $^{60}\text{Co(II)}$ ions [46].

$P_2$AACAN/ZnO and $P_5$AACMA/ZnO show high removal percentage compared with the other compositions of the prepared samples. Thus, the nano-composites of $P_2$AACAN/ZnO and $P_5$AACMA/ZnO selected for the aim of the study. The parameters affecting the sorption reaction as pH, contact time, initial metal ion concentration, ionic strength and temperature is studied.

**Table 3** The sorption efficiency (%) and Sorption amount of $^{60}\text{Co(II)}$ onto different compositions PAACAN/ZnO and PAACMA/ZnO nanocomposites

| Group | Sample        | Sorption efficiency (%) | Sorbed amount, $q_e$ (mg g$^{-1}$) |
|-------|---------------|-------------------------|-------------------------------------|
| A     | $P_1$AACAN/ZnO| 42.22                   | 4.222                               |
|       | $P_2$AACAN/ZnO| 48.75                   | 4.875                               |
|       | $P_3$AACAN/ZnO| 50.75                   | 5.075                               |
|       | $P_4$AACAN/ZnO| 50.75                   | 5.075                               |
|       | $P_5$AACAN/ZnO| 52.69                   | 5.269                               |
| B     | $P_1$AACMA/ZnO| 75.36                   | 7.536                               |
|       | $P_2$AACMA/ZnO| 78.34                   | 7.834                               |
|       | $P_3$AACMA/ZnO| 80.06                   | 8.006                               |
|       | $P_4$AACMA/ZnO| 84.18                   | 8.418                               |
|       | $P_5$AACMA/ZnO| 87.73                   | 8.773                               |
| C     | ZnO           | 26.35                   | 2.635                               |

**Table 4** The calculated kinetic parameters for the sorption of $^{60}\text{Co(II)}$ onto $P_2$AACAN/ZnO and $P_5$AACMA/ZnO nanocomposites [Co of Co(II) 100 mg L$^{-1}$, 25 °C, $V/m=0.1$ Lg$^{-1}$, pH = 4.5]

| Kinetic model                      | $P_5$AACMA/ZnO | $P_2$AACAN/ZnO |
|------------------------------------|----------------|----------------|
| Pseudo-first order                 |                |                |
| $q_e$ (mg/g) (calculated)          | 1.779          | 1.235          |
| $q_e$ (mg/g) (experiment)          | 8.60           | 5.11           |
| $K_1$ (min$^{-1}$)                 | 0.00377        | 0.002625       |
| $R^2$                              | 0.76           | 0.78           |
| Pseudo-second order                |                |                |
| $q_e$ (mg/g) (calculated)          | 7.52           | 4.282          |
| $q_e$ (mg/g) (experiment)          | 8.60           | 5.11           |
| $K_2$ (g/mg min)                   | 0.0123         | 0.0569         |
| $R^2$                              | 0.9985         | 0.999          |
| Film diffusion                     |                |                |
| $K_3$ (mg/g min$^{1/2}$)           | 0.096          | 0.043          |
| $C$ (mg/g)                         | −0.358         | −0.498         |
| $R^2$                              | 0.76           | 0.78           |
| Intraparticle diffusion            |                |                |
| $K_4$ (mg/g min$^{1/2}$)           | 0.096          | 0.043          |
| $C$ (mg/g)                         | 5.64           | 3.59           |
| $R^2$                              | 0.90           | 0.83           |
| Elovich kinetic model              |                |                |
| $\alpha$ (mg/g min)               | 1.383          | 232.73         |
| $\beta$ (g/mg)                    | 76.104         | 208.33         |
| $R^2$                              | 0.80           | 0.89           |

**Impact of [H$^+$] concentration**

Hydrogen ion concentration affects sorption reaction greatly. Studying the impact of the change of pH on the sorption of $^{60}\text{Co}$ onto $P_2$AACAN/ZnO and $P_5$AACMA/ZnO nanocomposites within (2–7.5) is shown in Fig. 5a. The sorption of $^{60}\text{Co}$ is increased by raising the pH values from (2–4.5).

After pH 4.5 the increase in removal percentage belongs to Co (OH)$_2$ precipitate rather than sorption of Co(II) and the differentiation between the amount of Co(II) adsorbed onto the nanocomposite and the precipitated amount become difficult. This is confirmed by the precipitation curve in Fig. 5b and Fig. 5c the speciation diagram of of Co(II) at different pH values in aqueous. It shows that $^{60}\text{Co}$ ions are presented at the optimum pH = 5; confirming that the $^{60}\text{Co}$ ions are sorbed as divalent species and at pH higher than 5; hydroxides started to appears [47]. The [H$^+$] compete $^{60}\text{Co}$ ions for °Ccupying the active sites as the value of pH is more acidic and thus the sorption of $^{60}\text{Co}$ is decreased. Increasing pH value led to a decrease in the competition. Therefore, the optimum pH value was chosen at pH = 4.5.
P5AACMA/ZnO nanocomposites, respectively. The optimum pH was chosen to be 4.5 for the two nanocomposites. The surface charge is positive at pH before 4.01 and 2.8 for P5AACAN/ZnO and P5AACMA/ZnO nanocomposites, respectively. At pH higher than pHZPC, the surface charge is negative so the sorption efficiencies increase after pH 4.01 and 2.8 for P5AACAN/ZnO and P5AACMA/ZnO nanocomposites, respectively. The lowest sorption efficiencies before this point were due to repulsion between surface of nanocomposites and radionuclides in addition to the competition of H+ as discussed previously. The higher sorption efficiencies of P5AACMA/ZnO nanocomposites than P5AACAN/ZnO are in agreement with pHZPC [48].

Impact of time

Figure 6 illustrates the impact of time studied of the sorption of 60Co(II) onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites within the range (5–4320 min). The point charge of P5AACAN/ZnO and P5AACMA/ZnO nanocomposites [Co of Co(II) 100 mgL⁻¹, 20 °C, V/m = 0.1 Lg⁻¹, Time = 24 h].
sorption equilibrium attained after 24 h. The sorbent amount increases gradually till the equilibrium time of the sorption process due to the accessible vacant active sites on the surface of the prepared nano-composites and high concentration of $^{60}$Co(II) at early time. The sorbed amount of $^{60}$Co(II) onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites at equilibrium time are 5.112 and 8.6005 mg g$^{-1}$, respectively.

**Impact of initial metal ion concentration and temperature**

The amounts of $^{60}$Co(II) sorbed onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites as a function of initial ion concentration are given in Fig. 7. This figure shows that the amount sorbed increases by increasing $^{60}$Co(II) metal ion concentration. The removal percentage decrease from 65.93% for $^{60}$Co(II) ion concentration 50 mg L$^{-1}$ to 22.28% with $^{60}$Co(II) ion concentration 300 mg L$^{-1}$ for P5AACAN/ZnO nanocomposites at 293 K and from 88.06% for $^{60}$Co(II) ion concentration 50 mg L$^{-1}$ to 54.26% with $^{60}$Co(II) concentration 300 mg L$^{-1}$ for P5AACMA/ZnO nanocomposite at 293 K. This is due to the fixed number of active sites and at higher concentrations of $^{60}$Co(II), the driving force to P5AACAN/ZnO and P5AACMA/ZnO nanocomposites surface $^{60}$Co(II) interface increases till the active sites become saturated [49, 50]. However, the relation of the amounts of $^{60}$Co(II) sorbed onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites are not linear due to the interactions between the particles [51].

The sorption of $^{60}$Co(II) onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites are affected by temperature. Variation of temperatures from 20 to 60 °C was carried out for the applied concentration range 50–300 mg g$^{-1}$ of $^{60}$Co(II) and pH 4.5. The effect of temperature on $^{60}$Co(II) sorption can be concluded from Fig. 10. The sorbed amount of $^{60}$Co(II) from aqueous solution was 88.05% and 65.932% at 293 °C for 50 mg g$^{-1}$ and increased to 99.3% and 99.60% at 233 K onto P5AACMA/ZnO and P5AACAN/ZnO nanocomposite, respectively. The increase in removal efficiencies with the increase in temperature shows that the sorption processes have endothermic nature.

**Impact of sorbent dose**

Sorbent dose is an important parameter in the sorption process controls on the economics of the process. To examine the effect of sorbent dosage on $^{60}$Co(II) sorption, different weights from 0.02 to 0.2 g of nanocomposites P5AACAN/ZnO and P5AACMA/ZnO were used at temperature of 20 °C and contact time of 24 h for initial $^{60}$Co(II) concentration of 100 mg L$^{-1}$. Experimental results plotted in Fig. 8 showed that the percentage of $^{60}$Co(II) removal increased from 54.2 to 86.4% for P5AACMA/ZnO and from 28.9 to 52.5% when the adsorbent dosage increased from 0.02 to 0.1 g and these behaviour related to the increase of available active sites. At higher dose of sorbent more than 0.1 g, the sorbed amount is nearly unchanged as the number of $^{60}$Co(II) ions in the solutions are not enough to completely combine with all active sites of the prepared nanocomposites [52].

**Impact of ionic strength**

Ionic strength effected the sorption of $^{60}$Co(II) onto P5AACAN/ZnO and P5AACMA/ZnO nanocomposites. The sorption experiment was conducted by equilibrating 0.1 g of the two nanocomposites with 100 mg/L of $^{60}$Co(II) and NaCl electrolytes with concentrations range from [0–102 mol
L$^{-1}$. Figure 9. Indicates that the sorbed amount decreased with increase in ionic strength of the NaCl salt solutions for the two prepared nanocomposites. The decrease in sorbed amount with increase in ionic strength is primarily attributed to the attraction affinity between Co(II) and Cl$^-$ ions and formation of CoCl$_2$. That is, the amount of $^{60}$Co(II) ions remaining in solution for sorption was reduced considerably and hence the low sorption of $^{60}$Co(II) in addition to competition of Na (I) for the sorption sites on the two nanocomposites. By the increase of NaCl concentration from 0 to 0.2 mol L$^{-1}$, the removal percentage decreases from 86.18 to 24.45% and from 51.31 to 14.4% in P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposites, respectively.

**Kinetic modeling**

Figure 10a shows the plot between $t$ with log $q_e$ for $q_e$. From the slope and the intercept of the linear relation, the constant of 1st equation $k_1$ and the capacity of sorption process at equilibrium $q_e$ respectively could be determined. Table 5 lists the values of $q_e$, $k_1$, and the $R^2$ of the $^{60}$Co(II). The straight line proposed the applicable of the pseudo 1st order model to conventional the experimental data during the adsorption process. Furthermore, the values of calculated capacity $q_e$ (cal.) must be resembled the values of the experimental $q_e$ (exp.). From Table 3 the values $q_e$ (cal.) are not in convenient with $q_e$ (exp.). So, it is obvious, the sorption of $^{60}$Co(II) onto the prepared nanocomposite do not fit pseudo 1st model.

Figure 10b shows the linear plot of $t$/$q_t$ versus $t$ for both nanocomposites. The coefficients of 2nd order model were listed in Table 3. The high values of $R^2$ and the plot of $t$/q$_t$ with t elucidate that, the sorption of $^{60}$Co onto P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite regulated by pseudo 2nd order kinetics. Also, the values of $q_e$ (cal.) are resembled the results of $q_e$ (exp.). These proved the sorption are chemisorptions process.

Film diffusion or Intra particle diffusion models fitting to the surface of nano composite are observed in Fig. (10 c, d), where the linear plot of log (1−F) versus $t$ in Fig. 10c has intercept and not equal zero. So, the sorption process is not controlled by film diffusion. The intra particle diffusion model is represented in Fig. 10d. The intra-particle diffusion kinetic model for the sorption process of the $^{60}$Co(II) occurred through three stages. The first stage expresses the diffusion of the $^{60}$Co(II) from the solution to the macro pores or boundary surface of P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite (from 5 to 40 min) and it is very fast stage. The second stage (from 60 to 360 min) designs the gradual sorption on the surface of the P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite or diffusion to micropores of the nanocomposite, which may be the rate-limiting step. The third one (from 1440 to 4320 min) is the equilibrium saturation. Table 5 includes the parameters obtained from the second part of the linear plot.

The plot in Fig. 10e elucidate that the chemical sorption process performed on the heterogeneous surface of P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite and they are successfully confirmed the second-order kinetic. The values of $R^2 \leq 0.8$ assure the confirmation of this explanation.

**Isotherm modelling**

The equilibrium sorption isotherm is fundamentally important in the design of sorption system mechanisms. Isotherm modelling studied at 293, 313 and 333 K. The linear plot of $C$/$q_e$ versus $C_e$ in Fig. 11a indicates the sorption process fitted with Langmuir postulates. Also, the high correlation coefficient, $R^2 = 0.96$, $q_{max}$ determined from the slope of the plot and listed in Table 4. The sorption capacity of $^{60}$Co(II) at 25 °C onto P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite are 18.248 and 7.107 mg g$^{-1}$, respectively. Furthermore, the increase of the sorption capacity with temperature rising confirming the endothermic nature of sorption reaction. The processes are favourable because $0<R_L<1$ for the two nanocomposites. The sorption capacities of $^{60}$Co(II) onto P$_5$AACMA/ZnO nanocomposite were 18.6284 and 18.587 and 18.857 mg g$^{-1}$ at 293, 313 and 333 K, respectively. On the other hand, the sorption capacities of $^{60}$Co(II) onto P$_5$AACAN/ZnO nanocomposite were 7.107, 11.1098 and 15.477 mg g$^{-1}$, respectively at the same temperatures.

Freundlich relation obtained by plotting log $q_e$ against log $C_e$ the straight lines had been obtained as shown in Fig. 11b. The values of Freundlich constants, $n$ and $k$, are calculated from the slope and intercept, respectively, and represented in Table 6. The numerical of $n>1$ suggested that P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposite bind
with a highest strength multiple binding sites of sorbent. The correlation coefficients of Langmuir isotherm model $R^2$ were higher than Freundlich isotherm model, for the two nanocomposites at the applied temperature range, thus the sorption process regulated with Langmuir model.

Applying Eq. (12) for Temkin modelling, the values of the maximum binding energies, $A_T$ were 0.623, 1.730 and 63.806 Lg$^{-1}$ for $P_5$AACMA/ZnO at 293, 313 and 333 K and 1.08, 0.729 and 266.488 for $P_5$AACAN/ZnO nanocomposite, respectively at the same temperatures. Thus, The difference between binding in $^{60}$Co(II) to $P_5$AACMA/ZnO and $P_5$AACAN/ZnO nanocomposite owing to the strong electron-withdrawing effect of the nitrile groups lowered the relative reactivity of $P_5$AACAN/ZnO [45].

**Thermodynamic study**

Applying Eqs. (12–15) in Fig. 12 to calculate the values of $\Delta G^0$ for $^{60}$Co(II) decreased from $-28.705$ kJ mol$^{-1}$ to $-35.965$ kJ on $P_5$AACMA/ZnO nanocomposite and from $-29.445$ to $-36.648$ kJ mol$^{-1}$ on $P_5$AACAN/ZnO coupled with a rise temperature from 293 to 333 K.
The endothermic processes of $^{60}$Co(II) sorption were investigated for the both two nanocomposites. The change of entropies ($\Delta S^0$) were 181.50 and 180.08 kJ mol$^{-1}$ K$^{-1}$ for $^{60}$Co(II) sorption on P$_3$AACMA/ZnO and P$_3$AACAN/ZnO nanocomposites, respectively. The positive values of $\Delta S^0$ suggested that the randomness...
increased at the P5AACMA/ZnO and P5AACAN/ZnO–60Co(II) metal ion interface. Thus, an increase of the surface affinity of the two nanocomposites toward 60Co(II) were presented.

**Desorption studies**

The desorbing of 60Co(II) from P5AACMA/ZnO and P5AACAN/ZnO nanocomposites are vital from an economic point of view. Desorption Experiments (Fig. 13) were performed by batch technique using three different desorbing agents 0.1 M HCl, 0.1 M EDTA, and 0.1 M AlCl3. The order of percentages of Co(II) eluted using the three desorbing agents from P5AACMA/ZnO and P5AACAN/ZnO nanocomposites were 0.1 M EDTA > 0.1 M HCl > 0.1 M AlCl3. HCl is a strong acid completely ionized to H+ that replaces metal ions adsorbed by ion exchange process [53]. AlCl3 also hydrolyse in water to give HCl and Al(OH)3. Desorption with EDTA solution indicated that an increase in desorption efficiency due to the chelating tendency is higher than ion exchange.

**Table 6** Thermodynamic parameters for the sorption of 60Co(II) onto P5AACMA/ZnO and P5AACAN/ZnO nanocomposite

| Nanocomposite          | Δ H kJ mol⁻¹ | ΔS Jmol⁻¹ K⁻¹ | Temperature (K) |
|------------------------|--------------|---------------|-----------------|
|                        |              |               | 293             |
|                        | ΔG kJ mol⁻¹   |               | 313             |
|                        |               |               | 333             |
| P5AACMA/ZnO            | 24.473       | 181.50        | −28.705         |
|                        |              |               | −32.335         |
|                        |              |               | −35.965         |
| P5AACAN/ZnO            | 23.318       | 180.08        | −29.445         |
|                        |              |               | −33.047         |
|                        |              |               | −36.648         |

**Fig. 12** Thermodynamic plot for fitting of 60Co(II) sorption onto P5AACMA/ZnO and P5AACAN/ZnO nanocomposites [C₀ of Co(II) = 100 mg L⁻¹, V/m = 100 mL g⁻¹, pH = 4.5, Eq. time = 24 h]

**Table 7** Comparison of maximum adsorption capacity of adsorbents reported in literature for Co(II) sorption

| Adsorbent                                      | pH | Temperature (°C) | qmax, mg g⁻¹ or mmol g⁻¹ | References |
|------------------------------------------------|----|------------------|--------------------------|------------|
| Date pits                                      | 6  | 25               | 0.052                    | [4]        |
| Sawdust                                        | 0.04                                     |                          |            |
| Mesoporous KZnFC                               | 4.5 | 25               | 10.5                     | [54]       |
| Aluminum silicate                              | 59.31                                    |                          |            |
| Non-living Chlorella vulgaris biomass          | 6  | 5                | 91.99                    | [55]       |
| Aluminum silicate/Chlorella vulgaris composites| 99.24                                    |                          |            |
| TiO₂(20,5)/SiO₂(20,5)                          | 5.5 | 25               | 2.892                    | [56]       |
| Poly-acrylamide based Ce(IV) phosphate         | 2  | 25               | 112.2                    | [57]       |
| Hazelnut shells                                 | 9  | 20               | 13.88                    | [58]       |
| Impregnated polymer [P(AM-AA-AN)-DAM/DtBB18C6] | 5  | 25               | 94.4                     | [59]       |
| Dolomite D-800                                 | –  | –                | 160                      | [60]       |
| I-0                                            | –  |                  | 0.01                     | [61]       |
| M-1                                            | –  |                  | 0.01                     |            |
| P-1                                            | –  |                  | 0.009                    |            |
| P5AACMA/ZnO                                    | 4.5 | 25               | 18.235                   | This study |
| P5AACAN/ZnO                                    |    |                  | 7.105                    |            |
Mechanism of the sorption reaction

A suggested mechanism dominating the polymerization and sorption of $^{60}$Co(II) onto the two prepared nanocomposites, Fig. 14. The main functional groups contributing in the sorption reaction. It can be noted that the sorption of $^{60}$Co(II) onto $P_5$AACMA/ZnO nanocomposite is more efficient than that onto its counterpart $P_5$AACAN/ZnO nanocomposite. This is due to the amount of adsorption sites of $P_5$AACMA/ZnO nanocomposite is higher than that on the $P_5$AACAN/ZnO nanocomposite. In addition to the strong electron-withdrawing effect of the nitrile groups stabilize negative charge of the carboxylate anion [44] and consequently, lowered the affinity of $P_5$AACAN/ZnO towards $^{60}$Co(II). Ion exchange is the main mechanism for the sorption reaction.

Comparative studies

$P_5$AACMA/ZnO and $P_5$AACAN/ZnO nanocomposites sorption capacity is compared with the sorption capacity of other adsorbents [4, 54–61] towards Co(II) and listed in Table 7. $P_5$AACMA/ZnO and $P_5$AACAN/ZnO nanocomposites have reasonable sorption efficiencies and could be used as a host for $^{60}$Co(II) radioactive waste.

Conclusion

Nano ZnO was successfully prepared using the bacterial culture Lactobacillus sp. Poly acrylic acid-co-maleic acid / ZnO nanocomposites or Poly acrylic acid-co-acrylonitrile / ZnO) nanocomposite were synthesized using different concentration of acrylic acid, maleic acid or acrylonitrile monomers and calcined ZnO nanoparticles via free radical polymerization using gamma irradiation of $^{60}$Co at a dose of 25 KGY. The study showed that the optimized nanocomposites $P_5$AACMA/ZnO and $P_5$AACAN/ZnO nanocomposites are efficient sorbents and recommended for the sorption of $^{60}$Co(II) the fission product. The sorption reaction was regulated with Langmuir isotherm model and the monolayer
adsorption capacities of $^{60}$Co(II) onto P$_5$AACMA/ZnO and P$_5$AACAN/ZnO were 18.235, 7.105 mgg$^{-1}$, respectively at pH 4.5 and 20 °C. The sorption process followed pseudo 2nd order kinetic mechanism. The sorption process was endothermic and spontaneous. Desorption of $^{60}$Co(II) from P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposites were performed using 0.1 M HCl, 0.1 M EDTA and 0.1 M AlCl$_3$. The maximum desorption efficiencies of 60Co(II) from P$_5$AACMA/ZnO and P$_5$AACAN/ZnO were 67.51 and 84.85%, respectively. Thus, P$_5$AACMA/ZnO and P$_5$AACAN/ZnO nanocomposites could be cost efficient alternative to radioactive waste management.

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