



Efficient removal of U(VI) from aqueous solution using the biocomposite based on sugar beet pulp and pomelo peel

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Abstract

A biocomposite sorbent composed of sugar beet pulp and pomelo peel was utilized for the biosorption of uranium (VI) from the aqueous solution. Parameters such as solution pH, biocomposite amount, contact time, temperature and initial concentration of U(VI) ions on the adsorption performance of biocomposite sorbent was studied. The equilibrium data fitted best to the Langmuir's isotherm model ($q_{e,max} = 79.36 \text{ mg g}^{-1}$). Obtained thermodynamic and kinetic parameters demonstrated that the biosorption process is spontaneous, exothermic, and fitted best to the pseudo-second order. The desorption study revealed that uranium recovery by 0.1 M NaHCO_3 was 99.24% in the first cycle for used biocomposite.

Keywords Uranium (VI) · Biocomposite · Equilibrium · Thermodynamics · Desorption

Introduction

Huge amounts of liquid radioactive waste are produced on a global level every day, and since the fissile isotope of uranium is the most used fuel in nuclear power plants, it appears as one of the main constituents of this type of waste. The presence of this radionuclide has a negative effect on all species and ecosystems due to its chemical and radiotoxicity, long half-life between 10^5 and 10^9 years and through the food chain, its toxicity to humans is unavoidable [1]. In natural waters, uranium is present in low concentrations, while in waters near mines and industrial plants, the concentration of uranium can reach significantly higher levels. The permissible discharge level of U(VI) in the wastewater of nuclear industries is in the range from 0.1 to 0.5 mg L^{-1} [2]. For this reason, uranium must be removed from such wastewater before discharging it into natural recipients.

Conventional methods for uranium removal from aqueous solutions include solvent extraction, chemical precipitation, reverse osmosis, ion exchange, ultrafiltration, and adsorption

[3]. All the mentioned methods have proven to be effective in the treatment of uranium-contaminated wastewater. However, the limitations of these methods, such as high energy consumption, the creation of secondary pollution and incomplete removal, are unfavourable for the widespread popularization and promotion of these methods. Among these methods, adsorption, and within it biosorption, has an advantage over others due to its high efficiency and environmental friendliness [4, 5]. Biosorption is the process of removing pollutants of interest from aqueous solution using biological material and it enables the treatment of large amounts of effluents with a low concentration of sorbates of interest, low capital and operating cost, reduced amount of chemical or biological sludge, selective removal of metal ions, reduced use of toxic chemicals, rapid kinetics of adsorption and desorption, metal recovery and possibility of the biosorbent regeneration.

Previous studies have shown that biosorbents like algae, bacteria, fungi [6–8], and in recent researches different kinds of nanomaterials [9–11] and mesoporous materials [12–14] have good adsorption abilities for U(VI) removal from aqueous solution. However, in comparison to these, there are only a few studies regarding the biosorption of uranium by agricultural waste [15–18], and among these, all studies have concentrated on the uptake of uranium by agricultural biomass as individual and separate entities in native or modified forms. Biowaste from agricultural industries represents a low-cost adsorbent that possesses a lot of compounds such

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as pectin (galacturonic acid), hemicellulose, cellulose and lignin which have various polar functional groups, like carboxylic and phenolic acid groups that can be involved in metal ions complexation [19]. Additionally, the lignocellulosic material has shown very good chemical stability and mechanical strength due to its crystal structure. The biocomposite sorbents represent a composite material comprising of two or more materials that have a biological origin. Removal of uranium and similar radionuclides by adsorption onto biocomposite sorbents is insufficiently explored [20, 21] and there is a great potential in composite sorbents because biocomposites may display better characteristics of the effective biosorbents than its components individually.

This study regards removal of U(VI) from aqueous solution by biocomposite sorbent prepared as a combination of native forms of sugar beet pulp and pomelo peel and as a supporting material has been used silica gel. Key adsorption parameters were optimized and the nature of binding mechanism of U(VI)-biocomposite system was evaluated.

Materials and methods

Chemicals

All used chemicals were of analytical grade. Uranyl nitrate hexahydrate, Arsenazo III, 60% perchloric acid and silica gel (70–230 mesh) were purchased from Merck (Darmstadt), 85% phosphoric acid from Kemika and potassium hydroxide from Alkaloid (Skopje). The stock solution of U(VI) (1000 mg L^{-1}) was prepared by weighing 2.1308 g (Metler Toledo balance, $\pm 0.1 \text{ mg}$) of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ salt analytical reagent grade. Working solutions were prepared by diluting the stock solution.

Batch experiments

Batch experiments were performed in a closed polypropylene bottle at 95–100 movements per min (mechanical shaker: Heidolph Duomax 2030) to study the biosorption of U(VI) onto biocomposite sorbent. The experiments were performed by adding prepared biocomposite sorbent (50–500 mg) into a 50 mL solution containing a certain concentration of U(VI) (50–120 mg U/L). The filtrates were collected at suitable time intervals (0–120 min) at different temperatures (25, 35, 45 and 55°C). The pH of each sample solution (3–9) was adjusted with 3.0 mol/L NaOH and measured by digital pH meter (CG 841 Schott-GERÄTE GmbH). All biosorption experiments were duplicated to make the results reliable and repeatable, and the mean values have been used for data evaluation because the variation of the experimental data was within the measurement error ($\pm 5\%$). The concentration of U(VI) ions in the filtrate, before and

after sorption was determined spectrophotometrically at 650 nm by Arsenazo-III method on a UV–Vis spectrophotometer (model: Varian Cary 50) [22].

The removal efficiency (R , %) and sorption capacity of U(VI) ions (Q , mg L^{-1}) were calculated according to Eqs. (1) and (2).

$$R(\%) = \frac{C_i - C_f}{C_i} \cdot 100\% \quad (1)$$

$$Q = \frac{C_i - C_f}{m} \cdot V \quad (2)$$

where C_i and C_f are initial and final concentrations of U(VI) ions at the filtrate (mg L^{-1}), respectively, V is the volume of solution (L), and m is the mass of the biocomposite sorbent (g).

Biocomposite preparation

Sugar beet pulp was provided as a waste (fraction which remains after the production of sugar) from the Sunoko Sugar Factory (Novi Sad, Serbia). It was first washed by the tap water, then several times with smaller quantities of distilled water, air-dried for 24 h, and then dried in an oven at $80^\circ\text{C} \pm 5^\circ\text{C}$ for 12 h. After cooling in a desiccator, sugar beet pulp was ground in a blender and powder was sieved through a standard sieve ($\varnothing = 0.25 \text{ mm}$) and stored in a hermetically sealed container.

Pomelo was purchased at a local supermarket. Exocarp from the pomelo was removed from the fleshy mesocarp and cut into pieces of 0.5–1.0 cm, washed first by tap water, then several times with smaller quantities of distilled water, air-dried for 24 h, and dried in an oven at $80^\circ\text{C} \pm 5^\circ\text{C}$ for 12 h. After cooling in the desiccator, the pomelo peel was ground in a blender, and the powder was sieved through a standard steel sieve ($\varnothing = 0.25 \text{ mm}$). In this form, the powder of the native pomelo peel was stored in a hermetically sealed container.

For the preparation of the biocomposite, the procedure previously described by Akar et al. (2009) was used with some modifications [23]. 100 mL of 7% (w/v) aqueous solution of KOH was mixed with 10 g of silica gel and heated at $70\text{--}80^\circ\text{C}$ for 30 min. After cooling to 20°C , the solution was mixed with a suspension containing 5 g of sugar beet pulp and pomelo peel. After a uniform mass was reached, 20% H_3PO_4 was added to the solution in a very small amount until a gel formulation was reached. The resulting gel was dried in an oven at 50°C for 24 hours. After drying, the biocomposite was ground with mortar and pestle, sieved through a standard steel sieve ($\varnothing = 0.25 \text{ mm}$) and stored in a hermetically sealed container for further sorption experiment.

Biocomposite characterization

To determine functional groups of prepared biocomposite from sugar beet pulp and pomelo peel, infrared spectra of the Fourier transform (FT-IR) were recorded on a Perkin Elmer BX FT-IR spectrometer using KBr pellet technique in a region from 4000 to 400 cm^{-1} . For the determination of the particle size distribution, the Malvern Mastersizer 2000 laser diffraction system with the wet dispersion unit Hydro 2000S (A) was used. This device is designed to determine the particle size distribution in the range of 0.2–2000 μm .

For the Energy Dispersive X-Ray Fluorescence (EDXRF) analysis, previously prepared biocomposite was additionally ground using mortar and pestle, sieved ($\varnothing = 45 \mu\text{m}$) and pressed into triplicate pellets weighing about 2 g with a diameter of 2.5 cm. Pellets were analysed using the EDXRF technique. The excitation source was a Siemens X-ray tube with Mo anode and Mo secondary target in orthogonal geometry. The tube operated at 45 kV and 35 mA and the irradiation time was 1000 s. Spectra were collected by a Canberra Si(Li) detector (3 mm thickness, 30 mm^2 active area, 0.025 mm Be window thickness) with a resolution of 170 eV (FWHM) at 5.9 keV. Samples were measured in vacuum. IAEA QXAS software was used for the analysis of spectra. Concentrations of K, Ca, Mn, Fe, Ni, Cu, Zn and Pb were determined by direct comparison of count rates using the IAEA-392 (Trace, minor and major elements in algae) standard reference material.

The value of pH_{pzc} (zero point of charge) is determined from the curve that intercepts the pH_i line of the plot ΔpH versus pH_i , where ΔpH represents a difference of $\text{pH}_{\text{final}} - \text{pH}_{\text{initial}}$ and pH_i is initial pH value of the buffered solution according to the previously described procedure by Zou and Zhao (2012) [24].

Desorption experiments

Desorption experiments were performed under optimal sorption parameters using the following desorption solutions: 1.0 M HNO_3 , NaHCO_3 and citric acid to recover the adsorbed uranium from biocomposite. The percentage desorption from the spent biocomposite was calculated from the dose of U(VI) adsorbed onto biocomposite and the final U(VI) concentration in the desorption solution. To investigate the reusability of the biocomposite, consecutive adsorption-desorption cycles were repeated three times with the same biocomposite using previously determined desorption solution at different concentrations (0.25; 0.5; 1.0 M).

Results and discussion

Biocomposite characterization

FTIR analysis

The obtained FTIR spectrum of the prepared biocomposite is presented in Fig. 1. The wide peak at 3427 cm^{-1} is assigned to the stretching of O-H vibrations due to the stretching of the alcohols, phenols and carboxylic acids in pectin, cellulose and lignin which are the main constituents of the pomelo peel and sugar beet pulp. This peak may also indicate the presence of N-H stretching vibrations [25]. A peak at 2928 cm^{-1} corresponds to the stretching of C-H bonds in saturated aliphatic constituents [26]. The absorption band at 2378 cm^{-1} indicates the presence of a triple C \equiv C bond [21]. A peak at 1736 cm^{-1} is assigned to the stretching of the C=O bond in the ester groups [27]. The band at 1650 cm^{-1} can be attributed to the symmetrical stretching of the C=O bonds of carboxylates ($\nu_s(\text{O}_2\text{C})$) and polyphenols ($\nu_s(\text{O}=\text{C})$). Additionally,

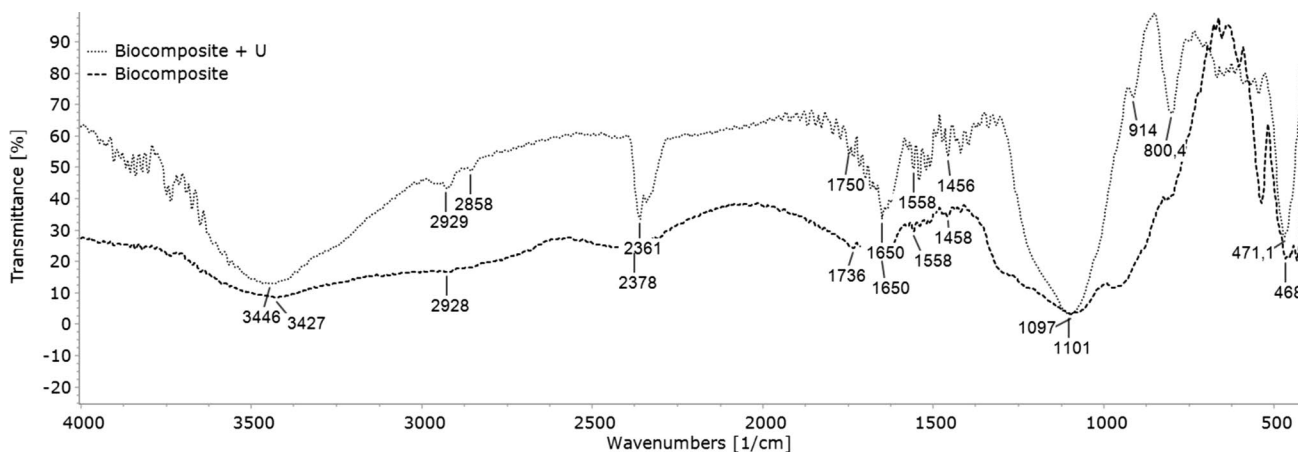


Fig. 1 FTIR spectra after and before sorption

the peak at 1650 cm^{-1} indicates a successful immobilization of the used components according to the study of Akar et al. [23] in which immobilization of *P. Vulgaris* was performed on activated silica gel. Symmetrical stretching of the C=C bonds of the aromatic systems of lignin and polyphenols can be attributed to a band of moderate intensity at 1558 cm^{-1} . A peak at 1458 cm^{-1} corresponds to the deformations of the C-H bonds of the various saturated cyclic rings, together with the deformations of the terminal methylene bonds (from $-\text{CH}_2\text{OH}$). This band predominantly originates from a symmetrical degenerate deformation of the methyl group of acetate and methoxy(poly)substituted aromatics, respectively. Since the immobilization of sugar beet pulp and pomelo peel was performed on activated silica gel, the band at 1097 cm^{-1} according to the literature indicates the presence of Si-O-Si bonds that are characteristic for silica gel. This peak was also found in the work of Akar et al. [23] and was taken as one of the indicators of successful immobilization.

The FTIR spectra of the loaded biocomposite showed which functional groups are involved in the sorption of uranyl ions (Fig. 1). The peak at 3427 cm^{-1} was shifted to 3446 cm^{-1} and the peak at 1736 cm^{-1} was shifted to 1750 cm^{-1} . Additionally, the change in band intensities in the region of 1600 to 1400 cm^{-1} could be observed. Furthermore, a new peak at 914 cm^{-1} is assigned to UO_2^{2+} stretching group which showed that sorption of uranyl ion was successful.

Zero point of charge (pH_{pzc}) of biocomposite

The pH_{pzc} of a biosorbent is a very important characteristic that determines the pH at which the biosorbent surface has net electrical neutrality. It is a characteristic of amphoteric surfaces and it is determined by the type of functional groups. Namely, the surface of the sorbent is positively charged in the case when the $\text{pH} < \text{pH}_{\text{pzc}}$ and shows an affinity for anions, while in the case when the surface is negatively charged, i.e. when the $\text{pH} > \text{pH}_{\text{pzc}}$, it shows an affinity for cations. The zero point of charge of the used biocomposite was 2.49 (Fig. 2), which is in accordance with the determined pH value (4). The obtained value indicates that below the pH value of the solution (2.49), the surface of the used biocomposite will be positively charged and will have a higher affinity for anions, while above this value surface of the biocomposite will be negatively charged and will have an affinity for positively charged cations, such as uranyl ions. Additionally, the obtained pH value is in accordance with the charge of the present functional groups on the surface of the used biocomposite.

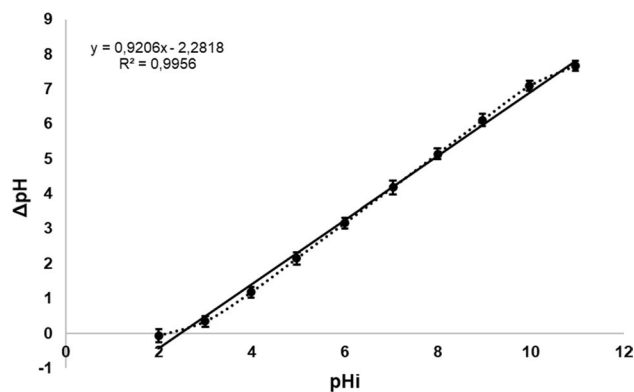


Fig. 2 The zero point of charge (pH_{pzc}) of biocomposite

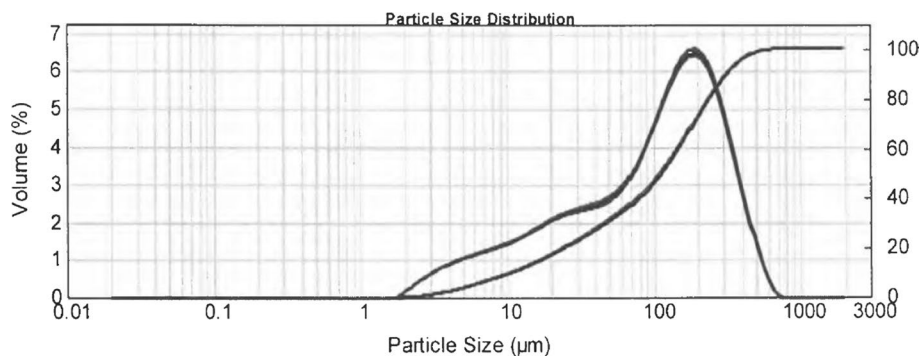
Particle size distribution analysis (PSA)

According to the obtained results for the particle size distribution of the prepared biocomposite (Fig. 3) the highest percentage of the particles of large ($\approx 90\%$) and medium diameter, and small particles are present in a small percentage. 10% of the particles are $< 10.69\text{ }\mu\text{m}$, 50% of the particles are $< 113.96\text{ }\mu\text{m}$ and 90% of the particle are $< 317.29\text{ }\mu\text{m}$. Regarding the obtained results for the adsorption capacity of U(VI) onto used biocomposite, this particle size distribution resulted in a favourable biosorption process for U(VI).

EDXRF analysis

In modern times, the popularity of eco-friendly and low-cost biosorbents has increased. Thus the number of research on the applicability of agricultural waste has also increased, although the elemental concentrations of such waste when used as a biosorbent, are not investigated often. In the rice husk waste and its ash, researchers found that the main elements were C, O, Si and K, with smaller amounts of Ca, P, Mg and Cl [28, 29]. In the ground coffee waste and husks, the main elements were (in order of abundance) O, C, K, P, Mg, Ca and [29, 30]. The banana peel biosorbent had high concentrations of K and C and smaller concentrations of O, Ca, Si, P, S and Cl [31–33] while the mango peel analysed with EDX had only the C, O, K and Ca peaks visible [34, 35].

Biosorbent made of pomelo peel and sugar beet pulp as individual entities were also analysed by other researchers. Using the elemental analyser, Zhao and Chen (2020) found that the mass ratios of C, H and N in the unmodified pomelo peel were 40.55%, 6.134% and 1.441%, respectively [36]. Dinh et al. [37, 38] identified the only two visible peaks in the EDX spectra of the pomelo peel as O (52.59 wt%) and C (47.41 wt%). Sugar beet pulp biosorbent for uranium sorption was analysed by Nuhanović et al. [17] and it was found that the unmodified sugar beet consisted mostly of O and C,

Fig. 3 The particle size distribution of the biocomposite**Table 1** Average values and standard deviations of the elemental concentrations measured in the pellets by EDXRF

Elemental concentrations	K (ppm)	Ca (ppm)	Mn (ppm)	Fe (ppm)	Ni (ppm)	Cu (ppm)	Zn (ppm)	Pb (ppm)
Biocomposite	70,060 ± 4762	3893 ± 394	7.1 ± 0.4	42.6 ± 2.6	0.65 ± 0.25	4.20 ± 0.56	6.94 ± 0.92	<MDL ^a

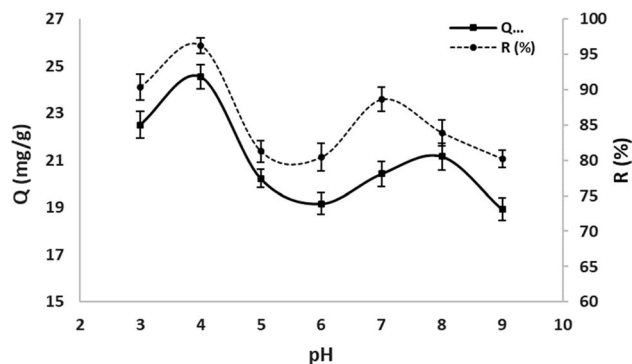
^a <MDL - below the minimum detection limit (1.1 ppm for Pb)

with Si, Ca, Cu, S, Mg and K in smaller amounts and with the maximum adsorption capacity of 19.78 mg g^{-1} [17].

In the current research, composite biosorbent was made of pomelo peel, sugar beet pulp and silica gel. Concentrations of the eight elements measured in the triplicate pellets are presented in the form of an average value \pm standard deviation and can be seen in Table 1. Using the EDXRF technique, it was found that the main elemental component of the used biocomposite was K, followed by Ca. In lesser amounts, Fe, Mn, Zn, Cu and Ni were also measured. Compared to the results of Nuhanović et al. [17], K is available in much higher concentration than Ca. Results obtained by EDXRF indicated that for uranium uptake by biocomposite except chemisorption involved mechanism could be also ion exchange.

Effect of solution pH

The pH of the solution is a crucial parameter for the sorption studies of metal ions, as it affects the solubility and speciation of metal ions, surface charge and binding characteristics of the adsorbent [39]. From Fig. 4 it can be observed that the maximum adsorption capacity (24.55 mg g^{-1}) and removal efficiency (96.28%) for U(VI) was achieved at pH 4. The pH value of the zero point of charge for a biocomposite based on pomelo peel and sugar beet pulp was 2.49 and this value is lower than the experimentally determined optimal pH value at which the maximum adsorption capacity is reached, which means that the surface of the biosorbent will take a negative charge at determined optimal pH value. This further means that at a pH of the solution greater than pH_{pzc} ($4 > 2.49$) the change in surface polarity results in a favourable attractive electrostatic interaction between the

**Fig. 4** Effect of pH on U(VI) adsorption capacity onto biocomposite ($m = 100 \text{ mg}$; $C_0 = 50 \text{ mg L}^{-1}$; $V = 50 \text{ mL}$; $T \approx 25 \text{ }^\circ\text{C}$; $t = 60 \text{ min}$; shaking at 95–100 movements per min)

negatively charged biosorbent surface and the positively charged uranyl species which was responsible for maximum adsorption performance at pH 4. Additionally, when the pH of the solution is low enough, divalent UO_2^{2+} is the dominant species of U(VI) in the solution. However, as the pH increases, the proportion of UO_2^{2+} in the solution decreases, while the proportion of monovalent hydrolysed species UO_2OH^+ and $(\text{UO}_2)_3(\text{OH})_5^+$ increases, and monovalent metal species have an even greater affinity in ion exchange with protons because they can replace individual protons at separate binding sites on the biomass. Therefore, the increase in pH indicates good agreement for U(VI) adsorption due to the increase in the number of monovalent ions [40]. The decrease in the removal efficiency of U(VI) at higher pH values based on pH_{pzc} is because at $\text{pH} > \text{pH}_{\text{pzc}}$ the surface of the biocomposite is negatively charged, which makes it, repulsive to anionic uranyl species such as

$\text{UO}_2(\text{OH})_3^-$, $\text{UO}_2(\text{OH})_4^{2-}$. Additionally, it could be due to the presence of insoluble inorganic carbon such as atmospheric CO_2 the formation of soluble ionic forms of uranyl carbonate ions, $\text{UO}_2(\text{CO}_3)_2^{2-}$ and/or $\text{UO}_2(\text{CO}_3)_3^{4-}$ as well as $(\text{UO}_2)_2(\text{CO}_3)(\text{OH})_3^-$, which become the dominant stable anionic forms of uranium and to which the biosorbent used as a cationic ion exchanger don't have an affinity [41]. Also, at high pH, insoluble forms of uranium are formed (e.g. schoepite, $4\text{UO}_3 \cdot 9\text{H}_2\text{O}$) which reduce the overall efficiency of U(VI) biosorption at higher pH values [42]. Regarding all the above, pH 4 was selected as the optimal value and used in further testing.

Effect of biocomposite amount

As presented in Fig. 5 the adsorption capacity decreased with the increase of biocomposite amount. The maximum adsorption capacity (46.2 mg g^{-1}) and removal efficiency (97.72%) was achieved with 50 mg of biocomposite dose. Decreased adsorption capacity of U(VI) ions can be due to partial aggregation and screening influence on the biocomposite surface which occurs at high biosorbent amount. Also, due to the increase of biocomposite amount mixing efficiency, the mass exchange could be slowed down [43]. Regarding the above, 50 mg was selected as the optimal mass of biocomposite for further biosorption experiments.

Adsorption kinetics

Kinetic modelling of the sorption process provides an insight into the velocity of the process and the mechanism of sorption, which includes mass transfer, diffusion and reaction on the surface of the biosorbent. The contact time influence onto uranium biosorption is presented in Fig. 6. Initially (0 to 15 min) the adsorption capacity increases to 44.8 mg g^{-1} . At the interval of 15–60 min these parameters value changes slowly, while after 60 min equilibrium

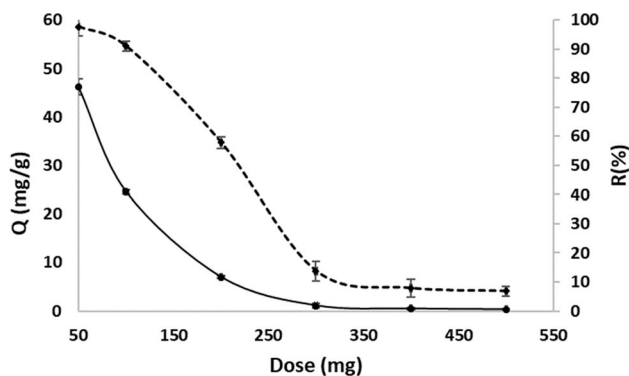


Fig. 5 Effect of biocomposite amount on U(VI) adsorption capacity onto biocomposite ($C_0=50 \text{ mg L}^{-1}$; pH 4; $V=50 \text{ mL}$; $T \approx 25^\circ\text{C}$; $t=60 \text{ min}$; shaking at 95–100 movements per min)

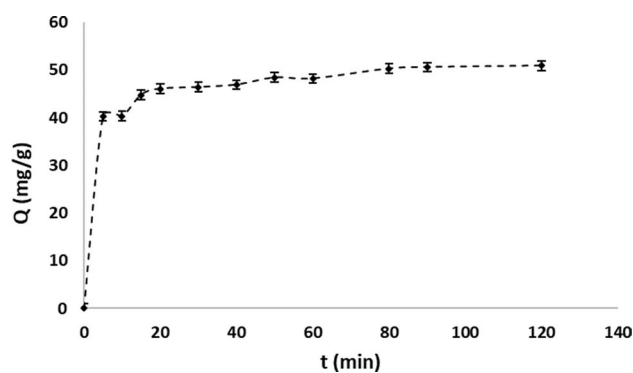


Fig. 6 Effect of contact time on U(VI) adsorption capacity ($C_0=50 \text{ mg L}^{-1}$; $m=50 \text{ mg}$; $V=50 \text{ mL}$; $T \approx 25^\circ\text{C}$; shaking at 95–100 movements per min)

is reached and maximum uranium sorption capacity of 48.2 mg g^{-1} was obtained. Finally, no significant changes in adsorption capacity were observed after 80 min. Based on the collected data, it can be concluded that the sorption process takes place through three stages. The first and fast stage (0–15 min) as a result of high free active sites concentration on the biosorbent surface and due to high initial metal concentration as a driving force. The second slower stage as a consequence of a free active centers number decrease and electrostatic repulsion which can also occur, by already bounded uranyl ions on the biocomposite [44]. At the last stage (80–120 min), a slight increase in the adsorption capacity (for only 2 mg g^{-1}) was the result of the mainly taken active sites on the biosorbents surface.

Applicability of the three commonly used kinetic models: Eq. (3) pseudo-first order model [45], Eq. (4) pseudo-second order model [46] and Eq. (5) intraparticle diffusion model [47] to the experimentally obtained results was investigated.

$$\ln(q_e - q_t) = \ln q_e - k_1 \cdot t \quad (3)$$

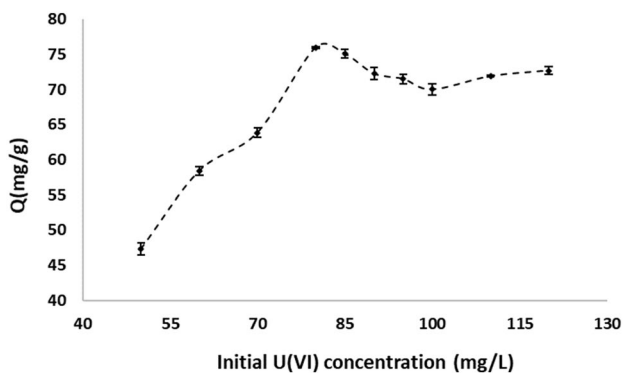
$$\frac{t}{q_t} = \frac{1}{k_2 q_c^2} + \frac{1}{q_e} \cdot t \quad (4)$$

$$q_t = k_{in} \cdot t^{\frac{1}{2}} + \quad (5)$$

where q_e and q_t (mg g^{-1}) are the amounts adsorbed at equilibrium and at time t , respectively and k_1 (min^{-1}), k_2 ($\text{g mg}^{-1} \cdot \text{min}^{-1}$) and k_{in} ($\text{mg g}^{-1} \text{min}^{-1/2}$) are the rate constants of the pseudo-first order, pseudo-second order and the intraparticle diffusion kinetic model, respectively, while C (mg g^{-1}) is the constant of the model in the function of the boundary layer thickness.

Table 2 Kinetic parameters for uranium (VI) removal by biocomposite at $25.0 \pm 0.5^\circ\text{C}$ and pH 4.0

Sorbent	$q_{e,\text{exp}}$	Pseudo-first order			Pseudo-second order			Intraparticle diffusion					
		$q_{e,\text{cal}}$	k_1	R^2	$q_{e,\text{cal}}$	k_2	R^2	k_{int1}	R_1^2	k_{int2}	R_2^2	k_{int3}	R_3^2
Biocomposite	51.9	12.3	0.033	0.946	51.5	0.00679	0.999	2.67	0.826	1.24	0.902	0.858	0.812

**Fig. 7** Effect of initial U(VI) concentration on adsorption capacity ($m=50$ mg; pH 4; $V=50$ mL; $t=60$ min; $t \approx 25^\circ\text{C}$; shaking at 95–100 movements per min)

Linear correlation coefficient (R^2) values, as a measure of experimental data matching with the each proposed kinetic model, are shown in Table 2.

Pseudo-second-order model correlation coefficient (0.999) was higher in comparison to pseudo-first-order (0.946) and intraparticle diffusion kinetic models (0.826; 0.902; 0.812) with the closest calculated q_e value (51.5 mg g^{-1}) to the experimental (51.9 mg g^{-1}). The limiting step of the process (according to this model) is a chemisorption with a fast dynamic (equilibrium reached after 60 min). Additionally, the second intraparticle diffusion models phase also showed a good agreement with the obtained results describing sequential ion sorption, whereby the intraparticle diffusion of the ions into internal biosorbents channels and cavities limits the process velocity. Furthermore, ion exchange with more difficult replacing ions occurs, as well as the analyte binding to the active centers of biocomposite.

A complex mechanism of this U(VI)-biocomposite removal process could be defined as kinetically controlled by chemisorption combined with complexation and ionic exchange [48].

Adsorption isotherm

The initial U(VI) concentration in solution was varied in interval 50–120 mg L^{-1} at room temperature ($25 \pm 5^\circ\text{C}$). According to the obtained results (Fig. 7), 80 mg L^{-1} was chosen as the optimal initial U(VI) concentration, with the maximum adsorption capacity of 75.99 mg g^{-1} .

Langmuir [49], Freundlich [50] and Temkin [51] adsorption isotherm models are expressed by linearized Eqs. (6, 8 and 9), respectively, fitting the experimental data:

$$\text{Langmuir: } \frac{1}{q_e} = \left(\frac{1}{K_L q_{\text{max}}} \right) \frac{1}{c_e} + \frac{1}{q_{\text{max}}} \quad (6)$$

Using Langmuir isotherm, a non-dimensional separating factor, RL , describes the affinity between the analyte and biosorbent indicating the nature of the biosorption mechanism, which is given by the equation:

$$RL = \frac{1}{1 + K_L c_0} \quad (7)$$

The RL values in the range of $0 < RL < 1$ mark the biosorption process as favorable [49].

$$\text{Freundlich: } \log q_e = \log K_F + \frac{1}{n} \log c_e \quad (8)$$

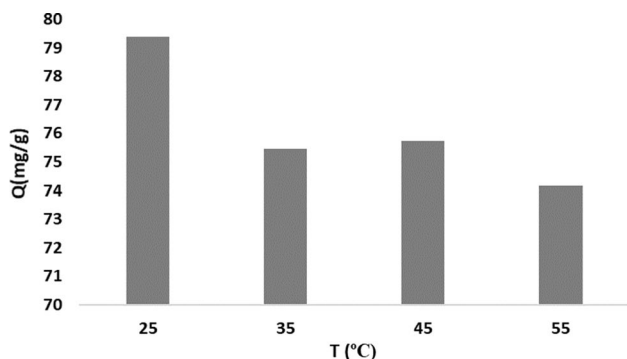
$$\text{Temkin: } q_e = \left(\frac{RT}{b} \right) \ln(A_T) + \left(\frac{RT}{b} \right) \ln(c_e) \quad (9)$$

where equilibrated amount of analyte adsorbed per unit weight of sorbent material is q_e (mg g^{-1}), equilibrium concentration after adsorption is c_e (mg L^{-1}), maximum biosorption capacity is q_{max} (mg g^{-1}), Langmuir's isotherm constant associated with free energy of adsorption is K_L (L g^{-1}), Freundlich's constants indicating adsorption capacity are K_F (mg g^{-1}) (L mg^{-1}) $^{1/n}$ and n (g L^{-1}), Temkin's equilibrium constant related to the maximum binding energy is A_T (L g^{-1}), the indicator of the heat of biosorption process is b (J mol^{-1}), the temperature is T (K) and a gas constant (8.314 J mol^{-1} K^{-1}) is R [51, 52]. The calculated adsorption isotherm parameters are presented in Table 2.

According to the obtained linear correlation coefficient values (Table 3), the sorption mechanism of U(VI) fits best to Langmuir model ($R^2=0.979$), regarding a sorption to a finite number of identical sites with a negligible interaction between adsorbed molecules. Therefore, U(VI) sorption onto biocomposite takes place on an energetically homogeneous surface (monolayer). The calculated maximum adsorption capacity, $q_{e,\text{max}}=79.36$ mg g^{-1} matches to the experimental value, $q_{e,\text{exp}}=75.99$ mg g^{-1} . Additionally, the sorption process of U(VI) onto used biocomposite is favorable due to separation coefficient value, $RL=0.0147$. Many similar studies reported that uranium sorption is

Table 3 Langmuir, Freundlich and Temkin isotherm parameters

Biosorbent	Langmuir				Freundlich			Temkin			$q_{e,exp}$
	$q_{e,max}$	K_L	R_L	R^2	K_F	$1/n$	R^2	RT/b	A_T	R^2	
Biocomposite	79.36	1.34	0.0147	0.979	53.06	8.38	0.814	320.65	9.34×10^{-4}	0.846	75.99

**Fig. 8** Effect of temperature on uranium adsorption capacity onto biocomposite ($C_0=80 \text{ mg L}^{-1}$, $t=60 \text{ min}$, $\text{pH } 4$, $m=50 \text{ mg}$, $V=50 \text{ mL}$, shaking at 95–100 movements per min)

better described by Langmuir's model than other models [17, 25, 53].

Adsorption thermodynamics

Temperature influence on U(VI) removal was investigated at four different values: 25, 35, 45 and 55 °C. The results shown in Fig. 8 indicate a temperature-independent U(VI) biosorption onto biocomposite with a maximum adsorption capacity of 79.38 mg g^{-1} . Regarding this, uranium adsorption capacity decreased as temperature raised. Therefore, the room temperature (25 °C) was chosen as an optimal and most suitable. As a not heat demanding process, the proposed removal system is very favorable in comparison to others [17, 54].

The following equations were used for a calculation of a thermodynamic parameters (shown in Table 4):

$$\Delta G = -RT \ln K_c \quad (10)$$

$$K_c = \frac{C_{Ac}}{C_e} \quad (11)$$

$$\log K_c = \frac{\Delta S}{2.303R} - \frac{\Delta H}{2.303R} \quad (12)$$

Table 4 Thermodynamics parameters (ΔH , ΔS , ΔG)

Biosorbents	$\Delta H \text{ (kJ mol}^{-1}\text{)}$	$\Delta S \text{ (J mol}^{-1} \text{ K}^{-1}\text{)}$	$\Delta G \text{ (kJ mol}^{-1}\text{)}$			
			298 K	308 K	318 K	328 K
Biocomposite	-38.637	0.1419	-72.62	-80.75	-82.50	-83.76

$$q_e = \left(\frac{RT}{b}\right) \ln(A_T) + \left(\frac{RT}{b}\right) \ln(c_e) \quad (13)$$

where change in the Gibbs free energy is $\Delta G \text{ (kJ mol}^{-1}\text{)}$, temperature is $T \text{ (K)}$, gas constant ($8.314 \cdot 10^{-3} \text{ kJ mol}^{-1} \text{ K}^{-1}$) is R , equilibrium constant is K_c , equilibrium concentration in the solution is $C_e \text{ (mg L}^{-1}\text{)}$ and the equilibrium concentration of the metal ion on the adsorbent is $C_{Ac} \text{ (mg L}^{-1}\text{)}$. Enthalpy change ($\Delta H, \text{ kJ mol}^{-1}$) and entropy change ($\Delta S, \text{ J mol}^{-1} \text{ K}^{-1}$) values were obtained from the slope and intercept of Vant Hoff's plots of $\log K_c$ versus $1/T$ [55].

Obtained thermodynamic data showed that the biosorption process of U(VI) by used biocomposite was spontaneous (high negative values of ΔG), exothermic (negative ΔH value $-38.637 \text{ kJ mol}^{-1}$), which also indicates that the energy released due to U(VI) binding onto sorbent is higher than the energy required for dehydration of metal ion [56]. Additionally, randomness increases as the reaction proceeds and the biosorption of U(VI) ions onto utilized biocomposite is a favorable process (suggested by positive ΔS value).

Desorption study

Desorption study is very important for the recovery of the adsorbed uranium and for the regeneration of the used biosorbent, which are important parameters for the practical application of the biosorption process. Additionally, the regeneration of the used biosorbents makes the process overall economically favorable. Three different desorption solutions were tested for the desorption study (Table 5).

NaHCO_3 showed the best results out of all tested eluents and was further investigated at three different concentrations (0.25; 0.5; 1.0). According to the obtained results, 1.0 M

Table 5 Removal efficiency after 1 cycle with different eluents

Eluents	1.0 M HNO_3	1.0 M NaHCO_3	1.0 M citric acid
Desorption efficiency (%)	44.18	95.98	43.35

NaHCO_3 showed the highest desorption efficiency and it could be used for at least three cycles (Fig. 9). Obtained results in this study are opposite with the results for the biocomposite composed of *Ulva* sp. and Na bentonite [14], amidoximated *Saccharomyces cerevisiae* [57] and *Trichoderma harzianum* [58] for which the highest desorption efficiency was obtained with eluents, HNO_3 and HCl .

These results have shown that biocomposite based on pomelo peel and sugar beet pulp has a very good potential for the removal of U(VI) ions and its recovery.

Mechanism modelling

According to the results obtained from modelling data (biocomposite characterizations; adsorption kinetics; adsorption isotherms; adsorption thermodynamics) it could be observed that the mechanism of the U(VI) binding to the biocomposite is a complex and multi-step process [59]. FTIR spectra after sorption indicated that predominantly hydroxyl and amino groups are participating in the binding of uranyl ions, and a new peak at 914 cm^{-1} showed a presence of UO_2^{2+} onto biocomposite after sorption. EDXRF analysis showed that on the surface of biocomposite are present some of metal ions which can be exchanged with uranyl ions (Ca, K etc.) indicating possibility of ion exchange as one of the steps of the mechanism.

Furthermore, data showed that the process takes place through the three phases; equilibrium is reached very fast; the limiting step in the process is probably chemisorption and considering obtained data for intraparticle diffusion model, kinetic is not only controlled by one but by diverse processes. The process is temperature independent and occurs on the monolayer surface (Langmuir model). Even though according to pseudo-first order model the limiting step should be chemisorption, thermodynamic data are suggesting contrary. Thermodynamic data revealed that the process is spontaneous (negative ΔG values) and exothermic (negative ΔH value). However, physisorption occurs if the change in Gibbs free energy is in the range:

-20 to 0 kJ mol^{-1} , both physisorption and chemisorption occurs if values of ΔG are in the range: -20 to -80 kJ mol^{-1} and chemisorption occurs if ΔG values are in the range: -80 to 400 kJ mol^{-1} ; so this data can indicate possible mechanism between sorbate and sorbent [60]. Calculated values of change in Gibbs free energy in the present study indicates both physisorption and chemisorption are occurring (Table 4). Furthermore, if $\Delta H > 0$ the main mechanism is a chemisorption, and if $\Delta H < 0$ the main mechanism is physisorption [61]. In the present study value obtained for ΔH (-38 kJ/mol) indicates exothermic nature of the process and physisorption. Considering that the capacity of adsorption (Fig. 8) decreased with an increase of temperature indicates also that physisorption occurred [62]. These results suggest both physisorption and chemisorption occurs on the surface of the used biocomposite at the same time and a layer of molecules may be adsorbed by physical forces on the top of an underlying layer with chemisorbed molecules [62–64].

With respect to all mentioned above, sorption of U(VI) onto biocomposite is a multi-phase process which is occurring with possible participation of both physisorption/chemisorption, ion-exchange, and surface complexation.

Comparison of adsorption capacities

The comparison of adsorption capacity (q_{exp} , mg g^{-1}) of biocomposite used in this research with the adsorption capacities of various biosorbents and adsorbents reported in previous researches are given in Table 6 [17, 21, 39, 65–67]. According to the results obtained in this study, biocomposite could be considered as a potential sorbent for U(VI) removal from wastewaters, due to the adsorption capacity (75.99 mg g^{-1}) which is higher than many other sorbent materials. In addition, biocomposite is comprised of two types of agricultural wastes, pomelo peel and sugar beet pulp which are biodegradable, abundant, and low-cost material for which preparation is required minimum chemical consumption.

Fig. 9 Desorption efficiency after I, II and III cycles of desorption/adsorption with the three different concentrations of the eluent (NaHCO_3)

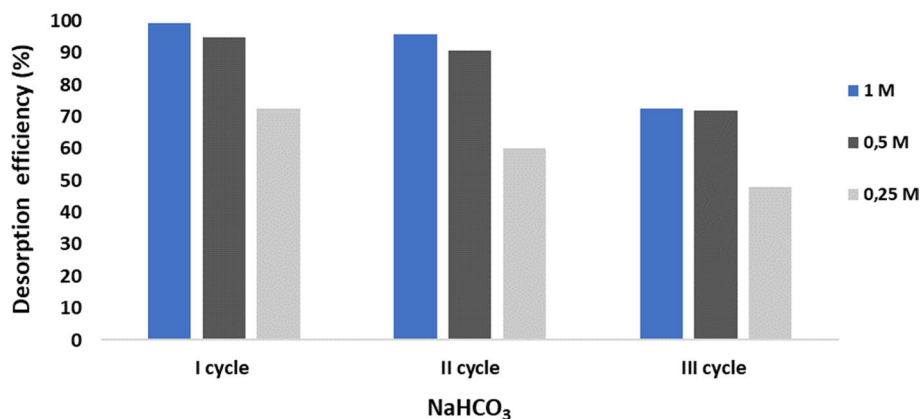


Table 6 Comparison of adsorption capacities of various sorbents for U(VI) removal

Sorbents material	Adsorption capacity (mg g ⁻¹)	References
Modified pomelo peel	270.71	[65]
Native sugar beet pulp	20.45	[17]
ACSD composite adsorbent (alginate, sepiolite, CaSO ₄ ·xH ₂ O and diatomite)	3.51	[66]
<i>Ceratophyllum demersum</i>	140.45	[39]
Chitosan-modified zeolite	16.4	[67]
Bi-functionalized biocomposite adsorbent	43.2	[21]
Biocomposite (pomelo peel and sugar beet pulp)	75.99	Present study

Conclusions

In this study, a biocomposite sorbent composed of sugar beet pulp and pomelo peel was employed for the biosorption of U(VI) from aqueous solution. According to the obtained results prepared biocomposite can be used as a sorbent for the removal of U(VI) from the aqueous solution under the optimum conditions of pH 4, biocomposite dosage 50 mg, initial uranium concentration 80 mg L⁻¹, contact time 60 min, temperature 298 K with the maximum adsorption capacity of 75.99 mg g⁻¹. The equilibrium data fitted best to the Langmuir isotherm model and pseudo-second order. The results of the thermodynamic analysis indicated that the biosorption process of U(VI) uptake was exothermic and spontaneous. The biocomposite characterization confirmed its removal efficiency due to the numerous functional groups, particle size distribution and pH_{pzc} value that is in accordance with the pH of the solution (negatively charged surface of biocomposite). Additionally, the adsorbed U(VI) desorbed quantitatively by 1.0 M NaHCO₃ and biosorbent can be reused for three and more cycles. Regarding all the above, the involved mechanism in uranium binding is complex and includes both physisorption/chemisorption, ion exchange and surface complexation. The investigated biocomposite is a low cost and environmentally friendly sorbent with high potential for efficient U(VI) removal and recovery from aqueous solution.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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