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ORIGINAL PAPER

Highly efficient and nature-based removal of radionuclides from environmental water

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Abstract

Radioactive elements released into the environment have been an imperative environmental issue and poses health hazards for living organisms. Activated carbon is one of the most common adsorbents involved in radioactive wastewater remediation due to its high specific surface area and high degree of surface reactivity. In this study, three alternative activated carbons, previously produced via thermo-chemical activation using phosphorous acid and derived from apricot (*Prunus armeniaca*), plum (*Prunus domestica*), and cherry/sour cherry (*Prunus avium*/*Prunus cerasus*) kernels were evaluated for their effectiveness in removing radioactive substances. Water from a uranium mine at Žirovski Vrh, characterised by high resolution gamma spectrometry, was chosen for the experiment due to its rather high content of radionuclides, especially uranium. The efficiency of these carbonous materials was compared to a commercial activated carbon. During the experiments the remediation performance was monitored by determination of the gross alpha/gross beta activity, which can be used as fast and cost-efficient screening method for the calculation of decontamination level. Activated carbon from plum kernels showed remarkable achievement lowering alpha and beta activity for 95% and 96%, respectively. Radionuclide removal was most effective under neutral to alkaline conditions, achieving efficiencies > 85% for alpha emitters and > 95% for beta emitters. Desorption was successful with a small amount of 0.1 M H_2SO_4 and 0.1 M H_3PO_4, respectively, proving that the tested materials can be used repeatedly for recovery of uranium from waters. Our results emphasised the importance of different alternative carbonous materials potential in radioactive pollutant removal and recovery.

Keywords Activated carbon from biomass · Adsorption · Environmental remediation · Radionuclide removal · Uranium recovery · Wastewater treatment

Introduction

Natural occurring radionuclides (NOR) are mostly present in the environment as primordial radionuclides (e.g., K-40), members of radioactive uranium, thorium or actinium decay chains or products of cosmic reactions with the Earth's atmosphere. Many industries such as uranium mining, the nuclear fuel cycle industry, the coal industry, the oil and gas industry, metal mining and smelting, mineral sand mining (i.e., rare earths, titanium, zirconium), the fertiliser industry (i.e., phosphate) or the construction industry use natural sources and generate waste with elevated or altered levels of NOR, mainly referring to members of the decay chains with numerous alpha and beta emitters, many of which have a high chemical and radio-toxicity (Hossain 2020; Mrdakovic Popic et al. 2023). Subsequently, the presence of NOR material (NORM) can pose a potential health risk to workers, the public and the environment; therefore its levels should be

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monitored and controlled. In addition, (low-level) radioactive liquid waste can be generated during the production of primary products or during remediation activities, which can pose a serious problem, especially due to their considerable volume (Chalupnik et al. 2001; Eriksen et al. 2006). Therefore, the content of contaminated substances should be reduced to clearance or an acceptable level before release into the environment otherwise insufficiently treated water can have negative effects on biota and human health (Turcanu et al. 2022; Popic et al. 2023).

To minimise the environmental impact of industries that generate radioactive liquid waste, cost-effective, more efficient and environmentally friendly methods should be investigated and promoted. There are various operations such as chemical precipitation, flocculation, filtration, ion exchange, electrochemical treatments, adsorption, membrane separation, etc. (Zhang et al. 2019; Hossain 2020; Chakraborty et al. 2022; Ma et al. 2023) that can be used. The advantages of adsorption are the simplicity of operation and high purification efficiency (Bhattacharyya and Gupta 2008). Commercially available activated carbon (AC) is a preferred adsorbent to remove impurities from liquid solutions. Its widespread use in water, air and soil purification and enrichment systems is limited due to its high cost in some parts of the world. Therefore, alternative, non-conventional adsorbents have been produced from natural, industrial and waste materials. Additionally, the use of waste products for water decontamination aligns with the principles of a circular economy promoted by the EU. These types of research have significantly increased (Zhang et al. 2020; Liu et al. 2022) in recent years, including radionuclide removal (Rae et al. 2019; Smječanin et al. 2022b; Banerjee et al. 2022; Xiong et al. 2025).

Recently, lignocellulosic biomass from plum (*Prunus domestica* L.) (Pap et al. 2017), apricot (*Prunus armeniaca* L.) (Turk Sekulić et al. 2018) and cherry/sour cherry (*Prunus avium* L./*Prunus cerasus* L.) (Pap et al. 2016) kernels have been used as a source for activated carbon production and for removal of heavy metals such as Cr^{2+}, Pb^{2+}, Cd^{2+} and Ni^{2+}. These materials are inexpensive and locally available from fruit plantations as residue from the food and beverage industry (Nieto-Delgado and Rangel-Mendez 2011; Pap et al. 2018). Since radionuclides can behave chemically the same or similar to heavy metals, such biomass-derived activated carbons may also be suitable for radionuclide removal from wastewater. In addition, desorption using diluted inorganic acids may allow recovery of valuable elements such as uranium.

To obtain a more realistic representation of the conditions in natural ecosystems and real-world applications, the adsorption capacity of the tested materials for the removal of radionuclides was investigated directly on water from closed Žirovski Vrh uranium mine, where elevated activity concentrations are expected. The obtained results were compared with commercially available AC. Further test such as effect of solution pH on removal efficiency as well as desorption of radionuclides were done for the most effective adsorbent.

Monitoring wastewater decontamination should be both rapid and economically viable. To determine radionuclide activity concentrations most often radiometric methods are used. Recently, the advantages of liquid scintillation counting (LSC) compared to gamma spectrometry and other techniques have been emphasised. It offers direct measurement of water samples, low background count rates and high efficiencies, up to 100% for alpha particles (Ajemigbitse et al. 2020). In addition, modern LSC instruments are equipped with alpha/beta discrimination, which allows simultaneous analysis of alpha and beta emitters by pulse shape analysis (Li et al. 2022). Therefore, the aim in this study is twofold: to evaluate the efficiency of fruit kernel-derived activated carbons for the removal and potential recovery of radionuclides from mine water and to assess the applicability of LSC for rapid and reliable monitoring of gross alpha and gross beta activity concentrations in treated and untreated water samples. The investigation began in 2017 and was later continued in 2023 with experiments focused on evaluating the performance of plum kernel based AC.

Materials and methods

Materials

The alternative activated carbons (AC) used were produced from apricot, plum and cherry/sour cherry kernels, washed with distilled water, crushed in a mechanical mill, dried and impregnated with 50% aqueous H_3PO_4 solution. The impregnated samples were then placed in an electric furnace. The final annealing was carried out at 500 ^{O}C. The exact preparation of AC was explained in previous studies (Pap et al. 2016, 2017; Turk Sekulić et al. 2018). For the comparison, commercially available AC (Sutcliffe Speakman Carbon Limited, Ashton-In-Makerfield, UK) was also used. This activated carbon produced from bituminous coal, is available as pellet grinded in the range 12 × 40 mesh and mostly used for wastewater treatment.

The water from the former uranium mine in Žirovski Vrh, Slovenia, contains higher activities of naturally occurring radionuclides, especially uranium isotopes.

All other chemicals used in the experiments were of analytical grade and were not further purified. The Milli-Q water used for the preparation of solutions was obtained from a Direct-Q 5 system (Millipore, Watertown, MA, USA).

Material characterisation

The carbonous materials have been fully characterised through determination of elemental content, specific surface area, morphology by SEM–EDX, surface chemistry by FTIR, etc. given in following papers: Pap et al. (2016) for AC produced from sour and sweet cherry kernels, Pap et al. (2017) for AC produced from plum and Turk Sekulić et al. (2018) for AC produced from apricot stones. The summarised characteristics of the materials, i.e., CCAC (from sweet and sour cherry), PAC (from plum), AAC (from apricot), and commercial AC (CAC) are provided in (Table S1), while their textural properties are given in (Table S2) in the Supplementary Material.

Material characterisation regarding radionuclide content was performed by high-resolution gamma spectrometry (HRGS) in an ISO 17025 accredited laboratory. The materials were filled into a tightly sealed plastic vessel (fi 90, 8×4) weighing 50 ± 12 g and set aside for 20 days prior to the measurements in order to establish a secular equilibrium between parent radionuclides and progenies. The manufacturers and specifications of the detectors, measurement time and sample geometry are given in (Supplementary Material, Table S3). Water sample from Žirovski Vrh was also characterised by HRGS prior the experiment. For this purpose, 8 L of water were evaporated to dryness using an automated evaporator under strictly dynamically controlled conditions, maintaining a temperature of 65 °C and precise regulation of airflow and pressure. The dry residue was pressed in plastic cylindrical container. The sample was measured after the secular equilibrium was established.

Adsorption experiment

For the adsorption experiments batch technique was used. In Erlenmeyer flasks, 100 mg of each type of AC was in contact with 50 mL of the water with higher radioactivity. Prior contact, to increase the adsorption performance, the pH of the water was adjusted to 6 with 0.5 mL of 1% HCl and a drop of 1% NH_4OH for pH correction. After shaking for 30 min (140 rpm) with a Heidolph Unimax 1010 horizontal shaker (Heidolph, Germany), the solution was separated from the activated carbon by filtration through Macherey–Nagel filter paper (MN 640 m) and prepared for liquid scintillation counting.

Removal efficiency of alpha or beta emitters was calculated according to Eq. (1):

$$Removal efficiency\left(A_{\alpha(\beta)}\right) = \frac{A_{\alpha(\beta)0sum} - A_{\alpha(\beta)sum}}{A_{\alpha(\beta)0sum}} \times 100 \quad (1)$$

where $A_{\alpha(\beta)0sum}$ is total activity of alpha (α) and beta (β) emitters of untreated water, respectively, $A_{\alpha(\beta)sum}$ is total activity of alpha (α) and beta (β) emitters of treated water, respectively.

Desorption experiment

AC with the best adsorption properties were selected for further tests of radionuclide desorption. After adsorption of radionuclides on PAC, desorption efficiency was tested by adding the two portions of 5 mL and 8 mL of 0.1 M hydrochloric, nitric, phosphate and sulfuric acid, respectively, to 0.1 g of PAC with adsorbed radionuclides. First portion of 5 mL was removed by filtration after 15 min, while the second portion of 8 mL was left in contact with PAC overnight. The filtrate was mixed with scintillation cocktail Ultima Gold AB, PerkinElmer, prior counting by LSC.

Desorption efficiency of alpha or beta emitters was calculated according to Eq. 2:

$$\left(A_{\alpha(\beta)}\right) = \frac{A_{\alpha(\beta)desorbed}}{A_{\alpha(\beta)adsorbed}} = \left(\frac{A_{\alpha(\beta)desorbed}}{\left(A_{\alpha(\beta)0}sum - A_{\alpha(\beta)}sum\right)/V_{measured} \cdot V_{total}}\right) \times 100 \quad (2)$$

where $A_{\alpha(\beta)desorbed}$ is total activity of alpha (α) and beta (β) emitters desorbed from the PAC, while $A_{\alpha(\beta)adsorbed}$ is total activity of alpha (α) and beta (β) emitters adsorbed on 0.1 g of PAC from 50 mL of the water sample, $V_{measured}$ is an aliquot taken for LSC analysis, V_{total} is sample volume in contact with PAC.

Optimization of pH

The efficiency of radionuclide removal by PAC was further investigated across different pH ranges (pH 3 to 9). Samples were acidified with HNO_3 or alkalised with NaOH to adjust the pH. The samples were then mixed with PAC and analysed as described in Sect. "Adsorption experiment".

Gross alpha and beta determination by liquid scintillation counting

The gross alpha and gross beta activities were determined in untreated and treated water samples to determine adsorption/desorption efficiency. For this purpose, 6 or 8 mL of each sample was transferred to low-potassium borosilicate glass vials (PerkinElmer, USA). The pH was adjusted to 1 with 1% HNO_3. The sample was mixed with 14 mL or 12 mL of Ultima Gold AB liquid scintillation cocktail, shaken vigorously and checked for sample homogeneity. The samples were counted five times for 80 min before adsorption and 400 min after adsorption. All experiments were carried out in duplicate.

Gross alpha and beta measurements were performed on ultra low-level Liquid Scintillation Counter (LSC), Quantulus 1220 (PerkinElmer). PSA was set to the optimal position by the usage of ^{241}Am and ^{90}Sr standard solution to distinguish between alpha and beta pulses. Am-241 was provided by CMI (Czech Metrology Institute, Czech Republic) and Sr-90 by Eckert & Ziegler Analytics, USA. The same standard solutions were also used for the determination of counting efficiency. A pulse shape analyser (PSA) was used to perform a simultaneous alpha/beta gross counting, which allows the distinction between alpha and beta pulses. Standard solution of Am-241 and Sr-90 as alpha and beta emitters were used for calibrating detection efficiencies. The counting windows for beta emitters were set from 100 to 1,000 channels and 500 to 1,000 for detection of alpha emitters. Counting efficiencies were determined as 98% for gross alpha and 96% for gross beta activity. For spectra analyses and identification of peaks, certified standard solutions (CRM) of U-238 (Eckert Ziegler Analytics), Th-232 (CMI), Ra-226 (CMI) were used and prepared as water samples. Activity of U-238 and Th-232 was around 5 Bq per sample, while Ra-226 activity was around 0.2 Bq per sample. Standard Quench Parameter was in all measured samples between 780 and 785, therefore no quench curve was necessary for correction of counting efficiencies.

Gross alpha and gross beta activity calculation

Activity concentration of gross alpha c_α or gross beta, c_β, were calculated according to ISO 11704 (ISO 2018) by following Eqs. (3) and (4):

$$c_{\alpha/\beta} = \frac{r_{g\alpha/\beta} - r_{0\alpha/\beta}}{m\varepsilon_{\alpha/\beta}} = \left(r_{g\alpha/\beta} - r_{0\alpha/\beta}\right) \cdot w_{\alpha/\beta} \tag{3}$$

$$w_{\alpha/\beta} = \frac{1}{m \cdot \varepsilon_{\alpha/\beta}} \tag{4}$$

where α refers to gross alpha activity and β for gross beta activity; $c_{\alpha/\beta}$ is alpha/beta activity per mass (Bq/kg (L)); $r_{g\alpha/\beta}$ is sample gross count rate, from the alpha and beta windows, respectively; $r_{0\alpha/\beta}$ is sample gross count rate, from the alpha and beta windows, respectively; m is mass of the test sample (kg (L));$\varepsilon_{\alpha/\beta}$ is counting efficiency for alpha and beta, respectively.

Results and discussion

Characterisation of the materials

Basic characteristics of alternative ACs are discussed in previously published papers (Pap et al. 2016, 2017; Turk Sekulić et al. 2018) and summarised in Table S1 (Supplementary Material). All materials had lower ash and moisture content compared to commercial AC. Moisture adsorbed in the structure of activated carbon blocks the pores of the material, making them unavailable for the ions in the solution. Ash in activated carbon is an impurity and an undesirable product, which reduce the mechanical strength and porosity of activated carbon (Pap et al. 2017).

FTIR analyses had shown that the ACs contain hydroxyl, carboxyl and phosphorous functional groups on their surface as well as aromatics, amines and hydrocarbons which can increase adsorption of cations. All produced ACs had acidic pH_{pzc}, which means they contain more acid functional groups (carboxylic, lactonic, and phenolic) which can be easily deprotonated at $pH > pH_{pzc}$ to facilitate out-sphere complexation (i.e., electrostatic attraction) with positively charged cations. SEM analyses revealed heterogeneous surface and porous nature of all ACs. A high number of pores shows good possibility for radionuclides adsorption into the pores. The main structural properties (Supplementary Material, Table S2) revealed that among produced AC, the one produced from apricot kernels has the highest BET surface area, 1099 m^2/g and total pore volume 0.51 cm^3/g, plum AC 829 m^2/g and 0.42 cm^3/g while AC from cherry 657.1 m^2/g and 0.25 cm^3/g, respectively.

Gamma characterisation of water sample and activated AC

The results of gamma spectrometry analysis of high activity water sample are presented in Table 1. The sampled water contains much higher values of U-238 than surface water in that area where annual average values for U-238, Ra-226 and Th-230 were 245–255, 15–25 and 0.5–1.5 Bq/m^3, respectively (Križman et al. 1995). Since U isotopes have higher chemical toxicity rather than radiotoxicity, often the amount of total U is expressed in concentration. In the analysed sample, activity concentration of 39.87 Bq/L of U-238 corresponds to 3.2 mg/L. For comparison, the concentrations of U in natural waters are typically less than 4 µg/L in river water, around 3.3 µg/L in open seawater, and usually less than 5 µg/L in groundwater (Smedley and Kinniburgh 2023). According to EU Directive 2020/2184, the maximum permitted concentration of total uranium in drinking water is 0.3 µg/L (Union 2020), whereas Council Directive 2013/51/Euratom, which regulates radioactive substances in water intended for human consumption, sets the maximum derived activity concentration of U-238 in drinking water is 3 Bq/L (Council of the European Union 2013).

The activity concentrations of radionuclides which can be released in the environment should be calculated depending

Table 1 Activity concentrations of radionuclides in water sample from Žirovski Vrh, produced AC and commercial AC prior the adsorption experiments

Radionuclide	Water sample		AAC		PAC		CCAC		CAC	
	Activity concentration (mBq/L)	MDAC (mBq/L)	Activity concentration (Bq/kg)	MDAC (Bq/kg)	Activity concentration (Bq/kg)	MDAC (Bq/kg)	Activity concentration (Bq/kg)	MDAC (Bq/kg)	Activity concentration (Bq/kg)	MDAC (Bq/kg)
K-40	528 ± 57	40	N.D	4	3.1 ± 1.4	6	4.8 ± 3.4	12	26.5 ± 4.9	13
Pb-210	N.D	40	N.D	1.5	N.D	2.6	N.D	6	18.8 ± 2.1	8
Ra-226	239 ± 25	90	3.5 ± 1.0*	8	4.0 ± 1.1*	8	3.8 ± 0.5*	9	32.6 ± 2.8	8
Ra-228	N.D	140	N.D	1	N.D	0.8	1.1 ± 0.7	2	6.6 ± 0.9	4.6
U-238	39,870 ± 1,190	1,500	N.D	3.2	4.6 ± 3.0	2.6	N.D	9	27.1 ± 2.8	10.5

Value determined from radon progeny assuming that the radon exhalation factor from the sample is the same as for CAC

N.D. *Radionuclide not detected*, MDAC *Minimum detectable activity concentration*

on volume of liquid effluent, isotopes present etc. and depends on the origin of its production. Subsequently, permitted activity concentration in discharged waters of U and other NOR is not unique. However, it can be concluded that the sampled water contains higher concentrations of U-238 than clearance level for effluents which makes it suitable for the purposed research. The presence of radionuclides was also determined in the functionalised ACs prior to their use. Natural radionuclides were determined to be below or slightly above detection limits. Detectable amounts of U-238 were only found in CAC. Measurable amounts of Ra-226 were found in all materials, whereby the highest value of 32.6 ± 2.8 Bq/kg was again determined in commercial AC. The activity concentrations of Ra-228 determined in AC produced from biomass were much lower, namely below 1 Bq/kg. Ra-228 was only detected in commercial AC. As can be seen, all produced AC from kernels contain fewer radionuclides than the commercial product. The purity of the AC ensures that there is no additional input of radioactive elements through a possible ion exchange process during the water decontamination.

Adsorption of radionuclides on alternative activated carbons

The tested functionalised AC adsorbents used in this study have shown the best adsorption capabilities for heavy metals at a dose of 2 g/L (Pap et al. 2016, 2017; Turk Sekulić et al. 2018). Therefore, the same ratio was used for radionuclide adsorption testing. The results of the adsorption of radionuclides from 50 mL of water (pH = 6) onto 0.1 g of different activated carbons are shown in Table 2.

As evident from the results, the gross alpha and beta activity determined by LSC for untreated water sample is higher than what can be calculated from the results obtained through gamma spectrometry. Gamma measurements indicate that U-238 is the primary contributor to the gross alpha activity in the sample. Since U-238 is the parent isotope in the natural uranium decay series, the sample contains numerous shorter-lived progenies. Many of these decay products either do not emit gamma radiation or emit it at such low intensities that they cannot be effectively detected by gamma spectrometry. U-238 is likely in equilibrium with its progenies (e.g., Th-234 (beta emitter), Pa-234m (beta emitter), U-234 (alfa emitter), Th-230 (alfa emitter)) which contribute to the overall alpha and beta activity. To account for this discrepancy, gross alpha and beta activity can be detected using liquid scintillation counting, a technique particularly sensitive to low-energy alpha and beta emitters that might otherwise go undetected by gamma spectrometry. By gamma spectrometry only U-238 was determined while U-234 does not have gamma rays with high enough intensity and probability

Table 2 Gross alpha and gross beta activity in water from Žirovski Vrh and after contact with different AC

Water samples treated with different AC	Alfa emitters			Beta emitters		
	c_A (Bq/L)	MDAC (Bq/L)	Removal efficiency (%)	c_A (Bq/L)	MDAC (Bq/L)	Removal efficiency (%)
Untreated water	90.6 ± 4.7	0.31	–	77.2 ± 6.5	2.57	–
AAC	31.6 ± 1.4	0.24	65	46.1 ± 2.7	2.05	40
PAC	4.6 ± 0.4	0.22	95	3.0 ± 1.1	1.91	96
CCAC	34.1 ± 1.5	0.22	62	9.9 ± 1.6	1.91	87
CAC	18.0 ± 0.5	0.22	80	9.5 ± 1.9	1.91	88

*The results are average values of 2 replicates determination. MDAC *Minimum detectable activity concentration*

to be detected. If it is assumed that activity concentration of U-234 is close to U-238 activity concentration or even higher (Benedik et al. 2015), then their total activity is probably around 80 Bq/kg or more, which is closer to gross alpha activity concentration determined by LSC. Furthermore, if Th and Ra isotopes and their progenies are included, it can be confirmed that the total activity corresponds to the activity determined by the gross alpha and gross beta methods. The gross activity by the LSC method is determined from only 6 mL of the sample, which was slightly acidified to obtain pH close to 1 and simply mixed with the scintillation cocktail and counted for a certain number of minutes (5 × 80 min). Compared to gamma spectrometry, where 8 L of the water sample was evaporated to dryness, this means that the gross alpha/beta method can indeed be a fast and simple screening method for the determination of total activity in liquid waste samples.

A comparison of the gross alpha and gross beta activities before and after adsorption on the tested AC shows that the ratio of adsorbed alpha and beta nuclides was different. The best adsorption properties were obtained for PAC with equal removal efficiency of alpha and beta emitters, where the ratio of removal of alpha and beta emitters is 1. On CAC, less radionuclides were adsorbed under the given conditions, but the ratio of alpha and beta emitters adsorbed is still close to 1, which could mean that this material has a lower capacity than PAC, but the same selectivity under the given conditions. On the other hand, the lower adsorption of alpha emitters to AAC and CCAC could be explained by the higher selectivity of the materials. The most dominant beta emitters in the spectra are short-lived decedents from U-238 chain, Th-234 and Pa-234m. Both, thorium and protactinium were not efficiently adsorbed.

The results of adsorption of radionuclides on different AC show their different removal efficiencies. The most promising is PAC which efficiently removes more than 95% of alpha and beta emitters at a given ratio of activated carbon to water (2 g/L). CAC was also efficient, removing 80% and almost 90% of the gross alpha and beta activity, respectively. CCAC showed good removal efficiency of beta emitters of 87%, but alpha emitters were not as efficiently removed, only 62%. AAC exhibited similar efficiency of alphas removal, 65%, while only 40% of beta emitters were removed.

Effect of solution pH on adsorption on activated carbon produced from plum kernels

Since PAC showed the best adsorption properties, it was further used for the protonation study on the removal of radionuclides from environmental waters (Fig. 1). The lowest result was obtained at pH 2, while the highest removal efficiency was achieved at neutral to alkaline condition.

The point of zero charge (pH_{pzc}) of PAC was found to be pH 4.12 (Supplementary Material, Table S2), indicating the net surface charge, pH_{PZC}, is a characteristic of amphoteric surfaces and is determined by the nature of the functional groups. If the pH is above this value, the functional groups are more negatively charged and have a high affinity for oppositely charged ions, i.e., cations (Babić et al. 1999; Fiol and Villaescusa 2009). Below this value, on the other hand, the functional groups on the activated carbon are protonated, which explains the lower efficiency of radionuclide removal.

Uranium shows various oxidation states, but most abundant are + 4 and + 6. It is normally associated with oxygen and in + 6 oxidation state it appears as uranyl ion (UO_2^{2+}). In strongly reduced conditions, for example in presence of biomass material, U^{6+} will be reduced by functional groups on the biochar to U^{4+}, which exhibit even stronger adsorption. This is commonly supposed to be due to a Coulomb effect making the binding of U^{4+} complexes stronger than for the UO_2^{2+} complexes (McGowan et al. 2022). This could contribute to a still high removal efficiency of alpha emitters of about 75%. At a pH values, pH > 4, both total alpha and total beta activity in the purified water was reduced by more than 90%. Since all radionuclides present act as cations, this pH value is more favourable. As a result, the electrostatic repulsion of the ions decreases, which in turn leads to better adsorption (Smječanin et al. 2022a). In basic and neutral pH condition, $UO_2(OH)^+$, $(UO_2)_3(OH)_5^+$, $(UO_2)_4(OH)_7^+$, UO_2CO_3, and $UO_2(OH)_2$ are the dominant species of uranium radionuclide respectively, thereby any kind of positively charged

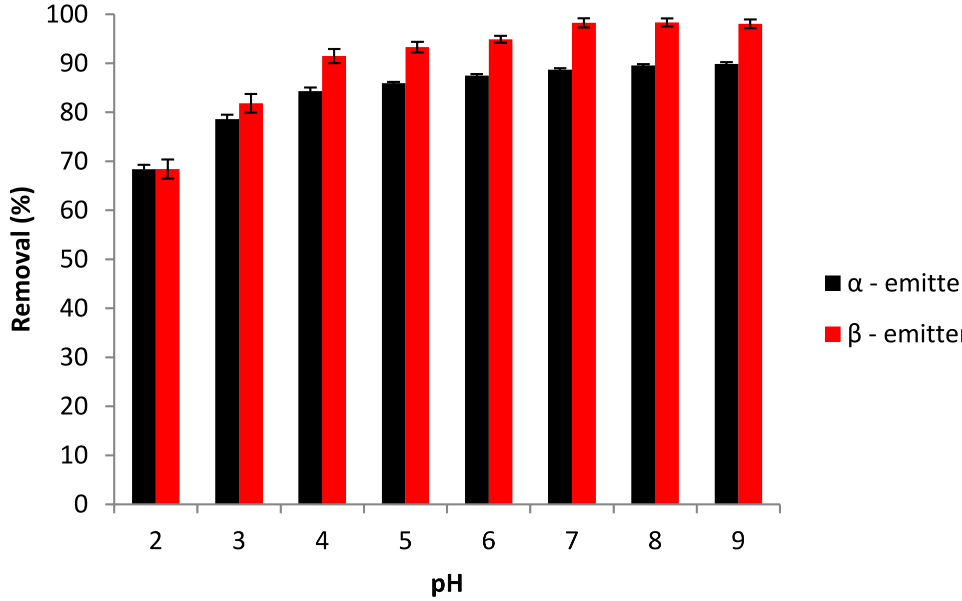

Fig. 1 Removal efficiency of PAC at different initial pH values of water sample from Žirovski Vrh (100 mg of PAC in 50 mL of the water shaking for 30 min at 140 rpm, the pH was adjusted with 1% HCl and 1% NH_4OH)

adsorbents can easily adsorbed U^{6+} ions through other chemical interactions (Banerjee et al. 2022) which results in high efficiency of U removal from the solution.

After the adsorption, the final pH value of the decontaminated water was around pH 5.5–6, as it is for deionised water, which means that no additional pH adjustment is required after treatment, but that in addition to removing the toxic chemical, a neutralisation process can be carried out at the same time. This is an important task of environmentally friendly and economical alternative materials used for so-called green wastewater treatment technologies and as a competitor to conventional approaches.

PAC was already proven to effectively remove heavy metals from water as well as organic contaminants such as chlorophenols and benzotriazole (Pap et al. 2017, 2023). This implies that this material has great potential to be used for removing multiple pollutants which can be present in liquid waste from NORM involving industries. For example, in uranium operations in addition to U and decay products release to the environment; metals such as cadmium, copper, bismuth, arsenic, and molybdenum; organic contaminants such as kerosene, amine, and isodecanol may be released in upset conditions (Bird 2012).

Gross alpha/gross beta screening method for monitoring discharge waters and determining decontamination level

Surface monitors and gamma spectrometry are commonly used for the rapid assessment of contamination levels. However, these methods are not always applicable to liquid samples. Gamma spectrometry is a valuable radiometric technique for identifying specific radionuclides, but the quick detection of naturally occurring radionuclides can be challenging due to their complex behaviour and low activity concentrations. Preconcentration is often required, and certain time (in days) should elapse after sample preparation before counting, due to progenies ingrowth. In addition, pure alpha or beta emitters, if present, are not detected. A useful technique that can be used for fast screening of gross alpha and beta emitters is LSC (Ajemigbitse et al. 2020). This method is mostly used for screening radioactivity in drinking water. Low detection limits can be achieved to meet the requirements set by the Directive, which are 0.04 Bq/L for gross alpha activity and 0.4 Bq/L for gross beta activity (Council of the European Union 2013). With reasonable counting times and the use of small sample quantities (mL), this method is suitable for determining whether certain process waters contain radionuclides below clearance levels and can be treated as normal waste and discharged into the environment. Even if this method cannot determine the specific activity concentrations of certain radionuclides, it can be used for rapid monitoring and decision-making. The disadvantage of the method is the poor resolution and overlap of the spectra compared to alpha spectrometry. For screening purposes, however, the simplicity of the procedure and the rapid response as well as the costs speak in favour of this method.

The gross alpha/gross beta method obtained by LSC, except for determining overall activity, can be used to identify dominant radionuclides by analysing the spectra. Figure 2 presents comparison of the spectra of analysed water sample with standard solutions of U-238, Ra-226 and Th-232 (along with their progenies). By the position and shape of the obtained peaks in the sample, the best match has been found with U standard solution, which is in accordance with gamma spectrometry measurements. By this way, it can be confirmed that U-234 and U-238 mostly contribute to the overall alpha activity in the sample, while their progenies, Th-234 and Pa-234, are the most abounded in the beta spectrum. However, due to low resolution, presence of other natural radioisotopes cannot be excluded and if present, their activity concentrations are much lower than uranium.

This procedure can be effective in any industry where radionuclide decontamination needs to be determined. When an initial characterisation is performed using more specific analyses, the spectra of gross alpha/gross beta activity can be monitored. The peaks in different channels correlate with the energies of the emitted isotopes. By comparing the spectra before and after treatment and analysing the counts in the alpha and beta regions, the degree of decontamination can be quickly assessed.

The advantage of this method lies in its near 100% detection performance for the alpha and beta emitters of interest. In the present study, the background counts in the alpha channels were 0.2 ± 0.05 cpm, while in the beta channels they were 3.5 ± 0.2 cpm. Therefore, with just 8 mL of the sample and 240 min of counting time, the minimum detectable activity concentration can be less than 0.16 Bq/L for beta emitters and 0.04 Bq/L for alpha emitters. This meets the strictest criteria for detection limits set by the Directive 2013/51/Euratom for drinking water.

Desorption of uranium from PAC

The results have shown that PAC efficiently removed radionuclides, thus helped in concentrating radionuclides. The

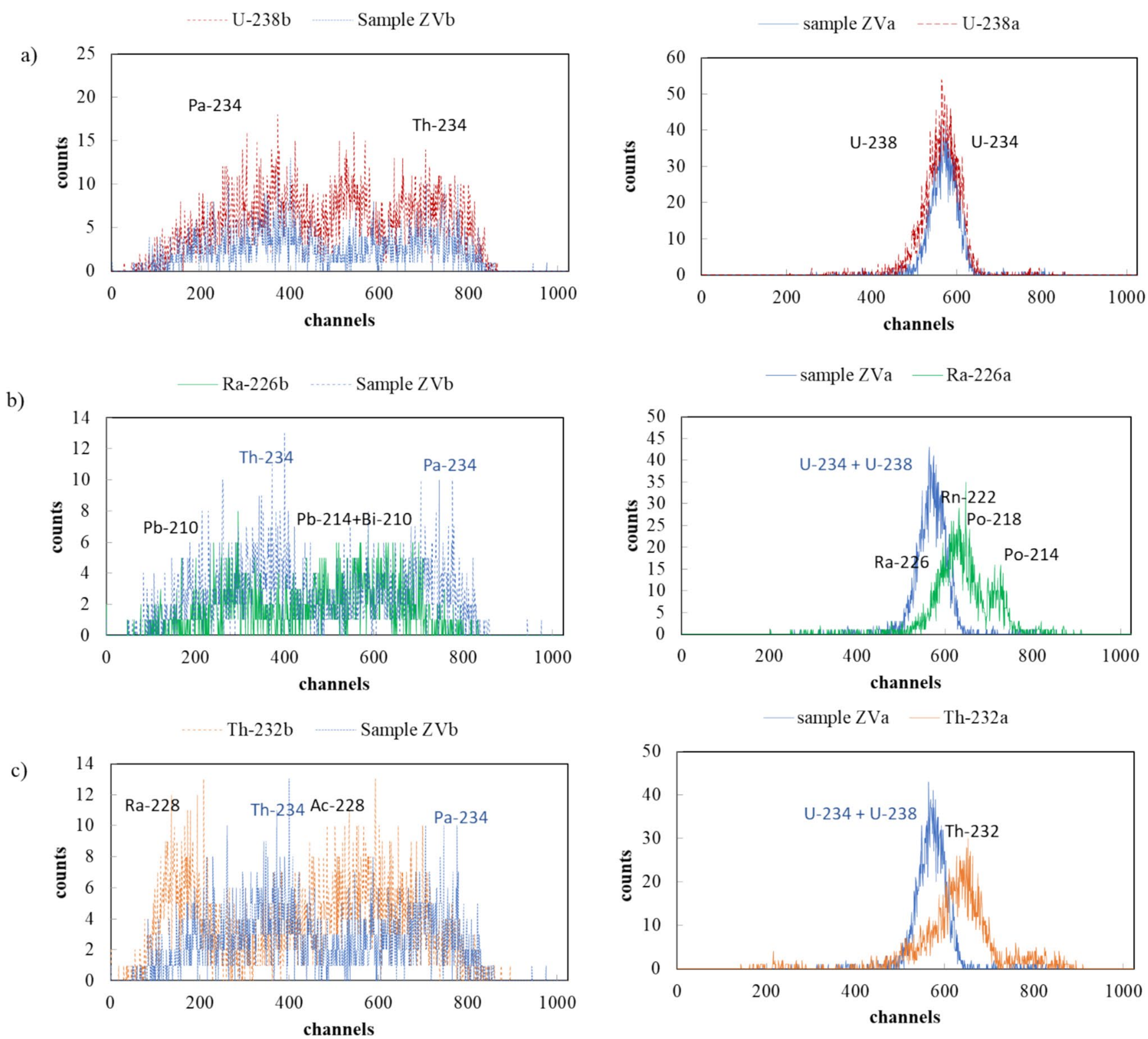

Fig. 2 Comparison of beta and alpha spectrum of water sample from Žirovski Vrh with standard solutions of a) U-238, b) Ra-226 and c) Th-232

Table 3 Efficiency of removing adsorbed alpha emitters (uranium) from PAC by different diluted acids

0.1 M Acid	% of U desorption in 5 ml after 15 min	% of U desorption in additional 8 mL after 24 h	Cumulative desorption
HNO_3	28.8 ± 0.03	30.0 ± 0.05	58.8 ± 0.05
HCl	34.7 ± 0.05	28.1 ± 0.04	62.8 ± 0.07
H_2SO_4	45.0 ± 0.05	50.9 ± 0.06	95.9 ± 0.08
H_3PO_4	47.8 ± 0.03	44.0 ± 0.07	91.8 ± 0.07

AC were proved to be recycled and reused for heavy metal adsorption by eluting cations with diluted HNO_3 or H_3PO_4. Similar approach was tested to determine recovery and recycling of alpha emitters (uranium isotopes). Since, in different NORM industries different acids are used, desorption from AC produced from plum, was tested with diluted HNO_3, HCl, H_2SO_4 and H_3PO_4. Short lived Th-234 and Pa-234 were not in the focus of this experiment.

The results in Table 3 suggest that uranium can be efficiently removed and recovered from wastewater using diluted phosphoric or sulfuric acid. However, further research may enhance uranium recovery with diluted nitric or chloric acid, where recoveries exceeded 50%. It is crucial to highlight that from 50 mL of contaminated water, radionuclides were removed using only 0.1 g of activated carbon produced from plum and recovered with less than 15 mL of diluted inorganic acid. With additional optimisation, this activated carbon has significant potential not only for decontamination but also as an alternative and cost-effective material for the recovery of critical minerals.

Conclusion

It was demonstrated that functionalised activated carbons produced from apricot, plum and cherry/sour cherry kernels by thermochemical activation with H_3PO_4 on 500 °C can be used for radionuclide removal from contaminated water. The tested materials have lower ash and moisture content in comparison with commercial activated carbon. They exhibit highly porous and heterogeneous structure and high BET surface which make them suitable material for adsorption of radionuclides. Gross alpha/beta measurements showed that all tested ACs were effective in removing radionuclides from contaminate water decreasing in order: PAC > CAC > CCAC > AAC. The efficiency of PAC has shown the best adsorption properties with removal efficiency of 95% for alpha and 96% for beta emitters, respectively. High adsorption efficiency was found in broad pH range, from pH 4 to pH 9. The results show that the material may be used for recovery of critical minerals by desorption with diluted inorganic acids, especially, phosphoric, or sulfuric acid. This material may be effective for the simultaneous removal of multiple stressors such as heavy metals, radionuclides, and organic compounds. The gross alpha and gross beta determination by LSC can be used as a fast, rapid, and relatively inexpensive method for screening of radionuclide contamination of (waste) water and efficiency of radionuclide removal from the sample.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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