

# INVESTIGATION OF URANIUM, CAESIUM AND STRONTIUM BY SOIL FROM WATER FOR REGION OF REPOSITORY FOR RADIOACTIVE WASTE

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*Sorption of U(VI), Sr(II) and Cs(I) with soil from the region of location of the radioactive waste repository was studied. The radionuclides  $^{237}\text{U}$ ,  $^{132}\text{Cs}$ ,  $^{91}\text{Sr}$  produced by the photonuclear reactions  $^{238}\text{U}(\gamma, n)$  and  $^{238}\text{U}(\gamma, f)$  on an electron accelerator, MT-25 microtron (FLNR, JINR), was used as tracers.*

Possible sources of uranium, caesium and strontium radionuclide contamination are enterprises for nuclear fuel reprocessing, enterprises for the recovery and processing of uranium and thorium, global fallout due to nuclear explosions and radioactive waste repositories. Experiments on determination of the distribution coefficients ( $K_d$ ) of radionuclides between water and soil, allowing evaluation of the radionuclide mobility in soil, are performed in territories that may be contaminated with radionuclides.

The distribution of nuclides between natural solutions and soil depends on many factors, including the soil composition [1 - 3].

In this study we examined the distribution of uranium, caesium and strontium between aqueous solutions and soil from the region of location of the Novi Khan radioactive waste repository in Bulgaria.

## Experimental

Sorption of U (VI), Cs (I) and Sr (II) were studied on samples of brown forest soil, taken from the region of location of the radioactive waste repository. The mean chemical composition (%) of this type of soils after A.P. Vinogradov [4] is given in Table 1.

**Table 1.**

Calcination loss (organic matter + CO <sub>2</sub> )	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	SO <sub>3</sub>
5.57	76.62	10.67	3.35	0.92	2.02	0.59	0.75	0.15

Soil samples were sieved through a 200-mesh sieve (particle size 0.074 mm) and dried at 100–105°C to constant weight.

In studying the sorption of uranium, caesium and strontium with the soil from aqueous solutions, we used the tracers  $^{237}\text{U}$  [ $T_{1/2} = 6.75$  d,  $E_\gamma = 59.54$  keV (34.5%), 208.00 keV (21.2%)],  $^{132}\text{Cs}$  [ $T_{1/2} = 6.479$  d,  $E_\gamma = 505.79$  keV (0.73 %); 630.19 keV (0.97 %); 667.718 keV (100 %)];  $^{91}\text{Sr}$  [ $T_{1/2} = 9.63$  h,  $E_\gamma = 652.9$  keV (8.0 %); 749.8 keV (23.61 %); 1024.3 keV (33 %)], which were produced by the reactions  $^{238}\text{U}(\gamma, n)$  and  $^{238}\text{U}(\gamma, f)$  on an electron accelerator, MT-25 microtron [5-7].

Sorption of U(VI), Cs(I) and Sr(II) on soil was performed in 50-ml plastic test tubes. A 0.2-g portion of soil was placed in a test tube, and 5 ml of a nitric acid solution with the pH measured with an accuracy of  $\pm 0.1$  using a pH-meter and 100  $\mu\text{l}$  of a solution containing U-237, Cs-132, Sr-91 were added. After that, the samples were kept for 24 h at room temperature ( $T = 20 \pm 0.5^\circ\text{C}$ ).

The activity in a solution aliquot (1 ml) before and after the sorption was measured with a multichannel amplitude pulse analyzer equipped with a HPGe detector with a resolution of 1.5 keV on the  $^{60}\text{Co}$  line at 1.33 MeV.

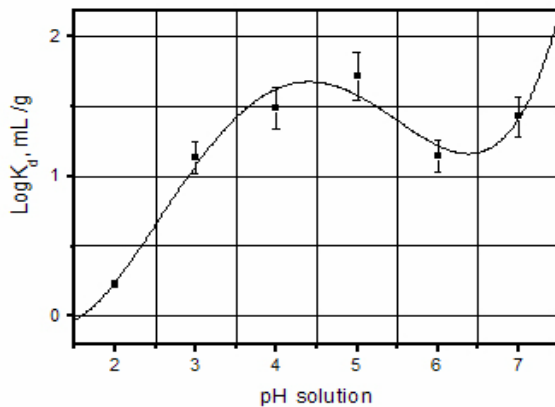
The distribution coefficient between the soil and solution was calculated from the activity of  $^{237}\text{U}$ ,  $^{132}\text{Cs}$  and  $^{91}\text{Sr}$  in the solution before and after the sorption, using the formula

$$K_d = (A_0 - A_i)V/(A_i m)$$

where  $K_d$  is the distribution coefficient,  $\text{ml g}^{-1}$ ;  $A_0$ , activity of the nuclide in solution before the sorption, Bq;  $A_i$ , activity of the nuclide in the solution after sorption, Bq;  $V$ , solution volume, ml; and  $m$ , weighed portion of the soil, g.

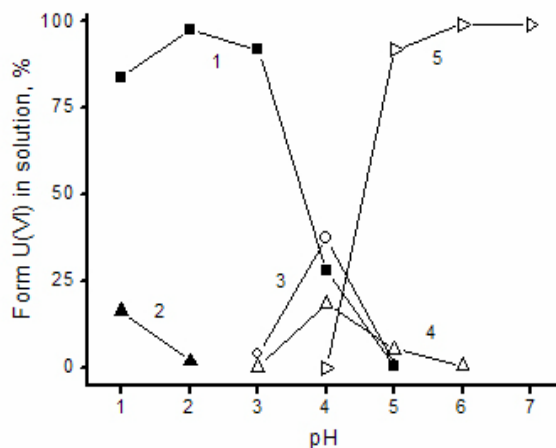
## Result and discussion

The uranium distribution coefficients ( $K_d$ , mean deviation  $\pm 2\%$ ) in the soil–solution system are shown in Fig. 1.



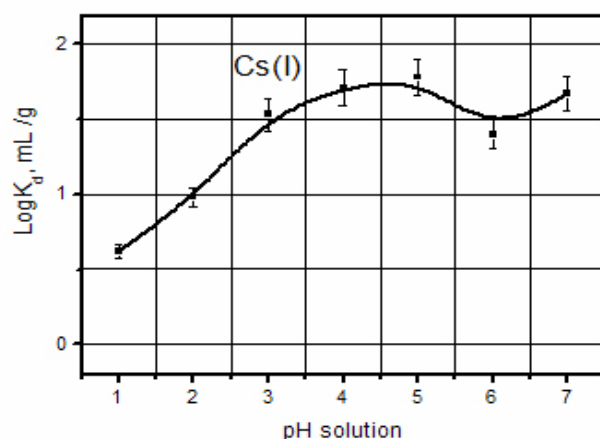
**Fig. 1.** Sorption of U(VI) on soil as a function of pH of solution at uranium concentration  $C_U = 1 \cdot 10^{-2} \text{M}$

In the examined brown forest soil, the uranium content was  $(2.6 \pm 0.1) \cdot 10^{-6} \text{g g}^{-1}$  [8]. The U(VI) speciation in solution is determined using the Speciation program [9] (Fig 2).

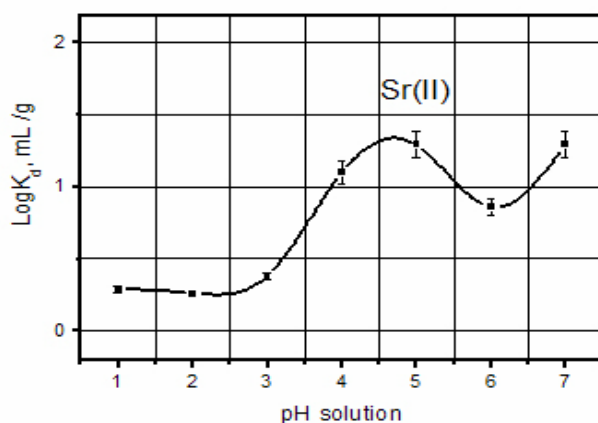


**Fig. 2.** Major U(VI) species in nitric acid solution.  $C_U = 1 \cdot 10^{-2} \text{M}$ :  
 (1)  $\text{UO}_2^{2+}$ , (2)  $\text{UO}_2\text{NO}_3^+$ , (3)  $(\text{UO}_2)_2(\text{OH})_2^{2+}$ , (4)  $(\text{UO}_2)_3(\text{OH})_5^+$ , (5)  $\text{UO}_2(\text{OH})_2(\text{solid})$

The caesium and strontium distribution coefficients ( $K_d$ , mean deviation  $\pm 2\%$ ) in the soil–solution system are shown in Figs. 3 and 4.



**Fig. 3.** Sorption of Cs(I) on soil as a function of pH of solution at caesium concentration  $C_{Cs} = 1 \cdot 10^{-16} - 1 \cdot 10^{-15}$  M



**Fig. 4.** Sorption of Sr(II) on soil as a function of pH of solution at strontium concentration  $C_{Sr} = 1 \cdot 10^{-16} - 1 \cdot 10^{-15}$  M

An increase  $K_d$  values of U(VI), Cs(I) и Sr(II) between investigated soil and solution are observed in the range pH 3 – 5. This showed that the studied soil samples from pH 3 to pH 5, can be characterized with cation exchange properties.

Thus, we have determined the uranium, caesium and strontium distribution coefficients  $K_d$  in the soil–solution system in the region of location of the radioactive waste repository. The sorption of various uranium species by the soil has been examined. The sorption characteristics that we determined for U(VI), Cs(I) and Sr(II) in the soil–solution system allow prediction of the migration behavior of uranium, caesium and strontium in the brown forest soil under the conditions of accidents.

## References

1. *Thibault D. H., Sheppard M. I., Smith P. A.* A Critical Compilation and Review of Default Soil Solid/Liquid Partition Coefficients,  $K_d$ , for Use in Environmental Assessments. AECL-10125. Whiteshell Nuclear Research Establishment. Atomic Energy of Canada Limited. Pinawa, Manitoba Canada. 1990.
2. Understanding Variation in Partition Coefficients,  $K_d$  values. Vol. 2., EPA 402-R-004B, U. S. Environmental Protection Agency, 1999.
3. *Takamiya K., Fukunishi T., Tsujito R.*, Investigation of adsorption behaviour of fission products in soil using multitracer technique, Czech. Journal of Physics, Vol. 50, 2006, P. D399 - D404.
4. *Vinogradov, A.P.*, *Geokhimiya redkikh i rasseyannykh khimicheskikh elementov v pochvakh* (Geochemistry of Rare and Trace Chemical Elements in Soils), Moscow: Akad. Nauk SSSR, 1950, p. 279.
5. *Maslov O. D., Bozhikov G. A., Ivanov P.I.* et al. JINR. P6-2009-30. Dubna, 2009.
6. *Sabelnikov A. V., Maslov O. D., Gustova M. V. et al.* JINR. P12-2004-193. Dubna, 2004.
7. *Gustova, M.V., Dmitriev, S.N., Maslov, O.D.*, et al., RF Patent 2 262 759, 2003, Byull. Izobret., 2005, no. 29.
8. *Filippov M. F., Maslov O. D., Bozhkov et al* // Radiokhimiya, 2009, Vol 51, № 1, P. 67-69
9. Academic Software: Sol-Eq, Aqueous Solutions <http://acadsoft.co.uk/>