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Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste

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- Radionuclide Diffusion in Water Saturated Graphite-
(FZJ-Contribution)

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Document title

Water saturation of the graphite pore system

Executive summary

The mobilisation of long-lived radionuclides like ^{14}C and ^{36}Cl is a key question for the long term safety of disposed irradiated graphite. The mobility of radionuclides under repository conditions prior to water access is an important concern. Diffusion of radionuclides in the graphite matrix after access of water to the pore space in graphite is studied in this report.

This report is focused on graphite from the A3-3 fuel matrix of HTR as well as on irradiated pieces from the AVR reflector structures.

The AVR reflector graphite is representative for other grades, which have been manufactured in the 1960th.

Revisions

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List of Content

1 Introduction 5
2 Diffusion of ^{241}Am, ^{230}Th, ^{234}U and ^{236}U radioactive tracers through HTR graphite 5
2.1 Experimental 5
2.2 Results and discussion 8
2.2.1 Nonradioactive samples from the HTR graphite fuel prototype 8
2.2.2 Radioactive samples from the HTR graphite fuel element (AVR-2) and from the AVR wall graphite block (AVR-1) 13
3 Conclusion 18
4 References 20
5 Appendix 21

1 Introduction

The AVR (Arbeitsgemeinschaft Versuchs-Reaktor) was the first High-Temperature Reactor (HTR) being built in Germany (Start of construction 1960). It operated between 1967 and 1988 and had a so-called pebble bed core. It had a power of 46 MWth and an electric power of 15 MWel. The coolant was helium and the operating temperatures were designed to be in the range of 850-950°C. The reactor served as a testbed for spherical fuel elements and for coated-particles, which were invented after fixing the design of the reactor. Fission product releases from the first fuel types was quite high. Thus, the contamination of the graphite structures is quite high.

For reasons of completeness, investigations made in the European RAPHAEL project on the A3 matrix of the fuel pebbles are also integrated into this report, whereas analyses on the AVR graphite structures are part of the CARBOWASTE project. However, only small broken pieces from the AVR reflector could be retrieved from the core, which made the related investigations rather difficult.

2 Diffusion of ^{241}Am, ^{230}Th, ^{234}U and ^{236}U radioactive tracers through HTR graphite

2.1 Experimental

The samples for the diffusion experiments were cut from the nonradioactive HTR graphite fuel prototype, as shown in Fig.1 a. In all cases, the samples diameter was 35 mm and thickness was about 10 mm. These samples are denoted as D1, D2 and D3. The density of the graphite fuel prototype was measured by the geometrical method, the value of density averaged by four spheres equals (1.76 ± 0.02) g/cm^3.

Moreover, two samples for the diffusion experiments were made from the radioactive graphite. First sample (denoted as AVR-2) was cut from the HTR graphite fuel element, as shown in Fig.2 a. In this case, the samples diameter was 11.4 mm and thickness was 4.7 mm. The density of the graphite disk was measured by the geometrical method; the value of density equals 1.80 g/cm^3. The initial sample contained the following radionuclides: ^{154}Eu – 397 Bq/g; ^{60}Co – 3526 Bq/g; ^{137}Cs - 23239 Bq/g.

Second sample (denoted as AVR-1) was cut from the AVR wall graphite block. The samples diameter and thickness were 11.2 mm and 5.6 mm, respectively. The density of the graphite disk was measured by the geometrical method; the value of density equals 1.72 g/cm^3. The initial sample contained the following radionuclides: ^{154}Eu – 391 Bq/g; ^{60}Co – 11157 Bq/g; ^{137}Cs - 16735 Bq/g.

Both radioactive samples were carefully washed in 20 ml distilled water during 30 minutes in ultrasonic bath. This operation was repeated eight times for the AVR-1 sample and ten times for the AVR-2 sample.

For preparing the rim, supporting the sample in diffusion cell, an epoxy resin of the type Araldit F and hardener HY956 (18 % weight of resin) were used. The sample was put into cylindrical Teflon mould and the resin was poured over it. After the total hardening (~ 10 day), the samples were taken out and their surfaces were carefully polished. Photos of the prepared samples are shown in Fig. 1b and Fig. 2b.

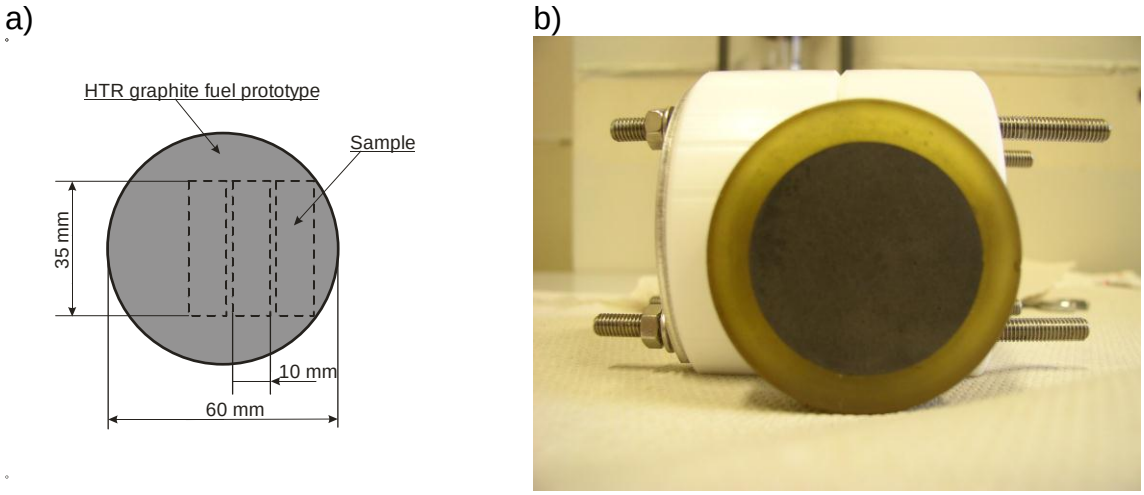


Figure 1 : Scheme of the making of D1, D2, D3 samples (a) and photo of sample (b)

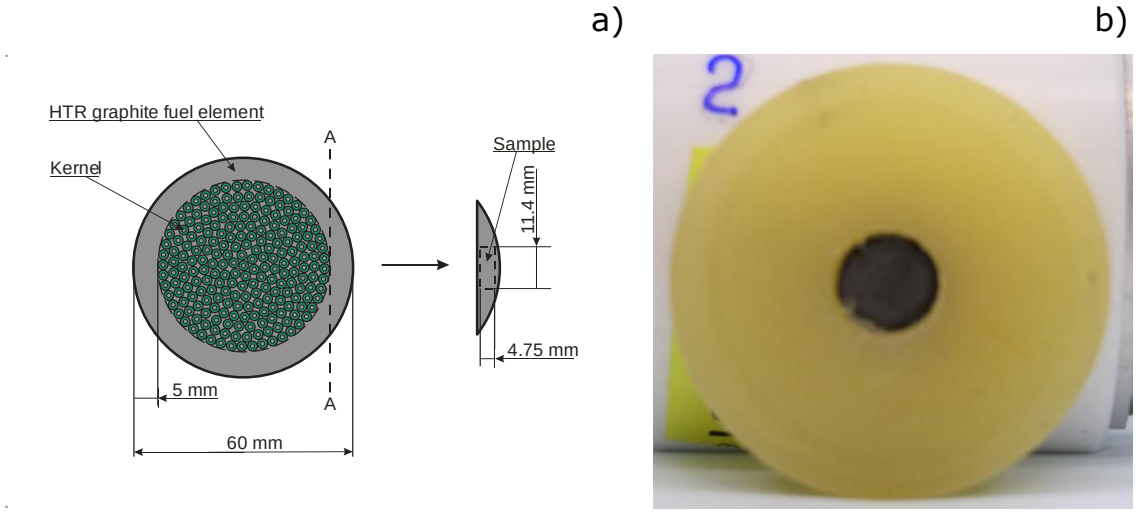


Figure 2. Scheme of the making of AVR-2 samples (a) and photo of sample (b)

The apparatus used for the diffusion experiments is shown in Figure 3

The water-saturated samples were mounted in between two 20 ml Teflon containers with rubber O-rings as sealing, Fig. 3a. Left and right containers were initially filled with distilled water. After one week, the distilled water was removed and the left container was filled with the radioactive tracers Brine-2 base solution, right container – pure Brine-2 solution.

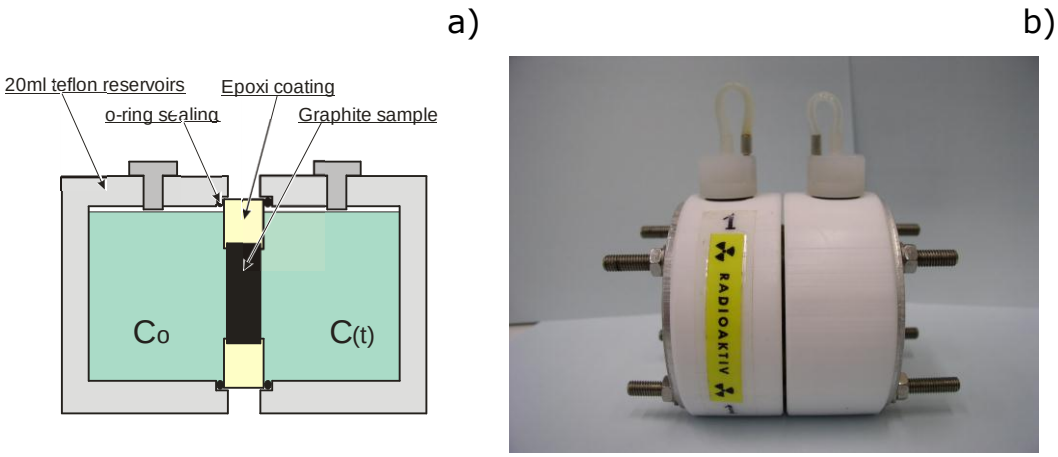


Figure 4: The apparatus for the diffusion experiments

Three diffusion cells were made for the through-diffusion experiment with the nonradioactive samples, denoted also as D1, D2 and D3. The values of the initial concentrations (*Co*) of radioactive tracers in Brain-2 solution were as following: ^{241}Am – 3087.8 Bq/ml; ^{230}Th – 728.3 Bq/ml; ^{234}U – 54.5 Bq/ml; ^{236}U – 714.4 Bq/ml.

The initial concentrations of radioactive tracers in Brine-2 solution in the left container was equal to *Co* for the diffusion cells D1 and D2 and *Co*/5 for the diffusion cell D3. The precise information about the initial concentrations of radioactive tracers is shown in Appendix 1 and 2.

Two diffusion cells were made for the through-diffusion experiment with the radioactive samples, denoted as AVR-1 and AVR-2. The values of the initial concentrations (*Co*) of radioactive tracers in Brain-2 solution were as following: ^{241}Am – 2970 Bq/ml; ^{230}Th – 322 Bq/ml; ^{236}U – 588 Bq/ml.

For all samples the diffusion experiments were carried out during three months. One time in week, 1 ml samples were taken from the right side of the cells for measuring of the current concentrations (*C*) of radioactive tracers. For the measuring of the initial concentrations (*Co*), 0.5 ml samples were taken from the left side of the cells at the start and the end of experiments only. All samples taken were weighed.

For the measurement-geometry correction, the volume of all samples was increased by addition of distilled water up to 10 ml. The concentrations of radioactive tracers in solutions were determined by gamma and alpha spectrometry methods (HPG Detector from EG&G Ortec and Canberra-Packard Alpha analyser, respectively).

The effective diffusion coefficient (*De*) was determined by analyzing the break-through curve using a mathematical solution for Fick's second law in assumption of one-dimensional diffusion [8]:

$$\frac{C(t)}{Co} = \frac{S}{V}\left(\frac{Det}{L} - \frac{\alpha L}{6} - \frac{2\alpha L}{\pi^2}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\exp\left(-\frac{Den^2\pi^2 t}{\alpha L^2}\right)\right) \qquad (1)$$

where *S*, *L* are the disk area and thickness, *V* – the container volume, α – capacity factor, *t* - time. At optimization, the fitting parameters were *De* and α.

2.2 Results and discussion

2.2.1 Nonradioactive samples from the HTR graphite fuel prototype

The measured values of the current concentrations (*C*) of radioactive tracers for all diffusion cells are presented in Appendix 1. The after-treatment of the experimental data is presented in Appendix 2, 3. In case of the diffusion cell D3, the measured values of the current concentrations (*C*) of ^{230}Th, ^{234}U, ^{236}U radioactive tracers were lower then detection level. Therefore, the current concentrations of ^{241}Am were considered in this work only.

The experimental data for the relative concentrations of the different radioactive tracer (^{241}Am, ^{230}Th, ^{234}U, ^{236}U) depending on time, and corresponding break-through curves are shown in Fig. 20a, b for the diffusion cells D1 and D2, respectively. The analytical solution of Fick's second equation for the geometry of the through-diffusion cells was fitted to experimental data by using effective diffusion coefficient (*De*) and capacity (α) as a fitting parameters. The values of the effective diffusion coefficient (*De*) and capacity factor (α) are summarized in Table 1.

a) b)

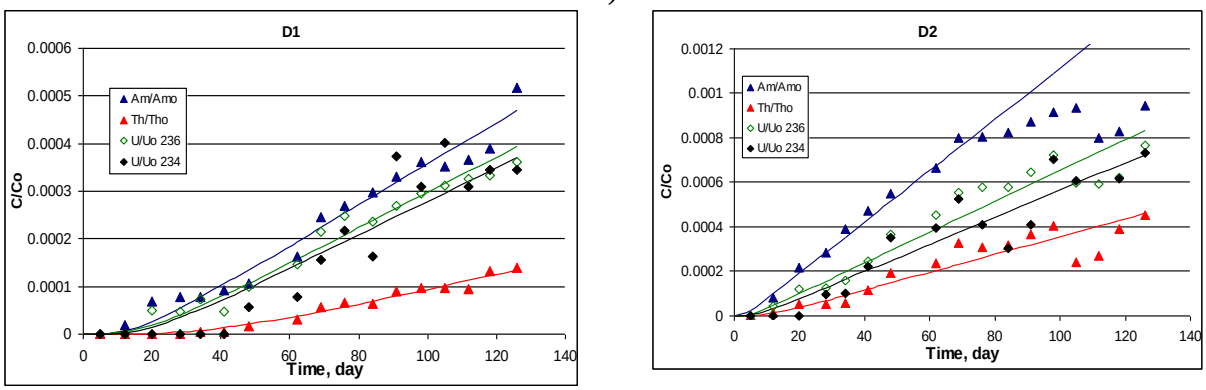


Figure 4: Through-diffusion of ^{241}Am, ^{230}Th, ^{234}U, ^{236}U tracers in diffusion cells D1 (a) and D2 (b).

For both diffusion cells, the effective diffusion coefficient (*De*) decreases in the following way:

^{241}Am > ^{236}U > ^{234}U > ^{230}Th

Sample		^{241}Am	^{236}U	^{234}U	^{230}Th
D1	De*1E-15,m^2/s=	10.71	9.22	8.78	4.10
	a*1E-3 =	0.96	0.97	0.98	0.98
D2	De*1E-15,m^2/s=	28.79	17.33	15.29	10.06
	a*1E-3 =	0.50	0.50	0.60	0.60
D3	De*1E-15,m^2/s=	4.23			
	a*1E-3 =	0.91			

Table 1: effective diffusion coefficient and capacity factor

Moreover, the ratio of De^{Am241}/De^{Th230} equals ~ 2.7 for both cells. For the uranium tracers, the values of *De* are nearly equal and, as usual, $De^{U234} \sim 0.9De^{U236}$. For all radioactive tracers, the diffusion coefficients for the diffusion cell D2 are on average in 2.2 times higher then *De* for the diffusion cell D1, it is shown in Table 2.

	^{241}Am	^{236}U	^{234}U	^{230}Th
De^{D2}/De^{D1}	2.7	1.9	1.7	2.5

Table 2: diffusion coefficients for diffusion cell D2

The capacity factors for the diffusion cell D2 are on average 2 times less than α for the diffusion cell D1 (α ~ 0.00055 and 0.001, respectively). Apparently, these differences may be determined by the non-uniformity of density to diameter of the HTR graphite fuel prototype. The experimental data on the relative concentrations of the different radioactive tracers allow to assume more complex dependence *C/Co = f(t)* than the assumption on one-dimensional diffusion (1), Fig. 3. At the times longer than 60 – 80 days, the considerable deviations from dependence (1) are observed. This effect takes place for both diffusion cells D1 and D2, but in the case of D2 this effect is more significant, especially, for the Am^{241} radioactive tracer, Figure 5a.

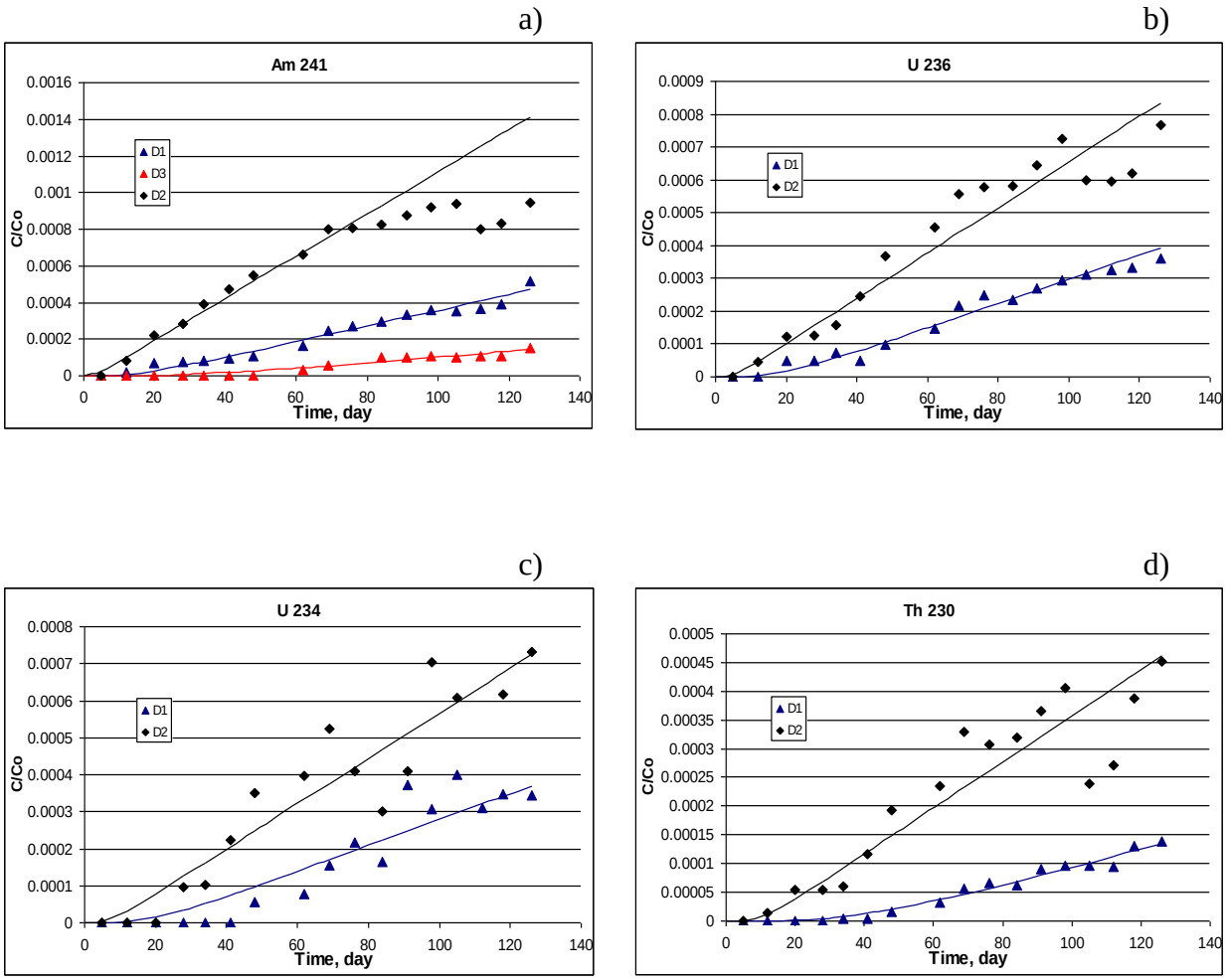


Figure 5: Through-diffusion of ^{241}Am (a), ^{236}U (b), ^{234}U (c) and ^{230}Th (d) tracers in diffusion cells D1, D2 and D3 (measured data (points) and fitted curves (solid lines)).

At the times less than 60 – 80 days, the dependence *C/Co = f(t)* was described by equation (1), and the effective diffusion coefficient (*De1*) was calculated. At the times longer than 60 – 80 days, the linear assumption of the diffusion equation was used, as described below.

The equation of the one-dimensional diffusion (1) can be divided on two parts:

$$\frac{C(t)}{Co} = \frac{S}{V}\left(\frac{Det}{L} - \frac{\alpha L}{6}\right) \qquad (2)$$

$$\frac{C(t)}{Co} = -\frac{2\alpha SL}{\pi^2 V}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^2}\exp\left(-\frac{Den^2\pi^2 t}{\alpha L^2}\right) \qquad (3)$$

As shown in Figure 6, at times less than *t** the behaviour of the diffusion curve is determined by both parts (2) and (3) of the one-dimensional diffusion equation (1). For the times longer than *t**, the linear behaviour of the diffusion curve predominates (equation (2)). In

this case, the effective diffusion coefficient (*De*) and capacity factor (α) can be estimated from the numerical parameters (*A*, *B*) of the linear dependence (*C/Co* = *At* + *B*):

$$De = \frac{AVL}{S} \tag{4}$$

$$\alpha = -\frac{6BV}{LS} \tag{5}$$

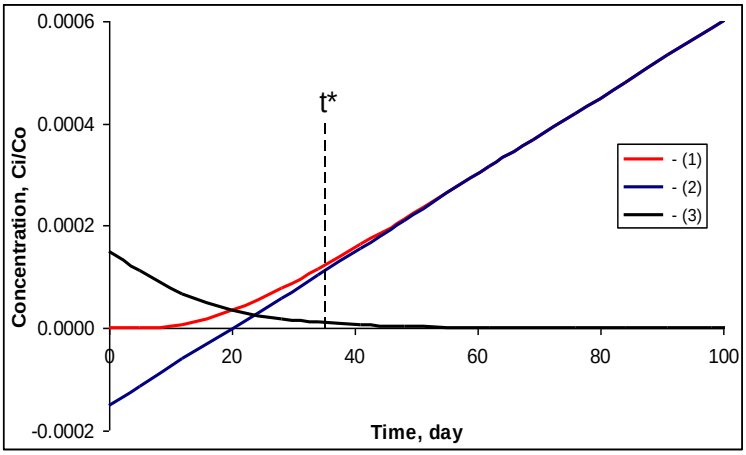


Figure 6: Through-diffusion curve.

Therefore, at the times longer than 60 – 80 days, the dependence *C/Co* = *f(t)* was assumed as linear (dotted line in Figure 22), and the effective diffusion coefficient (*De*) was estimated as $De2 = \frac{AVL}{S}$. The experimental data on the relative concentrations of the different radioactive tracer (^{241}Am, ^{230}Th, ^{234}U, ^{236}U) depending on time, and corresponding curves are shown in Fig. 7. The values of the effective diffusion coefficients (*De1* and *De2* for cell D2) are summarized in Table 3.

As shown in Table 3, the values of the effective diffusion coefficients for the diffusion cell D2 decrease by a factor of 2 to 3 (parameter $De1^{D2}/De2^{D2}$). The value of the effective diffusion coefficient for the diffusion cell D2 at the times later on 60 – 80 days is similar to that for the diffusion cell D1 (parameter $De2^{D2}/De^{D1}$).

Sample		^{241}Am	^{236}U	^{234}U	^{230}Th
D1	De*1E-15,m^2/s=	10.71	9.22	8.78	4.10
D2	De1*1E-15,m^2/s=	28.79	19.20	18.19	11.22
	De2*1E-15,m^2/s=	9.58	7.44	9.29	3.80
	$De1^{D2}/De2^{D1}$	3.01	2.58	1.96	2.96
	$De2^{D2}/De^{D1}$	0.89	0.81	1.06	0.93

Table 3: effective diffusion coefficient for the diffusion cell D2 at the times later on 60 – 80 days

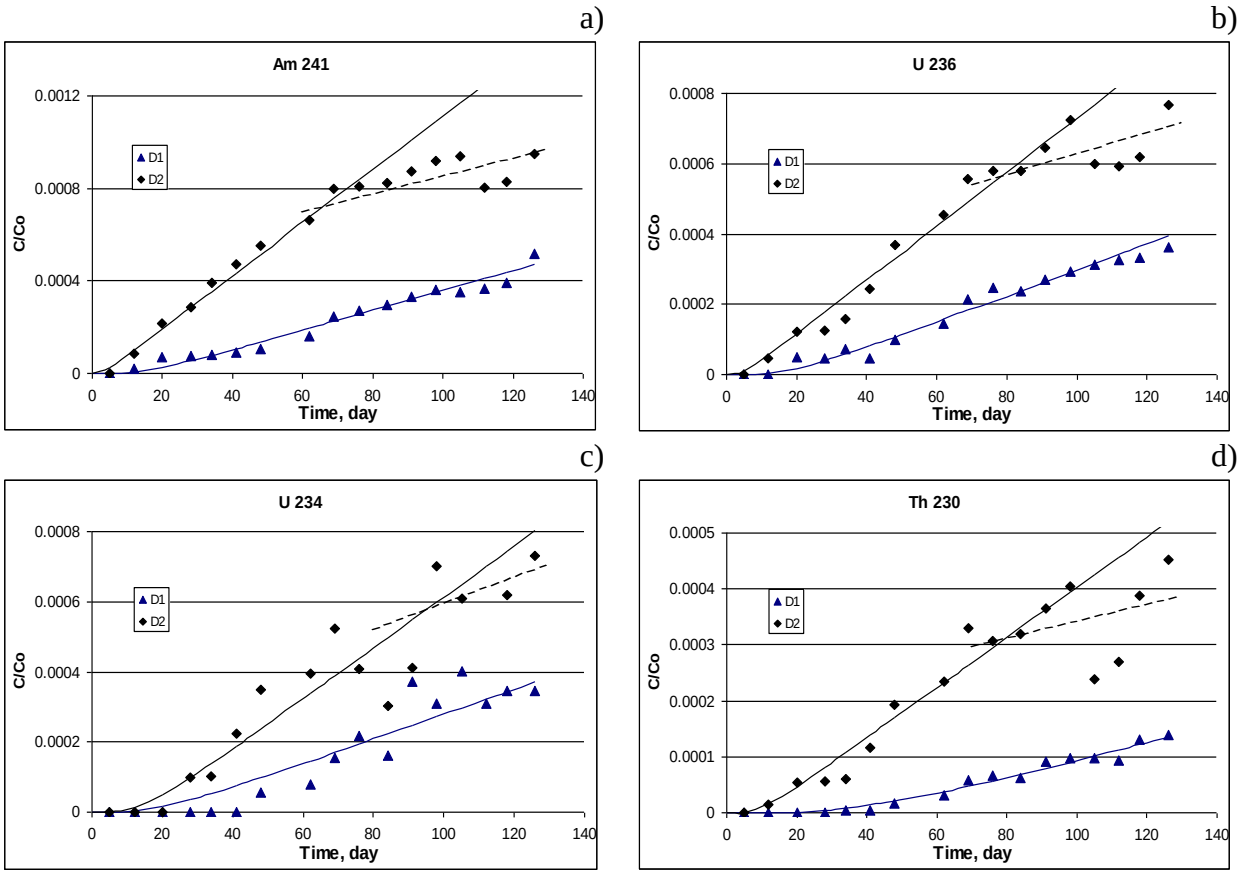


Figure 7: Through-diffusion of ^{241}Am (a), ^{236}U (b), ^{234}U (c) and ^{230}Th (d) tracers in diffusion cells D1 and D2 (measured data (points) and fitted curves (solid lines) linear assumption (dotted lines)).

Apparently, this change of the diffusion coefficient with the time may be explained by the sorption process of the radioactive tracers and brine salts on the surface of pores in graphite. Particularly, this is verified by the comparative analysis of radioactive tracers concentration in left and right containers for the diffusion cell D2, Table 4. For all radioactive tracers, the concentration in the left container decreases by 2 to 3%. However, the value of this decrease (dL/R2-17) is 30 ÷ 56 times higher than the tracer concentration in the right container.

Cell	Name	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
D2	L2-1	0	696.6	707.7	53.0	2996.3
	L2-2	126	681.3	690.4	51.4	2904.5
	dL(dL,%)	0-126	15.3 (2.2%)	17.3 (2.4%)	1.6 (3.1%)	91.8 (3.1%)
	R2-17	126	0.512	0.306	0.037	2.713
	dL/R2-17		30.0	56.4	44.0	33.8

Table 4: analysis of radioactive tracers concentration in left and right containers for the diffusion cell D2,

2.2.2 Radioactive samples from the HTR graphite fuel element (AVR-2) and from the AVR wall graphite block (AVR-1)

2.2.2.1 Sample AVR-2

The sample AVR-2 after the preparation was carefully washed in 20 ml distilled water during 30 minutes in ultrasonic bath. This operation was repeated ten times. The dependence of the radionuclide content in the sample on the operation number is shown in Fig. 8. After seven operations the level of the ^{60}Co and ^{137}Cs radionuclide content remains afterwards constant. The level of the ^{154}Eu radionuclide content decreased during all washed operations and indicate the leachability of this isotope. After washing of the sample, the contents of radionuclide decreases lightly: ^{154}Eu – 395.6 Bq, ^{60}Co – 3459.1 Bq; ^{137}Cs – 21966.2 Bq (for comparison, the radionuclides content in initial sample: ^{154}Eu – 396.9 Bq/g; ^{60}Co – 3526.2 Bq/g; ^{137}Cs – 23238.9 Bq/g).

At the next following diffusion experiment, the ^{154}Eu and ^{60}Co radionuclide contents in the right side of the diffusion cell were lower than the detection level. The small increasing on the 0.03% occurs for the ^{137}Cs radionuclide. The full quantities of the radionuclide releases are following:

^{154}Eu = 1.07% > ^{137}Cs = 0.55% > ^{60}Co = 0.39%

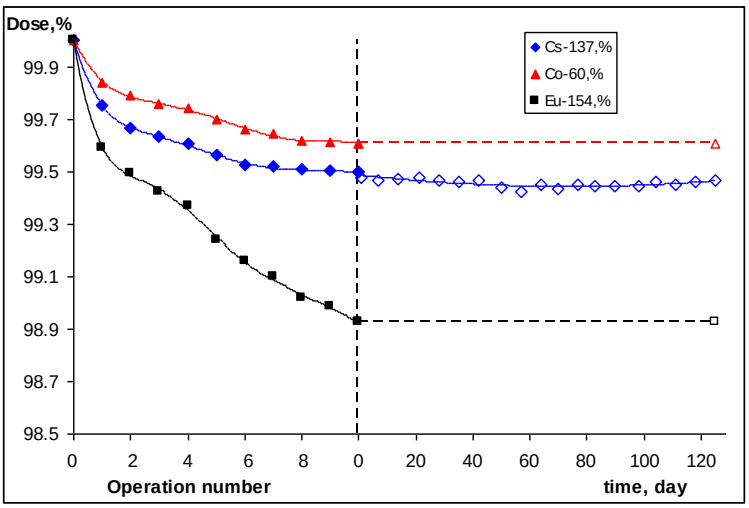


Figure 8: ^{60}Co, ^{137}Cs and ^{154}Eu radionuclide contents in AVR-2 sample depending on the washe diffusion experiment.

The diffusion experiments were carried out during 125 days. The after-treatment experimental data of the current concentrations (C) of radioactive tracers are presented in Appendix 4. During all time, the measured values of the current concentrations (C) of 230Th, 241Am, 236U radioactive tracers were lower than the detection level (< 0.003Bq/ml). This fact allows us to

assert the absence of radioactive tracers diffusion through the graphite covering of HTR fuel element. The highest possible value of the effective diffusion coefficients (De) may be estimated by using of equation (4) under the following assumption: after 125 day the concentrations of ^{230}Th, ^{241}Am, ^{236}U radioactive tracers equal detection level. The results of this estimation are presented in Table 5.

Sample		^{230}Th	^{241}Am	^{236}U
AVR-2	De*1E-15,m^2/s=	< 0.84	< 0.09	< 0.46

Table 5: radioactive tracers equal detection level

2.2.2.2 Sample AVR-1

The sample AVR-1 after preparation was carefully washed in 20 ml distilled water during 30 minutes in ultrasonic bath. This operation was repeated ten times. The dependence of the radionuclide content in the sample on the operation number is shown in Figure 9. The level of the ^{60}Co, ^{137}Cs and ^{154}Eu radionuclide contents decreased during all washed operations. After washing of the sample, the contents of radionuclide decreased significantly (~ 7 ÷ 15%) in comparison with the AVR-2 sample (~ 0.4 ÷ 1.1%), and equal ^{154}Eu – 351.2 Bq, ^{60}Co – 10350.8 Bq; ^{137}Cs – 15447.9 Bq (for comparison, the radionuclides content in initial sample: ^{154}Eu – 391.0 Bq/g; ^{60}Co – 11156.6 Bq/g; ^{137}Cs – 16734.9 Bq/g). Moreover, the order of the radionuclide loss is differ from AVR-2 sample as well:

$$^{154}\mathbf{Eu} = \mathbf{14.3\%} > {}^{60}\mathbf{Co} = \mathbf{7.9\%}\,{}^{137} > \mathbf{Cs} = \mathbf{6.8\%}$$

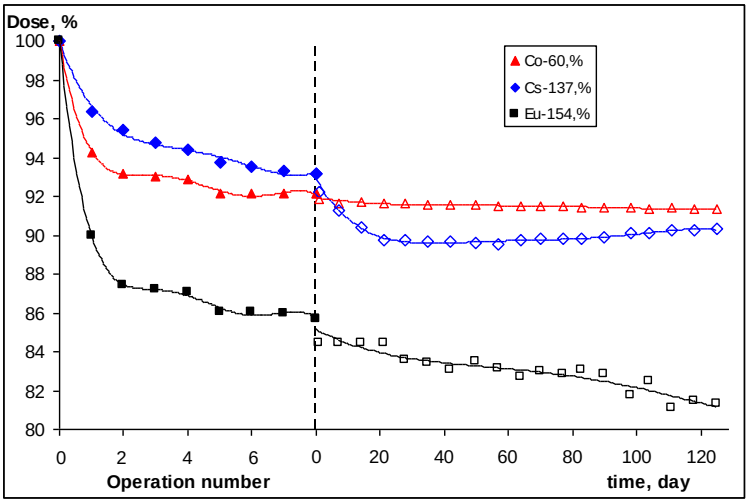


Figure 9: ^{60}Co, ^{137}Cs and ^{154}Eu radionuclide contents in AVR-1 sample depending on the washed operation number and time of diffusion experiment.

During the following diffusion experiment, the ^{60}Co radionuclide release is insignificant (~ 0.8%), Fig. 9. The significant 137Cs radionuclide release takes place during first 30 days

and later it remains constant. The continuous release during 125 days occurs for the ^{154}Eu radionuclide, Figure 25. After the diffusion experiment during 125 days, the order of the ^{154}Eu, ^{137}Cs and ^{60}Co radionuclide loss are similar to that for AVR-2, and the full quantities of the radionuclide releases are following:

^{154}Eu = 18.7% > ^{137}Cs = 9.7% > ^{60}Co = 8.7%

The diffusion experiments were carried out during 125 days. The after-treatment experimental data of the current concentrations (*C*) of radioactive tracers are presented in Appendix 4. The experimental data on the relative concentrations of the different radioactive tracer (^{241}Am, ^{230}Th, ^{236}U) depending on time are shown in Fig. 10. The measured values of the current concentrations (*C*) of ^{241}Am, ^{236}U radioactive tracers can be described as the sum of two linear dependences with break point at 19 and 30 days, respectively. Each linear dependence corresponds to the appropriate effective diffusion coefficient *De1*, *De2*, and capacity factor (α) was estimated by equations (4, 5). For the ^{230}Th tracer the measurements were made 21 days only. The values of the effective diffusion coefficient (*De*) and capacity factor (α) are summarized in Table 6.

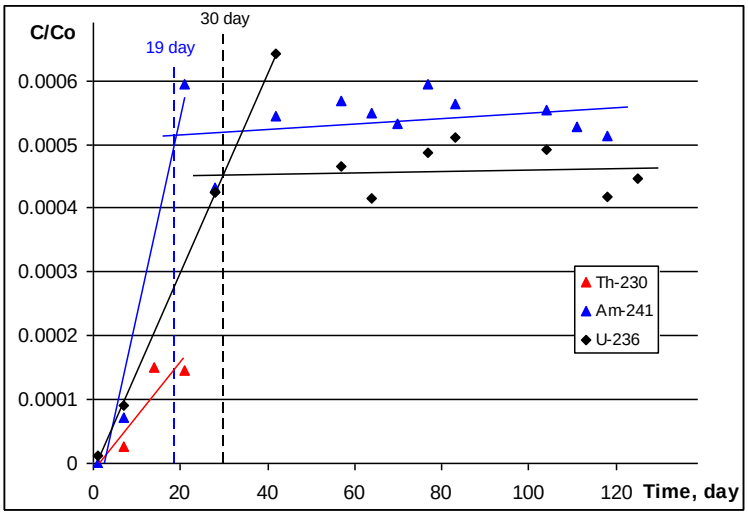


Figure 10: Through-diffusion of ^{241}Am, ^{236}U (b) and ^{230}Th (d) tracers in diffusion cells AVR-1.

Sample		^{230}Th	^{241}Am	^{236}U
AVR-1	De1*1E-15,m^2/s=	116.20	281.92	214.95
	a*1E-3	2.34	3.51	2.40
	De2*1E-15,m^2/s=	---	5.87	1.76
	De1/De2=	---	48.1	122.1

Table 6: diffusion coefficient of AVR-1 graphite

The values of the effective diffusion coefficient at short time (*De1*) are really high, what is apparently determined by the low density of AVR-1 samples (1.72 g/cm^3). The decreasing of the diffusion coefficient (*De2*) at long times can be also determined, similar to that in case of

the nonradioactive samples D1 and D2, by sorption process of radioactive tracers and brine salts on the surface of pores in graphite. However, in case of low density radioactive sample AVR-1, this change (parameter *De1/De2*) is significant (in 50 ÷ 120 times), shown in Table 10. For AVR-1 sample, the effective diffusion coefficient (*De*) decreases, similar to that in case of the nonradioactive samples D1, D2, in the following way:

$$^{241}\mathbf{Am} > {}^{236}\mathbf{U} > {}^{230}\mathbf{Th}$$

Moreover, the relation between the effective diffusion coefficients De^{Th230}/De^{Am241} and De^{U236}/De^{Am241} are near for all investigated samples, shown in Table 7.

Sample		^{241}Am	^{236}U	^{230}Th
AVR-1	$De1/De1_{Am}$	1	0.76	0.41
D2	$De1/De1_{Am}$	1	0.60	0.35
	$De2/De2_{Am}$	1	0.78	0.40
D1	$De1/De1_{Am}$	1	0.86	0.38

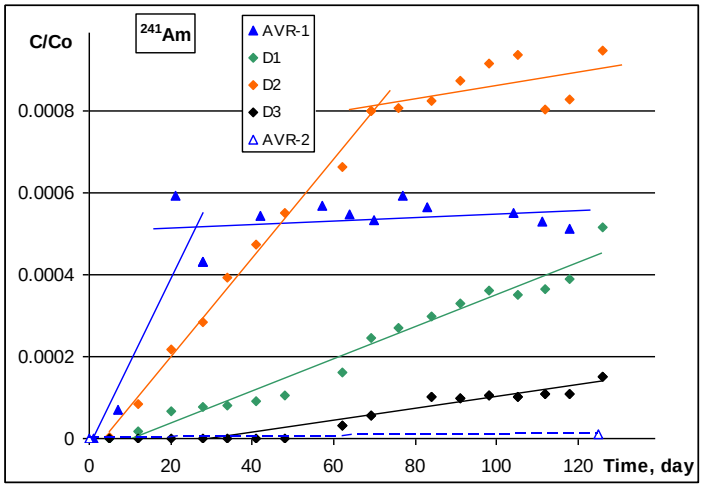
Table 7: relation between the effective diffusion coefficients De^{Th230}/De^{Am241} and De^{U236}/De^{Am241}

The experimental data for the relative concentrations of the different radioactive tracers (^{241}Am, ^{230}Th, ^{236}U) depending on time, and the corresponding break-through curves are shown in Figure 27 for all investigated samples. The values of the effective diffusion coefficient (*De*) and capacity factor (α) are summarized in Table 8.
The effective diffusion coefficients (*De*) for all radioactive tracers decreased at the increasing of the samples density in following way:

AVR-1 > D2 > D1 > D3 > AVR-2

This fact indicates the diffusion coefficient is mainly determined by the density of the samples.

a)



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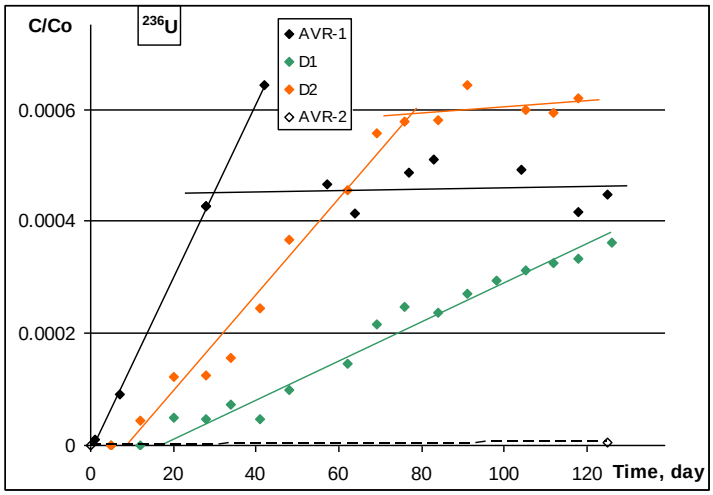

c)

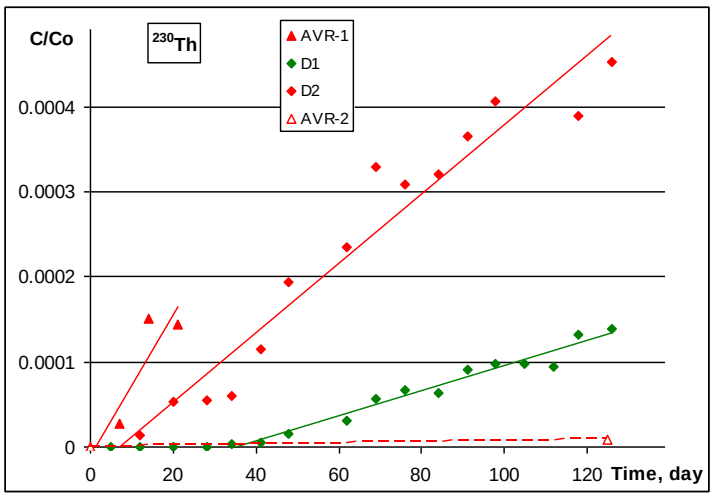

Figure 11: Through-diffusion of ^{241}Am (a), ^{236}U (b) and ^{230}Th (c) tracers for all investigated samples.

3 Conclusion

The diffusion of ^{241}Am, ^{230}Th, ^{234}U and ^{236}U radioactive tracers through HTR graphite was investigated in detail. The analytical solution of Fick's second equation (1) and linear diffusion assumption (4, 5) for the geometry of the through-diffusion cells was used for the description of the experimental data and calculation of the effective diffusion coefficient (*De*) and capacity factor (α). The calculated values of the effective diffusion coefficient (*De*) and capacity factor (*α*) for all investigated samples are summarized in Table 12.

Sample		^{241}Am	^{236}U	^{230}Th
AVR-1	De1*1E-15,m^2/s= a*1E-3	281.92 3.51	214.95 2.40	116.20 2.34
	De2*1E-15,m^2/s=	5.87	1.76	---
D2	De1*1E-15,m^2/s= a*1E-3	28.79 0.50	17.33 0.50	10.06 0.60
	De2*1E-15,m^2/s=	9.58	7.44	3.80
D1	De1*1E-15,m^2/s= a*1E-3	10.71 0.96	9.22 0.97	4.10 0.98
D3	De1*1E-15,m^2/s= a*1E-3	4.23 0.91	--- ---	--- ---
AVR-2	De1*1E-15,m^2/s=	< 0.09	< 0.46	< 0.84

Table 12: effective diffusion coefficients (De) and capacity factor (α) for all investigated samples

The performed investigations showed that the diffusion parameters (*De*, *α*) are mainly determined by the density of the samples. For the high density graphite covering of HTR fuel element (sample AVR-2), the absence of the diffusion of the radioactive tracers through was detected. Maximal diffusion takes place for the low density AVR-1 sample (AVR wall graphite). The change of the diffusion coefficients from *De1* to *De2* during the experiment is determined by sorption process of radioactive tracers and brine salts on the surface of pores in graphite.

For all investigated samples (nonradioactive HTR graphite fuel prototype, radioactive HTR graphite fuel element, and AVR wall graphite block), the effective diffusion coefficients (*De1* and *De2*) of the radioactive tracers decrease in the following way:

$$^{241}\mathbf{Am} > {}^{236}\mathbf{U} > {}^{230}\mathbf{Th}$$

Moreover, the relations between the effective diffusion coefficients for the investigated radioactive tracers are constant for the different type of graphite and equal:

$$De^{236U} = (0.6 \div 0.86)De^{241Am}$$

$$De^{230Th} = (0.35 \div 0.41)De^{241Am}$$

$$De^{U234} \sim 0.9De^{U23}$$

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5 Appendix

A1. The measured data of the current concentrations (*C* in Bq/ml) of ^{241}Am, ^{230}Th, ^{234}U, ^{236}U radioactive tracers (samples D1, D2, D3).

Start	17/01/2008					
Name	Data	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R1-1	21/01/2008	5	0	0	0	0
R1-2	28/01/2008	12	0	0	0	0.056
R1-3	05/02/2008	20	0.035	0	0	0.207
R1-4	13/02/2008	28	0.032	0	0	0.227
R1-5	19/02/1900	34	0.05	0.003	0	0.23
R1-6	26/02/2008	41	0.031	0.003	0	0.269
R1-7	04/03/2008	48	0.067	0.011	0.003	0.305
R1-8	18/03/2008	62	0.098	0.022	0.004	0.476
R1-9	25/03/2008	69	0.143	0.039	0.008	0.708
R1-10	01/04/2008	76	0.162	0.045	0.011	0.767
R1-11	09/04/2008	84	0.154	0.042	0.008	0.851
R1-12	16/04/2008	91	0.176	0.061	0.019	0.935
R1-13	23/04/2008	98	0.19	0.064	0.015	1.01
R1-14	30/04/2008	105	0.202	0.064	0.02	0.98
R1-15	07/05/2008	112	0.21	0.062	0.015	1.025
R1-16	13/04/2008	118	0.213	0.087	0.017	1.086
R1-17	21/04/2008	126	0.235	0.092	0.017	1.47
L1-1	17/01/2008	0	745	762	57	3235
L1-2	21/04/2008	126	619	630	47	2755
dL,%			16.9	17.3	17.5	14.8

Start	17/01/2008					
Name	Data	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R2-1	21/01/2008	5	0	0	0	0.007
R2-2	28/01/2008	12	0.031	0.01	0	0.246
R2-3	05/02/2008	20	0.081	0.036	0	0.619
R2-4	13/02/2008	28	0.081	0.036	0.005	0.792
R2-5	19/02/1900	34	0.101	0.039	0.005	1.086
R2-6	26/02/2008	41	0.157	0.076	0.011	1.299
R2-7	04/03/2008	48	0.235	0.126	0.017	1.498
R2-8	18/03/2008	62	0.288	0.151	0.019	1.803
R2-9	25/03/2008	69	0.347	0.21	0.025	2.148
R2-10	01/04/2008	76	0.359	0.193	0.019	2.15
R2-11	09/04/2008	84	0.36	0.202	0.014	2.202
R2-12	16/04/2008	91	0.389	0.224	0.019	2.265
R2-13	23/04/2008	98	0.44	0.25	0.033	2.39
R2-14	30/04/2008	105	0.361	0.143	0.028	2.46
R2-15	07/05/2008	112	0.361	0.168	0.014	2.08
R2-16	13/04/2008	118	0.386	0.249	0.03	2.22
R2-17	21/04/2008	126	0.468	0.28	0.034	2.481
L2-1	17/01/2008	0	705.5	716.8	53.7	3035
L2-2	21/04/2008	126	627.2	635.6	47.3	2674
dL,%			11.1	11.3	11.9	11.9

Start	17/01/2008					
Name	Data	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R3-1	21/01/2008	5	0	0	0	0
R3-2	28/01/2008	12	0	0	0	0
R3-3	05/02/2008	20	0	0	0	0
R3-4	13/02/2008	28	0	0	0	0
R3-5	19/02/1900	34	0	0	0	0
R3-6	26/02/2008	41	0	0	0	0
R3-7	04/03/2008	48	0	0	0	0
R3-8	18/03/2008	62	0	0	0	0.019
R3-9	25/03/2008	69	0	0	0	0.034
R3-10	01/04/2008	76	0	0	0	0.115
R3-11	09/04/2008	84	0	0	0	0.056
R3-12	16/04/2008	91	0	0	0	0.057
R3-13	23/04/2008	98	0	0	0	0.062
R3-14	30/04/2008	105	0	0	0	0.059
R3-15	07/05/2008	112	0	0	0	0.064
R3-16	13/04/2008	118	0	0	0	0.062
R3-17	21/04/2008	126	0	0	0	0.089
L3-1	17/01/2008	0	161.8	164.1	11.5	683.2
L3-2	21/04/2008	126	114.1	116.1	8	588
dL,%			29.5	29.3	30.4	13.9

* Denotation of samples XN-Z corresponds to: X - right (R) or left (L) containers of the diffusion cells; N – number of the diffusion cells D1, D2 or D3; Z – number of sample.
** It is necessary to note that wide disagreement between values of radioactive tracers concentrations in the left containers take place at the start and the end of experiments. The values of *Co* for the L1-2 and L2-2 are smaller that L1-1 and L2-1 by ~17 and ~11 % respectively.
This disagreement disappears after mass correction (Appendix 2).

A2. Mass correction of the measured data of the current concentrations (*C* in Bq/ml) of ^{241}Am, ^{230}Th, ^{234}U, ^{236}U radioactive tracers (samples D1, D2, D3).

For each sample, the mass (m) was measured by weighing using an analytical balance and the sample volume was calculated as Vs = m/ρ, where ρ is the density of Brine-2. In turn, the density of Brine-2 was measured by weighing of 25 mL solution using an analytical balance. The density of Brine-2 equals 1.326 g/cm^3.

Name	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R1-1	5	0.000	0.000	0.000	0.000
R1-2	12	0.000	0.000	0.000	0.057
R1-3	20	0.036	0.000	0.000	0.213
R1-4	28	0.033	0.000	0.000	0.235
R1-5	34	0.052	0.003	0.000	0.239
R1-6	41	0.032	0.003	0.000	0.279
R1-7	48	0.070	0.012	0.003	0.321
R1-8	62	0.103	0.023	0.004	0.500
R1-9	69	0.152	0.042	0.009	0.755
R1-10	76	0.173	0.048	0.012	0.820
R1-11	84	0.164	0.045	0.009	0.906
R1-12	91	0.189	0.066	0.020	1.005
R1-13	98	0.206	0.069	0.016	1.093
R1-14	105	0.218	0.069	0.022	1.059
R1-15	112	0.226	0.067	0.016	1.105
R1-16	118	0.232	0.095	0.019	1.182
R1-17	126	0.253	0.099	0.018	1.583
L1-1	0	732.2	748.9	56.0	3179.3
L1-2	126	676.1	688.1	51.3	3009.3
dL,%		7.7	8.1	8.4	5.3

Name	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R2-1	5	0.000	0.000	0.000	0.007
R2-2	12	0.031	0.010	0.000	0.249
R2-3	20	0.084	0.037	0.000	0.639
R2-4	28	0.084	0.037	0.005	0.818
R2-5	34	0.105	0.041	0.005	1.132
R2-6	41	0.165	0.080	0.012	1.361
R2-7	48	0.248	0.133	0.018	1.580
R2-8	62	0.304	0.160	0.020	1.906
R2-9	69	0.371	0.225	0.027	2.300
R2-10	76	0.383	0.206	0.020	2.296
R2-11	84	0.384	0.216	0.015	2.350
R2-12	91	0.429	0.247	0.021	2.496
R2-13	98	0.482	0.274	0.036	2.619
R2-14	105	0.392	0.155	0.030	2.670
R2-15	112	0.393	0.183	0.015	2.263
R2-16	118	0.411	0.265	0.032	2.364
R2-17	126	0.512	0.306	0.037	2.713
L2-1	0	696.6	707.7	53.0	2996.3
L2-2	126	681.3	690.4	51.4	2904.5
dL,%		2.2	2.4	3.1	3.1

Name	Day	^{236}U	^{230}Th	^{234}U	^{241}Am
R3-1	5				0.000
R3-2	12				0.000
R3-3	20				0.000
R3-4	28				0.000
R3-5	34				0.000
R3-6	41				0.000
R3-7	48				0.000
R3-8	62				0.020
R3-9	69				0.036
R3-10	76				0.124
R3-11	84				0.060
R3-12	91				0.062
R3-13	98				0.068
R3-14	105				0.064
R3-15	112				0.069
R3-16	118				0.068
R3-17	126				0.097
L3-1	0	157.2	159.5	11.2	664.0
L3-2	126	125.1	127.3	8.8	644.6
dL,%		20.5	20.2	21.5	2.9

* It is necessary to note that disagreement between values of radioactive tracers concentrations in the left containers is small at the start and the end of experiments.
** In section “Experimental”, the average values of the initial concentrations (*Co*) of radioactive tracers in Brain-2 solution (^{241}Am – 3087.8 Bq/ml; ^{230}Th – 728.3 Bq/ml; ^{234}U – 54.5 Bq/ml; ^{236}U – 714.4 Bq/ml) are presented. The averaging by L1-1 and L2-2 takes place.
*** Moreover, the correction of the current concentrations (*C*) of radioactive tracers on the value of the input sample was made.

A3. The corrected data of the current concentrations (*C* in %) of ^{241}Am, ^{230}Th, ^{234}U, ^{236}U radioactive tracers (samples D1, D2, D3).

		Do,Bq/ml	732.2	748.9	56.0	3179.3
Start	17/01/2008					
Name	Data	Day	^{236}U,%	^{230}Th,%	^{234}U,%	^{241}Am,%
R1-1	21/01/2008	5	0.00E+00	0.00E+00	0.00E+00	0.00E+00
R1-2	28/01/2008	12	0.00E+00	0.00E+00	0.00E+00	1.78E-03
R1-3	05/02/2008	20	4.92E-03	0.00E+00	0.00E+00	6.79E-03
R1-4	13/02/2008	28	4.77E-03	0.00E+00	0.00E+00	7.73E-03
R1-5	19/02/1900	34	7.33E-03	4.16E-04	0.00E+00	7.90E-03
R1-6	26/02/2008	41	4.76E-03	4.36E-04	0.00E+00	9.17E-03
R1-7	04/03/2008	48	9.86E-03	1.57E-03	5.63E-03	1.05E-02
R1-8	18/03/2008	62	1.46E-02	3.17E-03	7.79E-03	1.63E-02
R1-9	25/03/2008	69	2.15E-02	5.71E-03	1.56E-02	2.46E-02
R1-10	01/04/2008	76	2.47E-02	6.71E-03	2.18E-02	2.70E-02
R1-11	09/04/2008	84	2.36E-02	6.31E-03	1.63E-02	2.98E-02
R1-12	16/04/2008	91	2.70E-02	9.07E-03	3.73E-02	3.31E-02
R1-13	23/04/2008	98	2.94E-02	9.70E-03	3.08E-02	3.60E-02
R1-14	30/04/2008	105	3.13E-02	9.72E-03	4.01E-02	3.51E-02
R1-15	07/05/2008	112	3.25E-02	9.41E-03	3.09E-02	3.65E-02
R1-16	13/04/2008	118	3.33E-02	1.31E-02	3.46E-02	3.90E-02
R1-17	21/04/2008	126	3.62E-02	1.39E-02	3.44E-02	5.17E-02

		Do,Bq/ml	696.6	707.7	53.0	2996.3
Start	17/01/2008					
Name	Data	Day	^{236}U,%	^{230}Th,%	^{234}U,%	^{241}Am,%
R2-1	21/01/2008	5	0.00E+00	0.00E+00	0.00E+00	2.36E-04
R2-2	28/01/2008	12	4.51E-03	1.43E-03	0.00E+00	8.33E-03
R2-3	05/02/2008	20	1.22E-02	5.33E-03	0.00E+00	2.18E-02
R2-4	13/02/2008	28	1.26E-02	5.52E-03	9.74E-03	2.84E-02
R2-5	19/02/1900	34	1.57E-02	6.02E-03	1.03E-02	3.92E-02
R2-6	26/02/2008	41	2.44E-02	1.16E-02	2.23E-02	4.74E-02
R2-7	04/03/2008	48	3.68E-02	1.94E-02	3.49E-02	5.51E-02
R2-8	18/03/2008	62	4.55E-02	2.35E-02	3.96E-02	6.64E-02
R2-9	25/03/2008	69	5.56E-02	3.29E-02	5.25E-02	8.01E-02
R2-10	01/04/2008	76	5.78E-02	3.08E-02	4.09E-02	8.06E-02
R2-11	09/04/2008	84	5.80E-02	3.20E-02	3.02E-02	8.25E-02
R2-12	16/04/2008	91	6.44E-02	3.65E-02	4.10E-02	8.74E-02
R2-13	23/04/2008	98	7.24E-02	4.05E-02	7.03E-02	9.18E-02
R2-14	30/04/2008	105	5.99E-02	2.40E-02	6.08E-02	9.37E-02
R2-15	07/05/2008	112	5.94E-02	2.70E-02	3.18E-02	8.02E-02
R2-16	13/04/2008	118	6.20E-02	3.88E-02	6.18E-02	8.29E-02
R2-17	21/04/2008	126	7.66E-02	4.52E-02	7.32E-02	9.47E-02

		Do,Bq/ml	157.2	159.5	11.2	664.0
Start	17/01/2008					
Name	Data	Day	^{236}U,%	^{230}Th,%	^{234}U,%	^{241}Am,%
R3-1	21/01/2008	5	0	0	0	0
R3-2	28/01/2008	12	0	0	0	0
R3-3	05/02/2008	20	0	0	0	0
R3-4	13/02/2008	28	0	0	0	0
R3-5	19/02/1900	34	0	0	0	0
R3-6	26/02/2008	41	0	0	0	0
R3-7	04/03/2008	48	0	0	0	0
R3-8	18/03/2008	62	0	0	0	3.04E-03
R3-9	25/03/2008	69	0	0	0	5.64E-03
R3-10	01/04/2008	76	0	0	0	1.89E-02
R3-11	09/04/2008	84	0	0	0	1.00E-02
R3-12	16/04/2008	91	0	0	0	9.84E-03
R3-13	23/04/2008	98	0	0	0	1.07E-02
R3-14	30/04/2008	105	0	0	0	1.02E-02
R3-15	07/05/2008	112	0	0	0	1.10E-02
R3-16	13/04/2008	118	0	0	0	1.08E-02
R3-17	21/04/2008	126	0	0	0	1.52E-02

A4. The corrected data of the current concentrations (C in Bq/ml) of 241Am, 230Th, 236U radioactive tracers (samples AVR-1, AVR-2).

AVR-1

Name	Day	^{230}Th	^{236}U	^{241}Am
R1-1	1	0.045	0.006	0.003
R1-2	7	0.006	0.053	0.213
R1-3	14	0.048	0.278	1.510
R1-4	21	0.044	0.399	1.691
R1-5	28	---	0.230	1.199
R1-6	35	---	0.392	0.000
R1-7	42	---	0.358	1.491
R1-8	50	---	0.240	0.000
R1-9	57	---	0.262	1.529
R1-10	64	---	0.230	1.543
R1-11	70	---	0.306	1.502
R1-12	77	---	0.271	1.687
R1-13	83	---	0.286	1.587
R1-14	90	---	0.239	---
R1-15	98	---	---	---
R1-16	104	---	0.263	1.474
R1-17	111	---	0.222	1.487
R1-18	118	---	0.233	1.443
R1-19	125	---	0.250	1.284
L1-1	1	325.4	594.2	3001.3
L1-2	2	324.4	592.4	2992.2
L1-19	125	322.9	589.7	2978.6

AVR-2

Name	Day	^{230}Th	^{236}U	^{241}Am
R2-1	1	<0.003	<0.003	<0.003
R2-2	7	<0.003	<0.003	<0.003
R2-3	14	<0.003	<0.003	<0.003
R2-4	21	<0.003	<0.003	<0.003
R2-5	28	<0.003	<0.003	<0.003
R2-6	35	<0.003	<0.003	<0.003
R2-7	42	<0.003	<0.003	<0.003
R2-8	50	<0.003	<0.003	<0.003
R2-9	57	<0.003	<0.003	<0.003
R2-10	64	<0.003	<0.003	<0.003
R2-11	70	<0.003	<0.003	<0.003
R2-12	77	<0.003	<0.003	<0.003
R2-13	83	<0.003	<0.003	<0.003
R2-14	90	<0.003	<0.003	<0.003
R2-15	98	<0.003	<0.003	<0.003
R2-16	104	<0.003	<0.003	<0.003
R2-17	111	<0.003	<0.003	<0.003
R2-18	118	<0.003	<0.003	<0.003
R2-19	125	<0.003	<0.003	<0.003
L2-1	1	326.9	596.9	3015.1
L2-2	2	325.9	595.1	3005.9
L2-19	125	324.9	593.3	2996.7