

INVESTIGATION OF RADIONUCLIDE SORPTION ON PEAT

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Organic sorbents like peat can be successfully used for the purification of radioactive waste waters and are favoured by economical and technical considerations. The peat filters are also friendlier to the environment, since the manufacturing of such filters is a low emission process. This study examined the sorption of radionuclides ¹³⁷Cs, ⁵⁵Fe, ⁶³Ni and ¹⁴C on peat samples. The initial radioactivity of the peat samples was studied before the experiments. Peat samples were taken from different bogs. Radionuclide solutions were prepared using deionised water and appropriate radionuclide composition from the Salaspils research reactor stock water. It was found that in all cases of peat samples, the main natural radionuclides were ²¹⁰Pb, ⁴⁰K and ²¹⁴Bi. The artificial radionuclides ¹⁵²Eu, ¹³⁷Cs and ⁶⁰Co were also found in the peat samples. Radionuclide sorption studies indicated that the initial radioactivity of the solution decreased in the case of radionuclides ⁶³Ni, ¹⁴C, ⁵⁵Fe and ¹³⁷Cs. This confirmed that peat samples can effectively remove the radionuclides from radioactive water solutions.

Key words: *radionuclides, peat, sorption, radioactive waste.*

INTRODUCTION

The sorption of radionuclides on peat materials was investigated by [1–3]. Author [4] reported that humic substances are formed during the humification process of the peat. Humic substances can be divided into humic acids (HA), fulvic acids (FA) and humin [5]. The authors [6, 7] stressed that humic acids strongly affect a wide range of environmental processes, due to complexation ability, including complexation with radionuclides. HA participate in complex formation with metals ions and have an influence on immobilization of these pollutants by peat. It was shown [6], that the presence of HA influence the sorption of Eu(III) on alumina surface, depending on pH values. Sorption process is one of the methods for studying interaction behaviour between peat and radionuclides. Authors [8–11] investigated the distribution of radionuclides in the environment and the influence of humic substances on migration of radionuclides.

In this paper, the sorption of radionuclides ¹³⁷Cs, ⁵⁵Fe, ⁶³Ni and ¹⁴C on peat samples was studied for radioactive waste management purposes. Organic sorbents like a peat can be successfully used for purification of radioactive waste waters, and are favoured by economical and technical considerations. The

peat filters are also friendlier to the environment since the manufacturing of such filters is a low emission process.

EXPERIMENTAL

Peat samples were acquired from Dzelves, Svetupes and Viku bogs, using peat from different layers. The samples were oven dried during 24 h at 105 °C and were ground and sieved to obtain peat fractions with size: <0.25 mm, 0.25–0.5 mm, 0.5–2.0 mm and >2.0 mm. Analytical grade chemicals were used. Radionuclide solutions were prepared using deionised water and the appropriate radionuclide composition from Salaspils research reactor liquid radioactive waste water. Solution pH and conductivity were measured using a universal Mettler Toledo pH meter/ conductivity meter.

Batch sorption studies were performed in isothermal conditions at different temperatures using mechanical mixer and a 350 mL glass flask. A 5 g peat sample was inserted into 250 mL of radioactive solution and mixed for the entire duration of experiment. A 2 mL aliquot of the solution was taken after a defined time interval and was centrifuged for 10 min at 10000 rotations per minute.

Flow experiments were performed by using a sorption column loaded with 50 g of peat. The radioactive solution was pumped through the column at the flow rate of 0.4 L/h. An Ortec gamma spectrometer with high purity germanium probe was used for the measurement of γ -radionuclide concentration in solutions and peat samples. Radioactivity of β - and α -radionuclides was measured using a Canberra–Packard TriCarb 2700/SL liquid scintillation counter.

RESULTS AND DISCUSSIONS

Experimental studies indicated that the natural peat samples already contained gamma radionuclides which must be taken into account for correct explanation of the results (Table 1).

Table 1. Content of radionuclides in different layers of bogs

No.	Sampling depth, cm	²¹⁰ Pb	¹⁵² Eu	¹³⁷ Cs	⁴⁰ K	⁶⁰ Co	²¹⁴ Bi
Dzelves bog							
Milled peat, mBq/g							
1	50	126.0	4.0	8.0	76.0	<MDL	48.0
Near surface peat, mBq/g							
2	20	155.0	4.0	53.0	49.0	0.06	32.0
Svetupes bog, mBq/g							
3	0–100	35.0	0.5	448.0	39.0	0.06	15.0
4	140–150	15.0	0.9	28.0	15.0	<MDL	3.5
Viku bog, mBq/g							
5	0–100	27.0	0.5	101.0	146.0	<MDL	7.5
6	100–110	19.0	6.5	29.0	17.0	0.1	3.5
7	180–200	87.0	1.5	628.0	158.0	0.6	6.0

It was found that for all cases of peat samples, the main natural radionuclides were ²¹⁰Pb, ⁴⁰K and ²¹⁴Bi. The artificial radionuclides ¹⁵²Eu, ¹³⁷Cs and ⁶⁰Co were

also found in the peat samples from all the investigated bogs. Experimental results indicated that near surface peat samples has an increased radioactivity of ^{137}Cs radionuclide, but in the case of Viku bog, the radioactivity of the layer at 1.8 m–2.0 m depth had the highest value.

Batch sorption mode studies indicated that during the first 5 hours of the experiments, the initial radioactivity of the solution decreased in the case of ^{63}Ni by a factor 5 (Fig. 1). It proved that peat samples can effectively remove this radionuclide from radioactive water solutions, and the sorption ability of the Dzelves bog peat samples did not depend on the fraction size in the interval up to 2.0 mm.

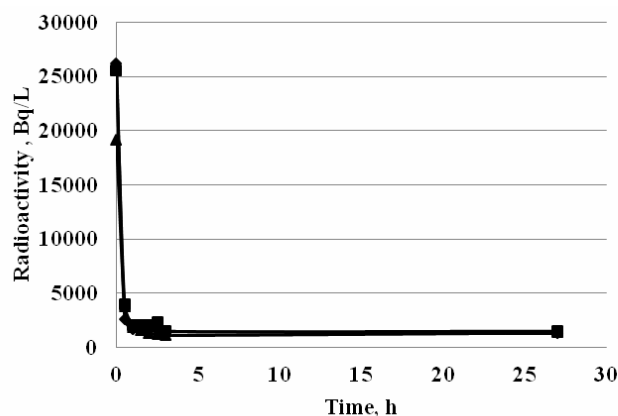


Fig. 1. The ^{63}Ni radioactivity sorption curves for the batch sorption studies: 1 – fraction <0.25 mm; 2 – fraction 0.25–0.5 mm; 3 – fraction 0.5–2.0 mm. Near surface peat samples from Dzelves bog. Deionised water temperature was 20 ± 0.1 °C.

Similar results [3] were observed in the case of the batch sorption studies for the radionuclides ^{14}C and ^{55}Fe . The initial radioactivity of the solution decreased in the case of ^{14}C by a factor 6 and in case ^{55}Fe – by a factor 10. Experimental results confirmed that the pH of the water phase had a significant influence on the radionuclide sorption on the peat samples. The results obtained (Fig. 2) using ^{63}Ni radionuclide showed comparable results for the samples at temperatures 5 °C and 30 °C with the same pH value of the solutions. Increasing the pH value up to 7.1 resulted in increased sorption ability of the peat sample.

The same results were observed for the flow experiments using the radionuclide ^{137}Cs . It was shown that ^{137}Cs sorption ability of the peat samples decreased significantly during the first 3 h (Fig. 3). Experimental data indicated that the pH of the solution also decreased from 9.0 to 3.8 in the same time period. The pH changes in the water solution occurred due to the acidic properties of the peat samples. It was shown (Fig. 3) that the sorption efficiency stabilized around pH = 4.0, but the rise of pH at the next stages of experiments caused a decrease in the sorption efficiency. The observed experimental data confirmed the high sorption efficiency of the radionuclides by peat from the Dzelves bog. The results of both experiments types were in a good agreement with the results of previous investigations [1–3, 6, 7]. The obtained results confirmed the role of humic substances in the immobilization of radionuclides.

It was an important consideration for developing new purification systems for radioactive waste management purposes.

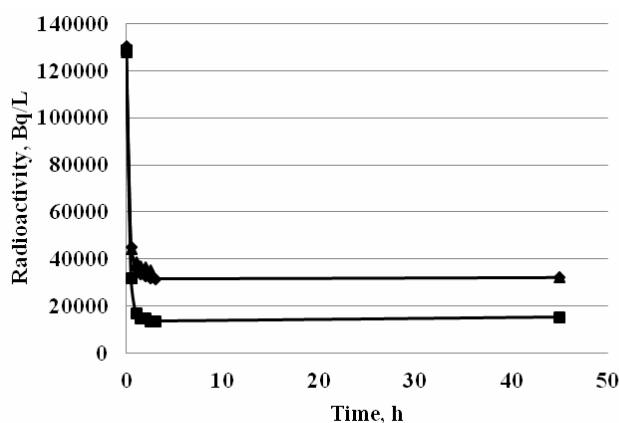


Fig. 2. Radionuclide ^{63}Ni sorption curves for the batch sorption studies. Fraction 0.25–0.5 mm: 1 – temperature 30°C , $\text{pH}=3.8$; 2 – temperature 30°C , $\text{pH}=7.1$; 3 – temperature 5°C , $\text{pH}=3.8$. Dzelves bog near surface peat samples.

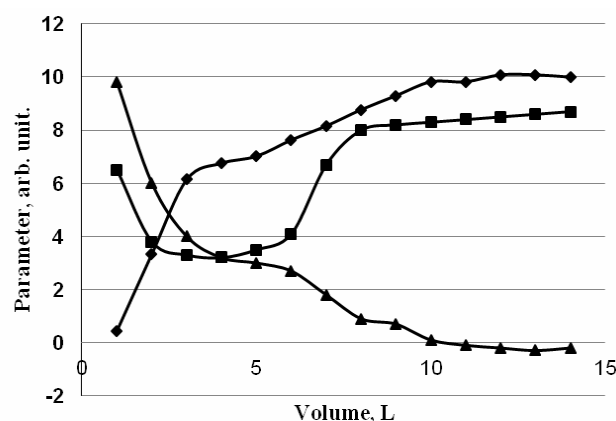


Fig. 3. Sorption parameters changes during the flow experiments with the radionuclide ^{137}Cs : 1 – sorption curve for ^{137}Cs ; 2 – pH changes during experiment; 3 – sorption efficiency $\times 10$, %. Dzelves bog near surface peat samples, fraction 0.25–0.5 mm. Temperature $20 \pm 1^\circ\text{C}$.

CONCLUSIONS

1. The radioactivity of peat from different bogs was studied. The artificial radionuclides ^{152}Eu , ^{137}Cs and ^{60}Co were found in the peat samples.

2. It was shown that the initial radioactivity of solution decreased during the batch and flow sorption experiments in the case of ^{63}Ni , ^{14}C , ^{55}Fe and ^{137}Cs radionuclides up to 10 times. This confirmed the high sorption ability of the investigated peat samples.

3. Radionuclide sorption ability by the peat samples during batch and flow sorption experiments significantly depended on the solution pH . Experimental studies indicated a complex effect of solution pH values on radionuclide immobilization processes.

4. Further research is warranted to establish the dependence of radionuclide sorption on external factors.

Acknowledgments

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KŪDRAS RADIONUKLĪDU SORBCIJAS PĒTĪJUMI

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K O P S A V I L K U M S

Organiskie kūdras un tās atvasinājumu sorbenti var tikt lietoti šķidro radioaktīvo atkritumu attīrīšanai gan ekonomisko, gan tehnisko apsvērumu dēļ. Kūdras filtri ir daudz draudzīgāki apkārtējai videi, to ražošanā izmanto process, kuros ir maza siltumnīcgāzu izmete. Darba mērķis bija izpētīt radionuklīdu ^{137}Cs , ^{55}Fe , ^{63}Ni un ^{14}C sorbciju kūdras paraugos.

Pirms eksperimentu veikšanas tika noteikta dažādu purvu kūdras paraugu sākotnējā radioaktivitāte. Radioaktīvie šķīdumi tika sagatavoti, izmantojot dejonizēto ūdeni un atbilstošo radionuklīdu kompozīciju no Salaspils kodolreaktora šķidro radioaktīvo atkritumu tvertnēm.

Visu purvu kūdras paraugos atrasti dabīgie radionuklīdi ^{210}Pb , ^{40}K un ^{214}Bi , kā arī mākslīgie radionuklīdi ^{152}Eu , ^{137}Cs un ^{60}Co . Radionuklīdu sorbcijas pētījumi stacionārajā un plūsmas režīmā parādīja, ka tiek novērota radionuklīdu ^{63}Ni , ^{14}C , ^{55}Fe un ^{137}Cs sākotnējās radioaktivitātes būtiska samazināšanās ūdens fāzē. Tas apstiprina pieņēmumu, ka kūdra var tikt efektīvi izmantota šķidro radioaktīvo atkritumu attīrīšanai no radionuklīdiem.