



Sorption behavior of ^{137}Cs , $^{152+154}\text{Eu}$ and ^{131}Ba from aqueous solutions using inorganic sorbent loaded on talc

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Abstract

Iron phosphate@talc (IPT) sorbent was fabricated by precipitation method and utilized for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorption from aqueous media. Different analytical apparatuses such as XRD, FT-IR, SEM, and XRF were used to find the structure, morphology, and functional groups of IPT sorbent. Different parameters like shaking time, pH, temperature, and initial metal concentrations were studied. The obtained data reveal that the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba was pH, time-dependence, and kinetics following pseudo-2nd-order kinetics. The Langmuir and Freundlich equations were used as model isotherms. Thermodynamics parameters were calculated to prove that the sorption reaction was spontaneous and endothermic.

Keywords ^{137}Cs · $^{152+154}\text{Eu}$ · ^{131}Ba · Talc · Inorganic sorbent · Iron phosphate

Introduction

Because of modern science and technology development, nuclear energy is now widely used for civilian purposes in fields including industry, agriculture, medicine, and scientific research. Moreover, the nuclear industry is rapidly expanding and provides a significant amount of solid, liquid, and gaseous radioactive waste, which should be treated properly before being adequately disposed of [1]. These radioactive waste forms are classified into low, intermediate, and high levels, with liquid radioactive waste representing a large

segment of the total [2]. Radionuclides in liquid radioactive waste are normally categorized into two categories fission products (such as ^{137}Cs , ^{90}Sr , $^{152+154+155}\text{Eu}$, ^{133}Ba , etc.) and activation products (such as ^{63}Ni , ^{60}Co , ^{56}Fe) [3]. The most problematic constituents in radioactive liquid wastes are barium, europium, strontium, and cesium because they are long-lived radionuclides [4, 5]. Due to this, it is extremely important to get eliminate them from liquid waste streams before they are discharged into the environment. However, some beneficial radionuclides are recovered for use in various applications, such as creating a $^{137}\text{Cs}/^{137\text{m}}\text{Ba}$ radioisotope generator used in industry and medicine for quality control and the production of sealed sources employing $^{152+154}\text{Eu}$ for use in industry and health [3, 6]. Ion exchange [7, 8], chemical precipitation [9], membrane separation [10, 11], solvent extraction [12], and adsorption [13, 14] are examples of techniques and methodologies that have been developed for the treatment of aqueous radioactive waste. Adsorption is an efficient and cost-effective technique to concentrate and remove contaminants from the aqueous phases. The contaminants are recovered from the aqueous solutions and bound in artificial or natural adsorbent, where they can be properly disposed off or retrieved [15].

Silicate clay, a naturally occurring nanoparticle adsorbent produced by silicate weathering on the earth's surface, is present in deposits totaling more than 50 million tons worldwide. Owing to its negatively charged surface, large specific surface area, robust cation exchange, and chemical

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pollution-free properties, clay minerals have become one of the most studied activations, modification, and functionalization topics among many scientists [16].

A typical silicate clay mineral with significant deposits and high magnesium content is talc (T) having the chemical formula of $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$. Talc is distinguished by its 2:1 sheets structure, which consists of a layer of $\text{Mg}(\text{OH})_2$ and two layers of the Si–O tetrahedron [17]. After being broken up into talc powder and processed, the talc powder is separated. It is widely used in a variety of industries, including ceramics, paper, plastic, rubber, food, medicine, and food processing [18]. Talc's high porosity, substantial specific surface area, rod-like structure, and the profusion of hydrophilic Si–OH make it an excellent adsorbent for the decontamination of radioactive wastewater [19–21]. Talc composites have been enhanced with other substances including P(AA-AN)-talc [17], $\text{Fe}_3\text{O}_4/\text{Talc}$ [22], and modified talcum [23]. Investigators haven't looked into the impregnation of talc layers with iron ions or phosphate groups. In this study, iron phosphate@talc (IPT) was developed as a novel substance and used to filter out ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba from radioactive wastewater. Using sorption techniques, the sorption efficiency (% S.E.) of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba from liquid waste was investigated [24, 25].

The purpose of this research is to reduce the negative effects of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba by applying the sorption technique to IPT sorbent prepared by the precipitation method. Fourier-transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), X-ray diffraction (XRD), and X-ray fluorescence (XRF) were used as analytical techniques to characterize IPT sorbent. Batch experiments were used to calculate the optimal sorption parameters for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba , which include time, pH, temperature, and initial metal concentration.

Experimental

Materials

The suppliers of BaCl_2 and FeCl_3 are Loba Chemie (India), KH_2PO_4 Alpha Chemika (India), $\text{Na}_5\text{P}_3\text{O}_{10}$ Goway (China), and HNO_3 Merck (Germany). NaOH , NH_3 , and NaCl El-Nasr Company (Egypt). For all works, double-distilled water

(DDW) was used. The reagents employed in this study were of the analytical grade without any further purification.

Radioisotopes tracers

Radioactive tracers ^{131}Ba and $^{152+154}\text{Eu}$ were obtained via the activation reaction of (n, γ) by neutron irradiation of 0.05 g weight of metal chloride salts wrapped in double aluminum foil and goes to the vertical irradiation channels inside the core of the second Egyptian Training Research Reactor ETRR-2 for 3 h at a neutron flux of $1 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ to obtain the desired radioactivity level. Then, they were left to cool for a week before dissolving and counting using the gamma spectrometry technique, to allow all undesirable short-lived radionuclides to decay. ^{137}Cs was obtained from a standard radioactive solution purchased from Eckert and Ziegler company with an initial activity of $3.7 \times 10^6 \text{ Bq}$ ($\approx 100 \mu\text{Ci}$).

Preparation

For the preparation of talc (T), talc phosphate (TP), and iron phosphate@talc (IPT) sorbents, a simple and ambient precipitation procedure was used through the final stage.

- (i) Preparation of talc solution by addition of 7.5 g $\text{Na}_5\text{P}_3\text{O}_{10}$ as dispersing agent to 75 g talc powder with the addition of DDW to reach 750 mL with constant stirring for 120 min.
- (ii) Preparation of 0.2 M KH_2PO_4 and 0.2 M FeCl_3 solutions by dissolving 14.2 g KH_2PO_4 with 500 mL DDW and 8.11 g FeCl_3 with 250 mL DDW.
- (iii) Different ratios of the equimolar solutions from FeCl_3 and KH_2PO_4 were added to the talc solution as the following (0:0:1, 1:0:1, 1:1:1) respectively, details are listed in Table 1.

Adding the prepared solutions together and stirring consistently for 2 h, adding 10% NH_4OH solution to the mixture, as observed a color gel began to form at a pH of 7.5. The formed gel was left undisturbed overnight before being washed with DDW, dried at $60 \pm 1 \text{ }^\circ\text{C}$, and finally ground to get a fine powder.

Table 1 Conditions for the synthesis of T, TP, and IPT sorbents and % S.E. of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba [$C_i = 50 \text{ mg L}^{-1}$, $V/m = 0.1 \text{ L g}^{-1}$, and shaking time 24 h]

Sorbents	FeCl_3 , [mL]	KH_2PO_4 , [mL]	Talc, [mL]	^{131}Ba		^{137}Cs		$^{152+154}\text{Eu}$	
				pH	% S.E.	pH	% S.E.	pH	% S.E.
T	0	0	250	6.0	66.5	6.0	49.85	4.0	41.8
TP	0	250	250		75.31		61.2		56.6
IPT	250	250	250		96.22		83.1		80.9

Preliminary studies for sorbents selection

To perform the measurements, 5 mL each of radioactive solutions of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba were added to 0.05 g of three different prepared samples and shaken at 298 ± 1 K in a thermostat shaker (Kottermann D-1362, Germany). The solution and the solid are decanted as soon as the shaker is turned off after 24 h.

The activities of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba were measured for 3600 s using a coaxial p-type HPGe detector (GEM-series, ORTEC, USA) connected to a multichannel analyzer system (MCA, Inspector 2000 Series, Canberra, USA). The % sorption efficiency (% S.E.) can be computed by using Eq. (1) [26, 27];

$$\% \text{ S.E.} = \left(\frac{A_i - A_f}{A_i} \right) 100 \quad (1)$$

A_i and A_f are the initial and final activity of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba , respectively.

Instruments for sorbent characterization

Germany's Bruker XRD diffractometer D2 Phaser II was used to examine the crystal structure of the IPT sorbent. FT-IR measurements were made in the wavenumber range of 4000–400 cm^{-1} using an automated spectrophotometer Alpha II Bruker (Germany). The Philips sequential X-ray spectrometer-2400 was used to identify the elemental composition of IPT sorbent. Using the Super-Q quantitative application program, the percentages of Mg, Al, Si, P, Ca, K, and Fe were estimated. SEM JSM-6510A Model (Japan) was used to study the surface morphology of the IPT sorbent material.

Chemical stability

Different solvents such as (DDW, ethanol, methyl ethyl ketone, HNO_3 , and NaOH) were used to study the solubility percentage of the IPT by constant shaking 0.1 g of solid and 10 mL of studied solvents for 3 days at 25 ± 1 °C. The amount of IPT sorbent left in the solution was measured gravimetrically by decantation of filtrate and then dried [24, 28].

pH titration

IPT sorbent was examined using the Topp and Pepper method to detect pH titration by the NaOH – NaCl system [29]. Each 10.0 mL of NaOH (0.1 M)– NaCl (0.1 M)

mixed by different volumetric ratios was contacted with 0.1 g of the prepared sorbent and then constantly stirred. After a standard 24 h interval, the pH of the solutions was recorded using a pH meter.

Sorption experimentation

All equilibrium measurements were carried out by shaking 0.05 g of IPT sorbent with 5.0 mL of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba at a baseline concentration of 50 mg L^{-1} in a shaker thermostat model Kottermann D-1362 (Germany), and $V/m = 100$ mL g^{-1} . The variation in sorption parameters such as pH (1–7), metal concentrations (50–500 mg L^{-1}), temperatures (30, 45, and 60 °C), and contacting time (5–300 min) were checked to get the optimum sorption conditions. IPT sorbent batch-wise interacted with ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba . Following sorption, the samples were separated by filtration. The amount sorbed q_e was calculated using the following equation [30–32]:

$$q_e (\text{mg g}^{-1}) = \left(\frac{A_i - A_e}{A_i} \right) \left(\frac{C_i V}{m} \right) \quad (2)$$

A_e , V , C_i , and m are the activity of a solution at equilibrium, the solution volume (L), initial concentration, and the IPT weight (g), respectively. The distribution coefficients K_d and separation factors α_B^A as a function of pH were determined with the subsequent equations [31, 33–35]:

$$K_d (\text{mL g}^{-1}) = \left(\frac{A_i - A_f}{A_f} \right) \left(\frac{V}{m} \right) \quad (3)$$

and

$$\alpha_B^A = \frac{K_d(A)}{K_d(B)} \quad (4)$$

V is the volume of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba (mL). $K_d(A)$, and $K_d(B)$ are the distribution coefficients for the two competing species A and B in the system.

Results and discussion

Preliminary studies

The sorption efficiency (% S.E.) of the studied radiotracer either onto talc, TP, and IPT sorbents is presented in Table 1. These results prove that an enhancement was attained in the sorption behavior of ^{137}Cs , $^{152+154}\text{Eu}$, and

^{131}Ba onto IPT sorbent than T and TP sorbents. So, IPT sorbent was used for all experiments.

XRD analysis

XRD was utilized to indicate the degree of crystallinity of IPT sorbent as presented in Fig. 1a. Figure 1a reveals that IPT sorbent has a uniform hexagonal crystalline nature and demonstrates the existence of characteristic peaks of talc addressed at 2θ values (8.33° , 14.46° , 16.71° , 19.86° , 24.65° , 25.18° , 28.69° , 35.35° , 36.45° , 39.4° , 42.6° , 45.22° , 55.1° , 58.12° , 60.45° , 66.3° , 67.99° , 73.3° , and 79.3°) related to Miller indices of hkl values (100, 2–10, 200, 101, 201, 300, 3–21, 4–31, 002, 2–12, 500, 302, 601, 2–13, 8–40, 8–61, 403, 8–52, and 2–14) respectively. Peaks of iron and phosphorus are significantly appeared at 2θ values (10.82° , 11.96° , 14.92° , 22.93° , 58.66° , and 67.24°). Moreover, some impurities are detected from Ca, Al, and K at 2θ values (9.57° , 12.61° , 18.91° , 19.54° , 25.19° , 26.79° , 28.02° , 28.75° , 34.72° , 35.37° , 36.79° , 55.31° , 60.18° , 61.60° , and 79.42°). This result gives a good estimation of the existence of iron and phosphorus in the present work-prepared IPT sorbent [36].

FT-IR spectrum

FT-IR analysis of IPT sorbent was existing in Fig. 1b. It is clear that from this Figure eight bands appeared in IPT

sorbent at wavenumbers (3749 , 3621 , 1630 , 1049 , 941 , 873 , 745 , and 577 cm^{-1}). The bands at 3621 and 947 cm^{-1} are due to Al–Al–OH (stretching and bending vibration, respectively) [37, 38]. The band at 3749 cm^{-1} is related to Al–OH–Mg bonds present in talc powder [39]. The bands at 3621 and 1639 cm^{-1} agreed to OH frequencies of the H_2O molecule (stretching and bending) respectively [24] or officially to P–OH [40, 41]. At the same time, bands at (1049 , 873 , and 745 cm^{-1}) are due to Si–O, Si–O–Al, and Si–O–Mg bending, respectively [37]. Bands from 941 and 577 cm^{-1} are related to M–O (where M is Al or Fe) [40]. The FT-IR analysis of IPT sorbent loaded by Cs(I), Ba(II), and Eu(III) ions presented in Fig. 1b has the same bands observed in the FT-IR of IPT sorbent. The band at 577 cm^{-1} shifted to 640 cm^{-1} for IPT loaded by Cs(I), shifted to 586 cm^{-1} for IPT loaded by Ba(II), and shifted to 660 cm^{-1} for IPT loaded by Eu(III). Whereas the intensity of bands at 3621 and 1630 cm^{-1} was increased after loading of IPT sorbent with Cs(I), Ba(II), and Eu(III) ions. These data confirmed that the combination between Cs(I), Ba(II), and Eu(III) ions with IPT sorbent was achieved.

SEM analysis

SEM images of the proposed sorbent material are shown in Fig. 2 at different magnification powers of X500, X1000, and X2000. Results show a heterogeneous distribution of the iron and phosphate particles (white color) on the talc

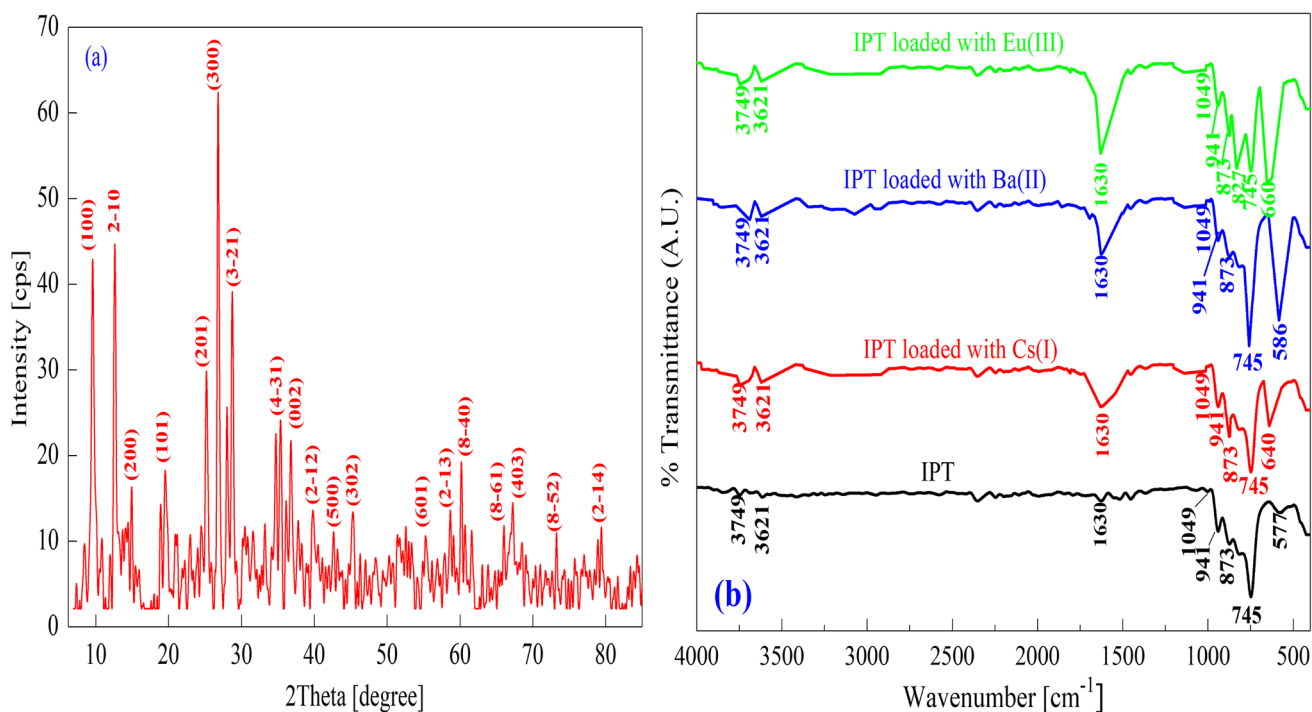


Fig. 1 a XRD pattern and b FT-IR spectrum of IPT sorbent

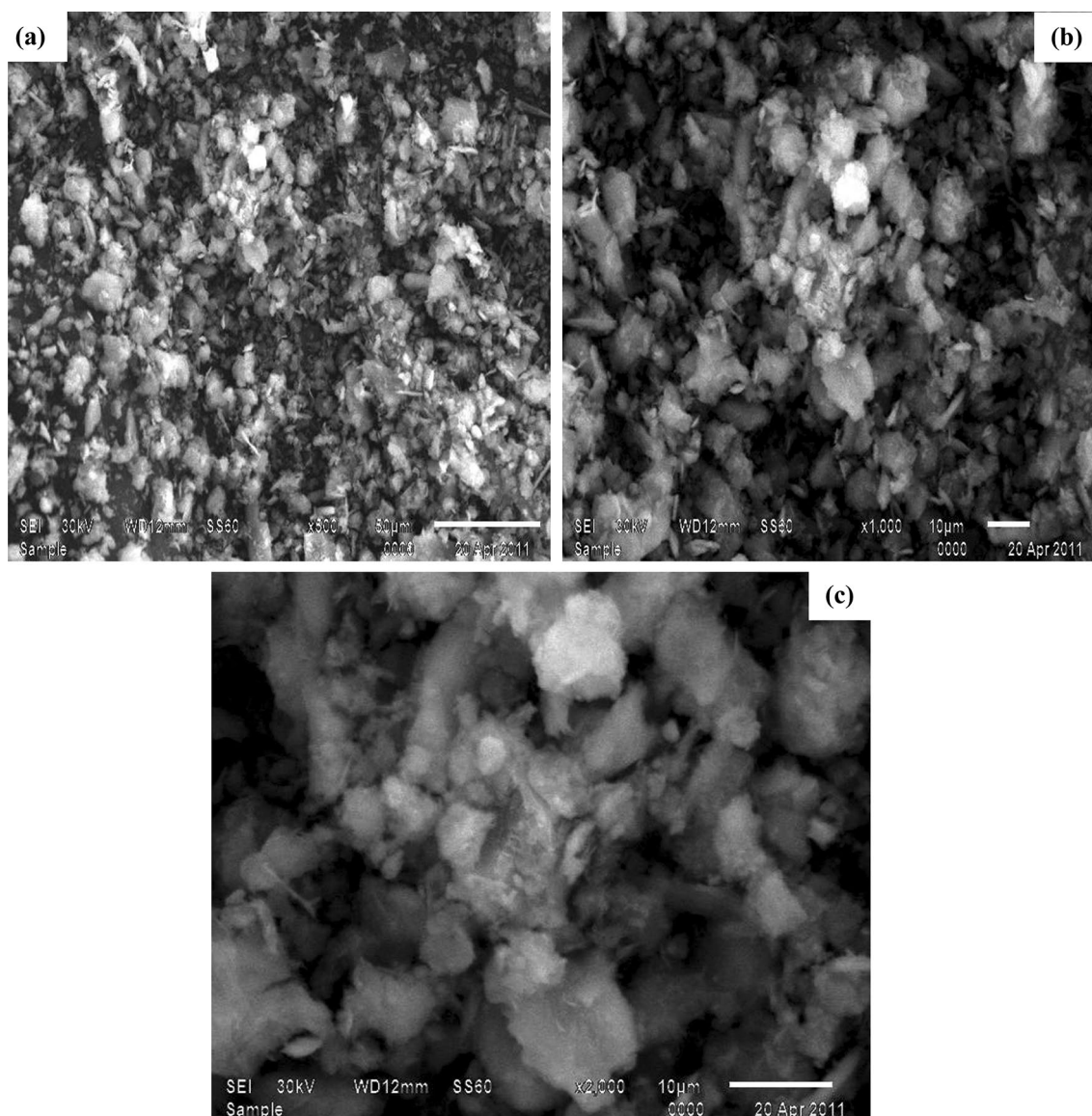


Fig. 2 SEM images for the proposed IPT sorbent material at different magnification powers **a** X500 **b** X1000 **c** X2000

media (grey color), simply they are similar to a bulk of tiny islands on the ocean surface. At small magnification power of X500, the surface appears to have very small porous, and by increasing the magnification power at X1000 and X2000, these particles are rugged, and sharp and have intermolecular distances that support the physical sorption process on the material.

XRF analysis

The percentage of metal present in IPT sorbent based on an X-ray fluorescence spectrometer was measured and revealed that the prepared IPT sorbent possesses the values

14.2, 4.31, 24.26, 15.11, 3.92, 3.38, and 34.82 for Mg, Al, Si, P, Ca, K, and Fe respectively. These data confirmed that the main constituents are Fe, Si, P, and Mg. Also, the reason for the appearance of Ca, K, and Al elements in XRF analysis is due to the presence of these elements in natural talc.

pH titration curve

Figure 3 displays the pH titration curve of IPT sorbent. The X-axis and Y-axis characterize the number of mL of NaOH solution interacting with 0.1 g of IPT sorbent, and the pH of the effluent, respectively. The pH titration curve for IPT

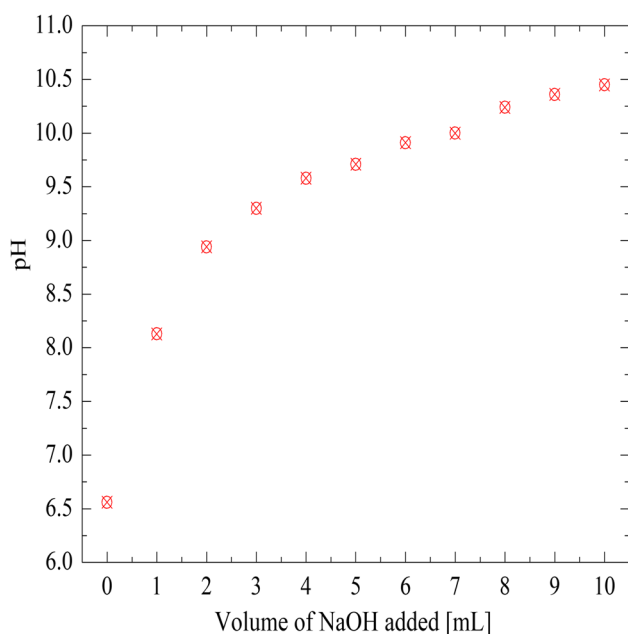


Fig. 3 pH titration curve of IPT sorbent

sorbent reflect only one inflection point at a pH value equal 10, revealing that it is a mono-functional sorbent. This is the same behavior as ZrSnP prepared by Abass et al. [24].

Chemical stability

The solubility test of the IPT sorbent is represented in Table 2 reflecting that the prepared sorbent was stable in DDW, ethanol, methyl ethyl ketone, HNO₃, and NaOH below 2 mol L⁻¹. Table 2 exhibits that IPT sorbent has relatively high stability to chemical reagents compared to other sorbents [42–44].

Table 2 Chemical stability of IPT sorbent in different solvents

Solvents	% Solubility
DDW	Below detection limit
Ethanol, [96%]	0.5
Methyl ethyl ketone	2.25
0.5 mol/L HNO ₃	2.12
1 mol/L HNO ₃	2.93
3 mol/L HNO ₃	5.56
0.5 mol/L NaOH	4.23
2 mol/L NaOH	24.62

Sorption studies

Influence of pH on the sorption efficiency and distribution coefficients (K_d)

The sorption of ¹³⁷Cs, ¹⁵²⁺¹⁵⁴Eu, and ¹³¹Ba onto IPT sorbent in different pH values was studied from aqueous media as exposed in Fig. 4a. According to this Figure, the % S.E. increases with increasing pH until it reaches the maximum levels of 96.0, 83.2, and 81.0% for ¹³¹Ba, ¹³⁷Cs, and ¹⁵²Eu, respectively, at pH 6 for ¹³¹Ba and ¹³⁷Cs and pH 4 for ¹⁵²⁺¹⁵⁴Eu. The sorption of ¹³¹Ba, ¹³⁷Cs, and ¹⁵²Eu was found to be low at low pH values, which is likely caused by the protonation of the surface-active sites and the rise in H₃O⁺ ions in the aqueous solution [45]. As a result of competition for the accessible binding surface active site produced by the positively charged surface sites, uptakes of the radionuclides ¹³¹Ba, ¹³⁷Cs, and ¹⁵²Eu were reduced. With rising pH values, the concentration of H₃O⁺ ions is reduced while the concentration of OH⁻ is raised which causes deprotonation of the sorbent surface, such explanations mean that the surface of the IPT sorbent tends to have a negative charge. Hence, the attraction among the surface of IPT and the positive charge of ¹³¹Ba, ¹³⁷Cs, and ¹⁵²Eu in the solution was improved.

The speciation of Ba(II), Cs(I), and Eu(III) ions in a liquid solution at different pH values (1–7), at ionic strength 0.001 M and 25 °C was performed using the MEDUSA program which gives information about the distribution of species as a function of pH [46], as exposed in Fig. 4b–d. The results showed that the speciation of Cs(I) and Ba(II) ions have no precipitate at all pHs (1–7) [7]. H⁺ ions concentration was decreased with increasing pH values and at pH approximately 7, H⁺ ions disappeared and at pH 5, OH⁻ ions start to increase with increasing pH values. The speciation of Eu(III) ions has no precipitate at a pH range from 1 to 2.2 with no presence of these ions in the hydroxide form. The hydrolysis of Eu(III) ions begins at a pH value above 2.2, and different species can be shaped, such as EuOH²⁺ and Eu(OH)₂⁺ formed at pH 5, the uptake of Eu(III) increases with pH until 4.0 and then decreased due to formation of EuOH²⁺ and Eu(OH)₂⁺. After pH 6 Eu(III) precipitated as Eu(OH)₃. The above data of chemical speciation are compatible with the sorption of ¹³⁷Cs, ¹⁵²⁺¹⁵⁴Eu, and ¹³¹Ba onto IPT sorbent at pH 6 for ¹³¹Ba and ¹³⁷Cs and pH 4 for ¹⁵²⁺¹⁵⁴Eu.

Distribution coefficients (K_d) and separation factors (α_B^A) at different pHs (1–7) were calculated and tabulated in Table 3 and reflected that K_d has the affinity order: ¹³¹Ba > ¹³⁷Cs > ¹⁵²⁺¹⁵⁴Eu, this result supports that ¹³¹Ba and ¹³⁷Cs uptake was carried out in the case of ionic radii [¹³¹Ba and ¹³⁷Cs have ionic radius 0.142 and 0.165 nm, respectively] whereas the sorption of ¹⁵²⁺¹⁵⁴Eu was done as hydrated ionic radius [¹⁵²⁺¹⁵⁴Eu has an ionic radius

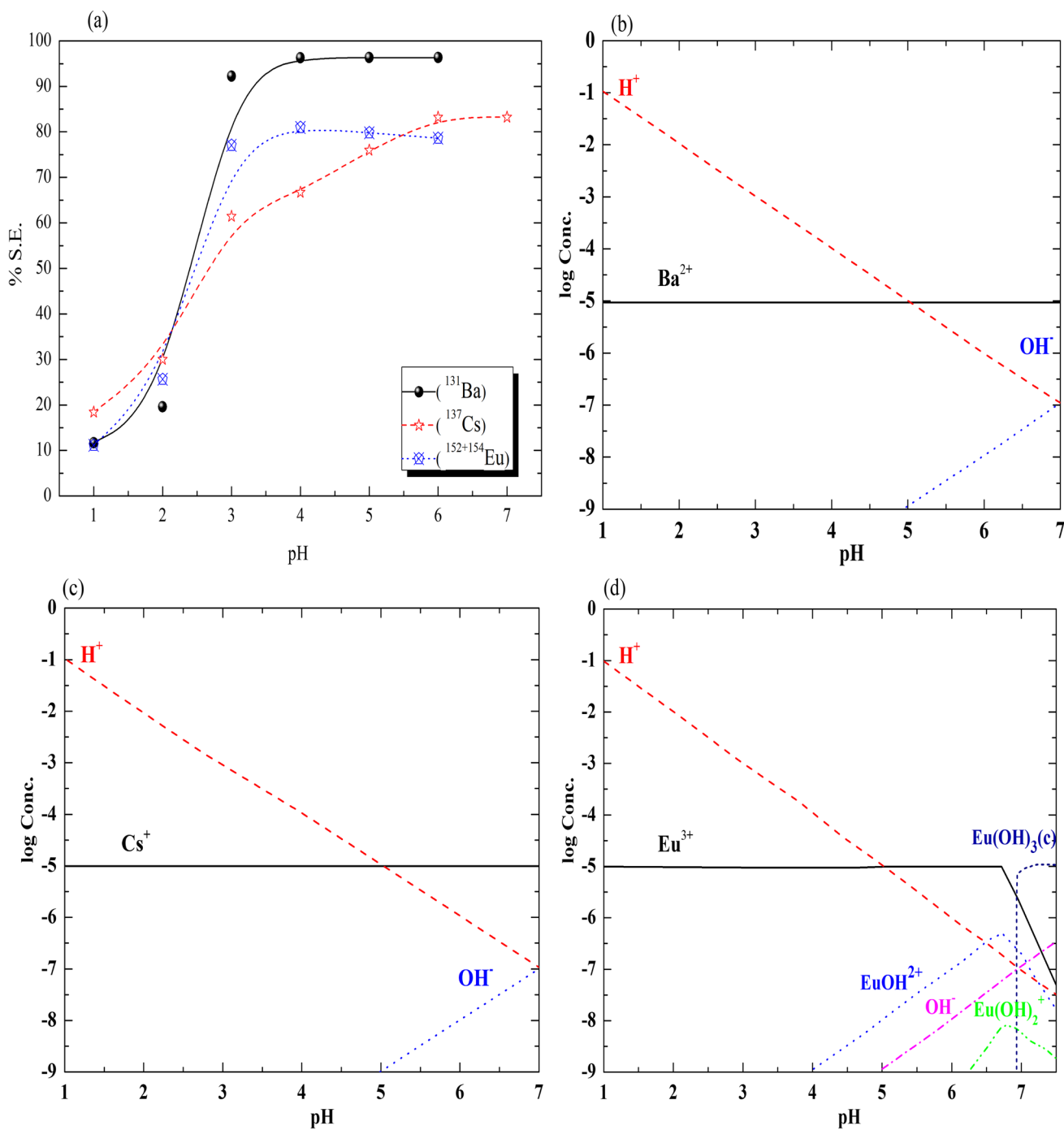


Fig. 4 a Effect of pH on the % S.E. of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent and chemical speciation of **b** Ba(II), **c** Cs(I), and **d** Eu(III) at pH range (1–7), ionic strength 0.001, and 25 °C

0.107 nm]. Radionuclides with lower ionic radius (^{131}Ba) easily enter the cavities of IPT sorbent increasing in sorption and consequently increasing K_d [47, 48]. Separation factors for studied radionuclides reflected that ^{131}Ba has a good separation factor at different pH values. Also, the data obtained in Table 3 reveal that the best separation factors

for studied radionuclides were at pH 4 (6.1 for the separation of ^{131}Ba from $^{152+154}\text{Eu}$ and 12.9 for the separation of ^{131}Ba from ^{137}Cs).

Nonlinear relationships between $\log K_d$ and pH were observed for the studied radionuclides, as shown in Fig. 5a. This relationship reveals the exchange reaction is

Table 3 Distribution coefficients and separation factors of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at 25 ± 1 °C

pH	K_d [mL g ⁻¹] and α_B^A	$^{152+154}\text{Eu}$	^{137}Cs	^{131}Ba
1	K_d	12.6	22.6	13.3
	α_B^A		1.8	1.1
2	K_d	34.5	42.9	24.4
	α_B^A		1.2	0.7
3	K_d	335.7	159.4	1190.8
	α_B^A		0.5	3.5
4	K_d	425.5	200.5	2581.6
	α_B^A		0.5	6.1
5	K_d	–	316.0	2636.0
	α_B^A		–	–
6	K_d	–	496.7	2637.0
	α_B^A		–	8.3
				5.3

non-ideal. The difference could be attributed to the prominence of a mechanism other than ion exchange, such as precipitation and/or surface adsorption [33].

Shaking time impact

The impact of contact time on ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorption onto IPT sorbent was studied at a fixed temperature (298 ± 1 K), initial concentration $C_i = 50$ mg L⁻¹, $V/m = 0.1$ L g⁻¹, shaking time (5 min–24 h), pH=4 for $^{152+154}\text{Eu}$, and pH=6 for ^{131}Ba and ^{137}Cs , and the data are shown in Fig. 5b. This Figure examined that the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent enhanced with contact time attain equilibrium at 1 h for ^{137}Cs and $^{152+154}\text{Eu}$ and 5 h for ^{131}Ba . The rate of ^{137}Cs and $^{152+154}\text{Eu}$ uptake onto IPT sorbent rapidly increases with time from 5 to 60 min reaching flattened behavior. Moreover, the rate ^{131}Ba uptake is different, it takes two stages to be flattened the first one start 5–90 min and then form rapidly rises from 150 to 300 min, and starts to flatten again till 1000 min achieving the maximum % S.E. Results can be concluded as there is no significant change of the uptake above 100 min for ^{137}Cs and $^{152+154}\text{Eu}$ but at 300 min for ^{131}Ba .

Kinetic studies

In the present work, pseudo-1st-order (Lagergren equation), and pseudo-2nd-order were applied to study and analyze the

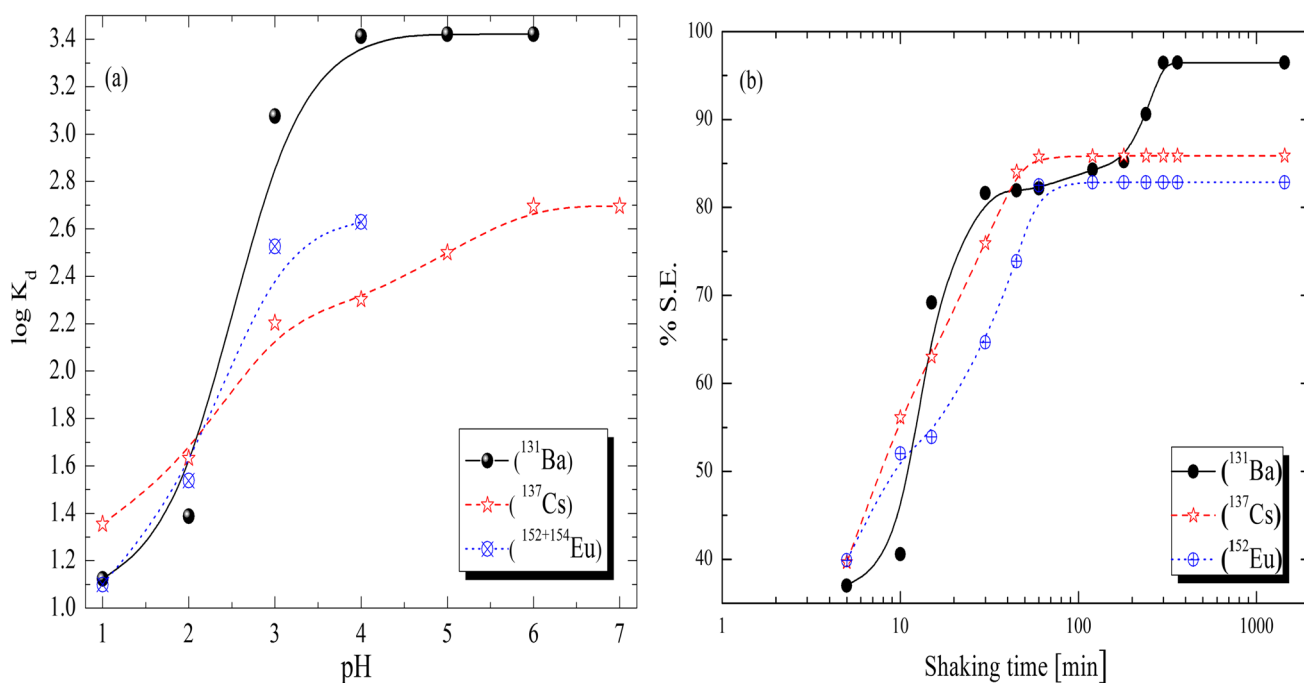


Fig. 5 Sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent, **a** plots of $\log K_d$ against pH at 25 ± 1 °C and **b** effect of shaking time on the % S.E.

obtained data from the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent, their equations are given below as [49, 50]:

$$\log (q_e - q_t) = \log q_e - \frac{K_f t}{2.303} \quad (5)$$

$$\frac{t}{q_t} = \frac{1}{K_s q_e^2} + \frac{t}{q_e} \quad (6)$$

in which, q_t is the value of the amount sorbed per unit mass (mg g^{-1}) at time t , K_f (min^{-1}), and K_s ($\text{g mg}^{-1} \text{min}^{-1}$) are the rate constants of two kinetic equations. By plotting $\log (q_e - q_t)$ versus t and t/q_t versus t , the obtained results are displayed in Fig. 6a and b. The result shows that the obtained experimental data are well-fitted with the pseudo-2nd-order model rather than the 1st-order model. The calculated constants of the two models are tabulated in Table 4. By comparing the model data with the obtained experimental data; it

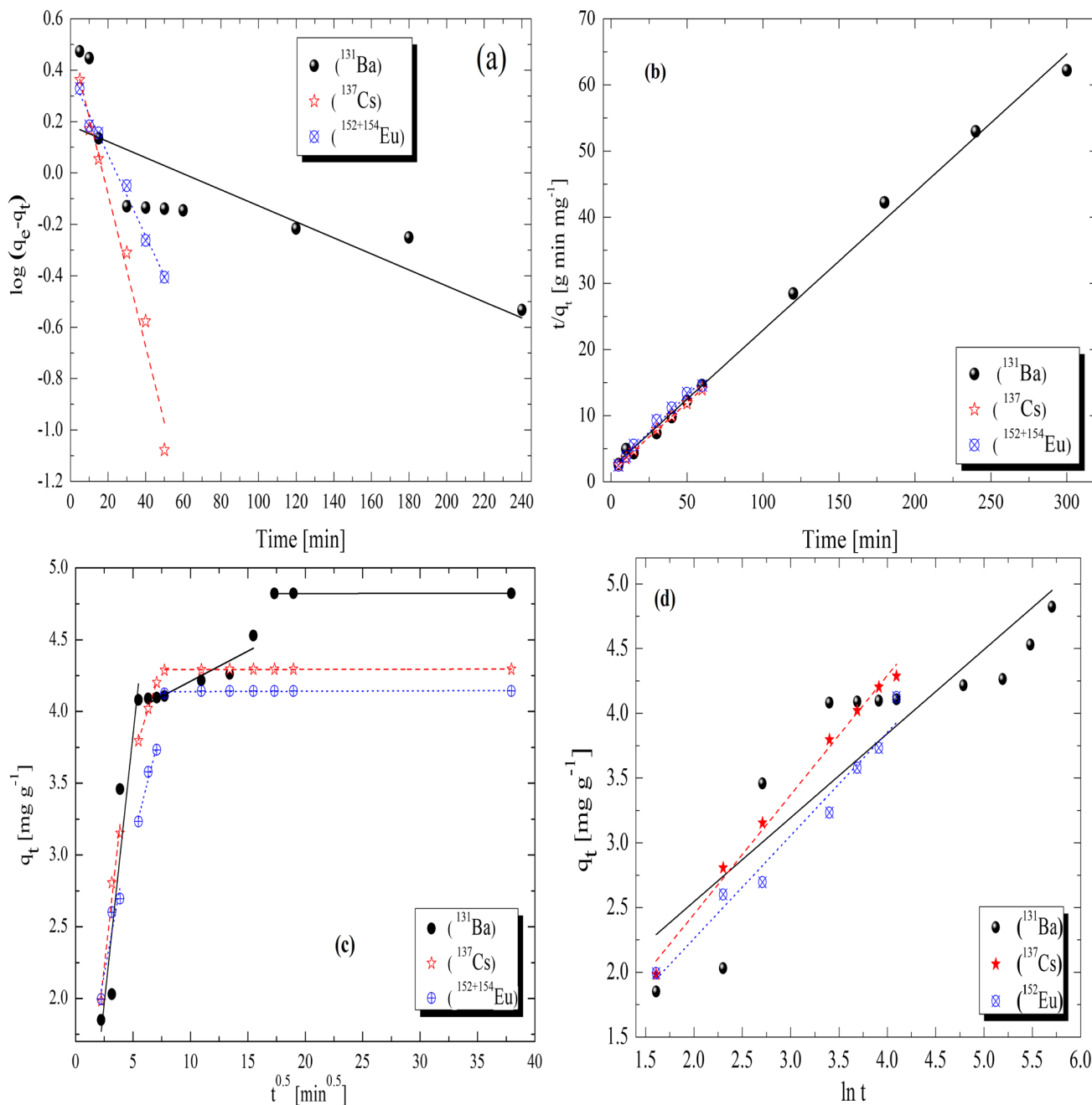


Fig. 6 Kinetic modeling fitting of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent, **a** pseudo-1st-order kinetic, **b** pseudo-2nd-order kinetic, **c** intra-particle diffusion, and **d** Elovich models at $25 \pm 1^\circ\text{C}$

Table 4 Kinetic parameters and correlation coefficients (R^2) for pseudo-1st-order, pseudo-2nd-order, intra-particle diffusion, and Elovich models for the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at 25 ± 1 °C

Radionuclides	q_e (exp) [mg g ⁻¹]	Pseudo-1st-order			Pseudo-2nd-order			
		q_e (cal) [mg g ⁻¹]	K_f	R^2	q_e (cal) [mg g ⁻¹]	h	K_s	R^2
^{131}Ba	4.82	1.53	-0.003	0.580	4.79	0.49	0.089	0.995
^{137}Cs	4.29	3.25	-0.030	0.976	4.81	1.55	0.028	1.000
$^{152+154}\text{Eu}$	4.14	2.43	-0.016	0.987	4.47	1.90	0.026	0.985
Intra-particle diffusion					Elovich			
Radionuclides	Time range [min]	K_{id}	C	R^2	α	β	R^2	
^{131}Ba	5–30	0.747	0.101	0.822	4.43	1.541	0.777	
	40–240	0.04209	3.69273	0.7626				
	300–1440	–	4.824	–				
^{137}Cs	5–15	0.72235	0.41523	0.9516	1.77	1.084	0.965	
	30–50	0.25484	2.40406	0.9987				
	60–1440	–	4.29077	–				
$^{152+154}\text{Eu}$	5–15	0.43951	1.0719	0.7983	1.84	1.255	0.967	
	30–50	0.31534	1.53129	0.9345				
	60–1440	–	4.13563	–				

proposes that pseudo-2nd-order sorption is the main mechanism, as data quality is related to R^2 (a goodness-of-fit measure for linear regression models); for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba are almost equal to one compared with its low values obtained from the pseudo-1st-order kinetic model. Also, the values of q_e calculated from the pseudo-2nd-order model were nearer to the values of q_e (experimental). This means the adsorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba was found to be a chemisorption process [51, 52].

The diffusion mechanism of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent was investigated by the intra-particle diffusion model which is presented in the next equation [53, 54].

$$q_t = K_{id}t^{0.5} + C \quad (7)$$

Which, K_{id} and C are the intra-particle diffusion rate constant (mg g⁻¹ min^{-0.5}) and the intra-particle diffusion constant which is directly proportional to the boundary layer thickness, respectively. The plot of q_t versus $t^{0.5}$ is assumed in Fig. 6c. The value of the rate parameter was calculated from the slope, and the value of R^2 is assumed in Table 4. Three steps were used to create the intra-particle diffusion model for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorption [55, 56]. (a) The diffusion of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba from the solution to the surface of the IPT sorbent (from 5 to 30 min) for ^{131}Ba and (from 5 to 15 min) for ^{137}Cs and $^{152+154}\text{Eu}$. (b) The gradual sorption on the surface of the IPT sorbent, which may be the rate-limiting step (from 40 to 240 min) for ^{131}Ba , (from 30 to 50 min) for ^{137}Cs and $^{152+154}\text{Eu}$. (c) The equilibrium

saturation (from 300 to 1440 min) for ^{131}Ba and (from 60 to 1440 min) for ^{137}Cs and $^{152+154}\text{Eu}$. The sorption mechanism of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent was found to be fast at the initial period of contact time and then to become constant with the increase in time. The sorption mechanism, which comprises both film and intra-particle diffusion, is primarily controlled by the multi-diffusion step.

The Elovich kinetic model is used to describe the kinetics of chemisorption on highly energetically heterogeneous solid surfaces. The mathematical linear form of this model is expressed as [57]:

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \quad (8)$$

where: α is the Elovich coefficient which represents the initial sorption rate (mg g⁻¹ min⁻¹) and β is a parameter related to the desorption constant (g mg⁻¹) during any experiment. The plots of q_t vs. $\ln t$ for the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent are shown in Fig. 6d. It gives a linear relationship with a slope of $1/\beta$ and an intercept of $1/\beta \ln(\alpha\beta)$. The values of Elovich prediction model parameters were determined from the obtained linear form and listed in Table 4. The data in Table 4 declared that the Elovich equation did not fit the experimental results based on the low values of R^2 . This validated the inapplicability of this model for describing the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent.

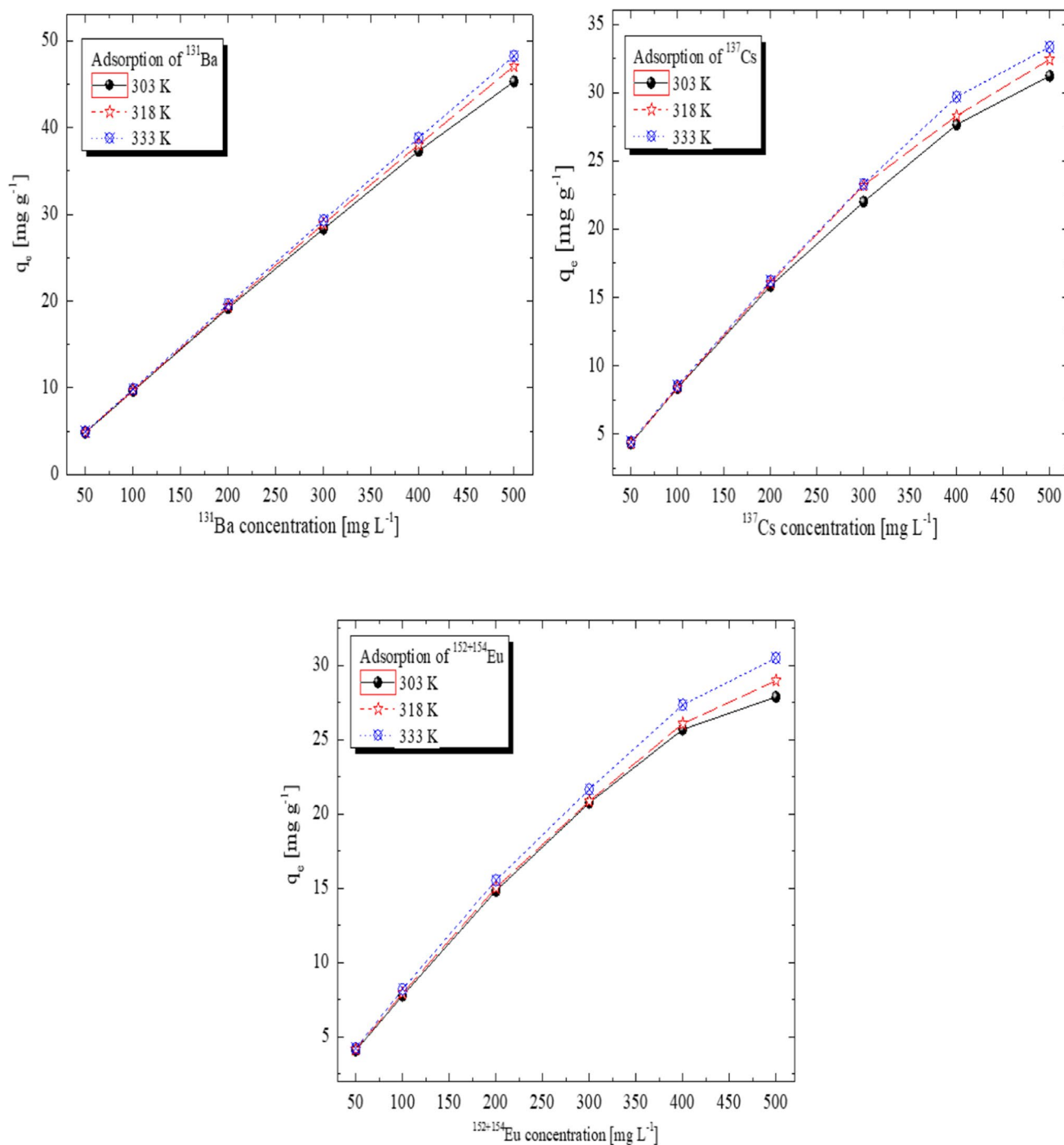


Fig. 7 Effect of initial concentration of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at different temperatures [$V/m=0.1 \text{ L g}^{-1}$, $\text{pH}=6$ for ^{131}Ba and ^{137}Cs and $\text{pH}=4$ for $^{152+154}\text{Eu}$]

Effect of concentrations

Figure 7 reveals the plots between q_e of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT and C_i at the range (50–500 mg L^{-1}) at different reaction temperatures. The q_e of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent rises as the initial concentration

of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba increases. As well as, the q_e increases by increasing reaction temperature indicating the endothermic nature of the sorption.

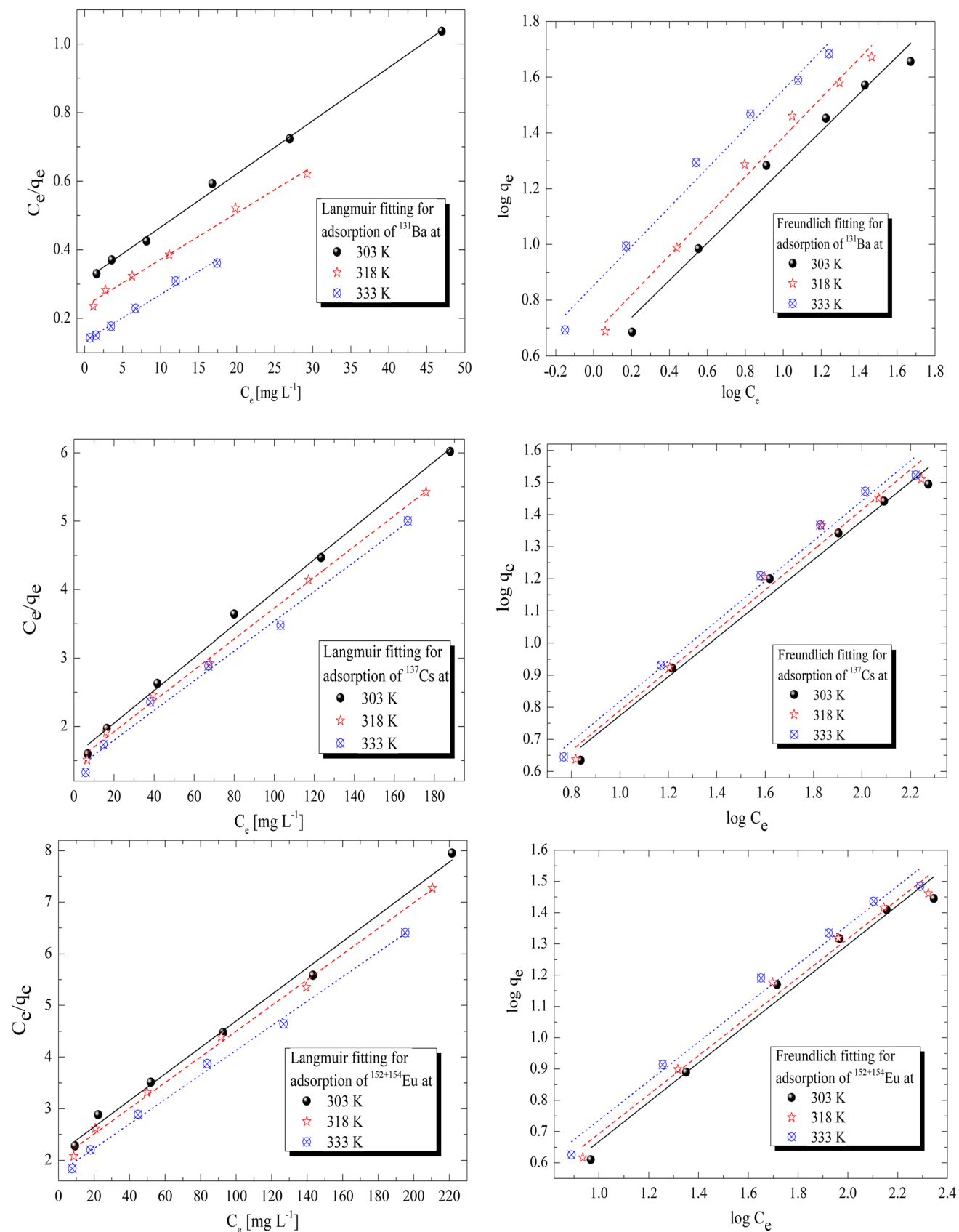


Fig. 8 Isothermal modeling fitting of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at different reaction temperatures [$V/m=0.1 \text{ L g}^{-1}$, $\text{pH}=6$ for ^{131}Ba and ^{137}Cs and $\text{pH}=4$ for $^{152+154}\text{Eu}$]

Isothermal modeling

The Langmuir isotherm model can be represented in the equation [3, 58]:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_{\max}} + \frac{1}{bQ_{\max}} \quad (9)$$

Q_{\max} and b are the theoretical monolayer capacity (mg g^{-1}) and the sorption equilibrium constant is related to the energy of sorption, respectively, and C_e is the equilibrium concentration. As shown in Fig. 8, plotting C_e/q_e and C_e gives straight lines for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at temperatures (303, 318, and 333 K). The calculated data from the slopes and intercepts of the linear form of the Langmuir model for ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorption onto IPT sorbent at 303, 318, and 333 K were presented in Table 5. The R^2 values confirm that the high applicability of Langmuir isotherm for the sorption of these radionuclides onto IPT sorbent. The Q_{\max} was (64.47 , 73.64 , and 73.75 mg g^{-1}), (41.1 , 44.37 , and 46.06 mg g^{-1}), and (39.0 , 40.2 , and 42.11 mg g^{-1}) for ^{131}Ba , ^{137}Cs , and $^{152+154}\text{Eu}$ at 303, 318, and 333 K, respectively. These data prove the endothermic nature of the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent because of rising

Q_{\max} for ^{131}Ba , ^{137}Cs , and $^{152+154}\text{Eu}$ with increasing reaction temperature.

The equilibrium parameter (R_L) can be obtained from the Langmuir constant b as follows:

$$R_L = \frac{1}{1 + bC_i} \quad (10)$$

The R_L values reflect the type of isotherm to be irreversible if, $R_L = 0$, favorable $0 < R_L < 1$, linear if $R_L = 1$, or unfavorable $R_L > 1$. Table 5 proves that the $0 < R_L < 1$ reveals the favorable sorption isotherms of studied radionuclides [3].

A linear form of Freundlich expression can be represented as:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (11)$$

In which, K_F and $1/n$ are the Freundlich constant and heterogeneity factors. It represents the amount of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT for unit equilibrium concentration. The values of $1/n$ signify the deviation from the linearity of sorption as follows: (i) $1/n = 1$ reflects that the sorption is linear; (ii) $1/n < 1$ implies heterogeneous surface structure with minimum interaction between the adsorbed atoms, and (iii) $1/n > 1$ implies homogeneous surface

Table 5 Isotherm parameters for sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent at different reaction temperatures

Radionuclides	Temp [K]	Langmuir constants				Freundlich constants		
		Q_{\max} [mg g^{-1}]	b [L mg^{-1}]	R_L	R^2	$1/n$	K_F [mg g^{-1}]	R^2
^{131}Ba	303	64.47	0.050	0.04	0.998	0.67	4.02	0.976
	318	73.64	0.058	0.03	0.991	0.71	4.77	0.988
	333	73.75	0.101	0.02	0.991	0.70	7.12	0.982
^{137}Cs	303	41.81	0.015	0.116	0.995	0.61	1.47	0.983
	318	44.37	0.015	0.116	0.996	0.63	1.46	0.979
	333	46.06	0.016	0.112	0.989	0.62	1.56	0.986
$^{152+154}\text{Eu}$	303	39.00	0.012	0.143	0.993	0.63	1.09	0.975
	318	40.21	0.012	0.139	0.996	0.62	1.18	0.982
	333	42.11	0.014	0.129	0.996	0.62	1.30	0.981

Table 6 Comparison of the monolayer capacity of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorbed onto various sorbents

Sorbents	Shaking time [min]	Q_{\max} [mg g^{-1}]			Ref. no
		^{131}Ba	^{137}Cs	$^{152+154}\text{Eu}$	
IPT	240	64.47	41.81	39.0	Current work
Dolomite powder	120	3.96	NR	NR	[59]
MXene	120	9.3	NR	NR	[60]
SP(M) (miswak powder)	60	34.97	NR	NR	[15]
SnSiMo	15	NR	16.14	NR	[65]
SnV	120	NR	26.68	NR	[61]
PAM/TiWSi	120	NR	30.7	26.6	[62]
Activated carbon	90	NR	NR	18.41	[63]
B:D composite	1440	NR	NR	26.87	[64]

structure and unfavorable Freundlich adsorption processes [52]. The results of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba sorbed onto IPT sorbent by Langmuir and Freundlich isotherms were shown in Fig. 8. As well as the constants of both isotherm models were calculated from the slope and intercept, and the data were tabulated in Table 5. The R^2 values obtained from Freundlich for ^{131}Ba , ^{137}Cs , and $^{152+154}\text{Eu}$ were lower than the Langmuir isotherm values. Based on R^2 data, the linear forms of the Freundlich model were less applicable than the Langmuir model.

Literature review of monolayer sorption capacity of different sorbents

The monolayer capacity (Q_{max}) of IPT sorbent for the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba was compared with other sorbents reported in the literature. As characterized in Table 6, the monolayer capacity of IPT is a good value compared with the previously reported values which recommended that this is a promising material to decontaminate ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba from aqueous media [15, 35, 59–66].

Effect of temperature studies

The linear form between $\ln K_d$ of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent and $1000/T$ was exposed in Fig. 9 according to Van't Hoff relation [24]:

$$\ln K_d = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT}$$

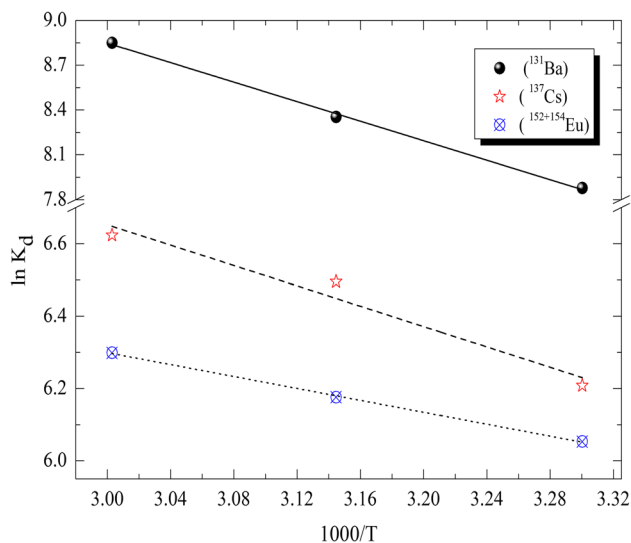


Fig. 9 A plot of $\ln K_d$ against $1000/T$ for sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent [$C_i=50 \text{ mg L}^{-1}$, $V/m=0.1 \text{ L g}^{-1}$, $\text{pH}=6$ for ^{131}Ba and ^{137}Cs and $\text{pH}=4$ for $^{152+154}\text{Eu}$]

Table 7 Thermodynamic parameters for the sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent

Radionuclides	Temp [K]	ΔH° [kJ mol ⁻¹]	ΔS° [J mol ⁻¹ K ⁻¹]	ΔG° [kJ mol ⁻¹]
^{131}Ba	303	27.2	155.0	-19.8
	318			-22.1
	333			-24.5
^{137}Cs	303	11.7	90.4	-15.7
	318			-17.0
	333			-18.4
$^{152+154}\text{Eu}$	303	6.9	72.9	-15.2
	318			-16.3
	333			-17.4

where R , ΔH° , T , and ΔS° are the gas constant, the enthalpy change of adsorption, the absolute temperature, and the entropy change of adsorption, respectively. As exposed in Fig. 9, $\ln K_d$ of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba increased with rising reaction temperature from 303 to 333 K. This enhancement is due to the acceleration of previously slow adsorption steps and the formation of new active sites on adsorbent surfaces [52]. From both slopes and intercepts of the straight lines, ΔH° and ΔS° were obtained and tabulated in Table 7. The positive values of ΔH° and ΔS° reflect the endothermic nature of the adsorption process and the increased randomness of the solid solution interface during the adsorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent, respectively [24, 52, 67]. The free energy change of specific adsorption (ΔG°) was obtained using the relation:

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (13)$$

The negative values of ΔG° reflect the adsorption process is spontaneous and reflects the preferable adsorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent compared with the H^+ ion [67].

Conclusion

The co-precipitation method was used to prepare IPT sorbent. IPT sorbent was characterized and employed for batch sorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba from an aqueous solution. The sorption data of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba reveal that IPT has an equilibrium time for ^{137}Cs and $^{152+154}\text{Eu}$ (1 h) and ^{131}Ba (5 h). The optimal $\text{pH}=4$ for $^{152+154}\text{Eu}$ and $\text{pH}=6$ for ^{131}Ba and ^{137}Cs . The pseudo-2nd-order kinetic described the kinetic data well, reflecting the presence of chemisorption. Isotherm models are more applicable for the Langmuir model with high monolayer

capacity for studied radionuclides. Thermodynamic functions show the adsorption of ^{137}Cs , $^{152+154}\text{Eu}$, and ^{131}Ba onto IPT sorbent was endothermic and spontaneous. Finally, IPT sorbent could be considered a promising sorbent possessing high sorptive abilities for the studied radionuclides.

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Declarations

Conflict of interest The authors whose names are listed immediately below certify that they have NO affiliations with or involvement in any organization or entity with any financial interest (such as honoraria; educational grants; participation in speakers' bureaus; membership, employment, consultancies, stock ownership, or other equity interest; and expert testimony or patent-licensing arrangements), or non-financial interest (such as personal or professional relationships, affiliations, knowledge or beliefs) in the subject matter or materials discussed in this manuscript.

Consent to publish Yes.

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