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- Water saturation of the graphite pore system -

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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. Access of water to the pore space in graphite as first step for the release of radionuclides from graphite is studied in this report as well as diffusion of radionuclides in the graphite matrix.

For water uptake tests a closed system was constructed. Graphite samples were put into salt brine under the absence of air and under negative pressure to facilitate the water ingress into the pores for ca.150 day tests. After several days of testing the uptake remains stable The pore system is saturated. In a bigger pore system (Carbon) brick the amount of up taken water is bigger and the time until a steady state is reached is longer.

For diffusion experiments water-saturated samples were mounted between two 20 ml Teflon containers with rubber O-rings as sealings. One container was filled with distilled water the other container was filled with radioactive tracers The performed investigations show that the diffusion parameters are mainly determined by the density of the samples. For high density graphite the absence of the diffusion of the radioactive tracers was detected. Maximal diffusion takes place for the low density sample (AVR wall graphite).

This report is focused on graphite from the thermal column of the German MERLIN (FRJ1) MTR, in Juelich, and on the A3-3 fuel matrix of HTR as well as on irradiated pieces from the AVR reflector structures.

Revisions

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1 Introduction

Actual data on water uptake have been retrieved from the two reactor types having been operated at the site of the Juelich research centre, i.e. the FRJ-1 Material Test Reactor (MTR) and the High-Temperature Test Reactor AVR.

The Jülich-1 Research Reactor (FRJ-1), also referred to as MERLIN (Medium Energy Research Light Water Moderated Industrial Nuclear Reactor), was a light-water moderated and cooled swimming pool reactor of British design. The cornerstone in the erection of the reactor building was laid on June 11, 1958. The core was moderated by light water using beryllium-reflectors. The maximum thermal neutronenflux was $1{,}1 \times 10^{14}$ n/cm^2 s. Reactor operation was started on February 23, 1962. The plant was last run at a thermal power of 10 MW and shut down for good in 1985 after 23 years of operation. After the fuel elements had been removed and most of the experimental installations dismantled, some first steps towards demolition were taken in 1995. Demolition on a large scale began in 1996. Graphite taken from the Thermal Column of this reactor serves as sample material for the investigations described in the following Chapters.

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 make the related investigations rather difficult.

Further graphite grades from other sources will be investigated, when samples will be made available.

2 Water-Uptake of Merlin-Graphite

2.1 Origin of MERLIN Graphite

The research reactor MERLIN was a light water moderated and cooled swimming-pool reactor of British construction type. In 1985 the research reactor was decommissioned. Within dismantling 10 t of graphite blocks from thermal columns had to be disposed. Figure 1 shows a

drawing of the research reactor MERLIN (Medium Energy Research Light Water Moderated Industrial Nuclear Reactor). The thermal column is seen here also.

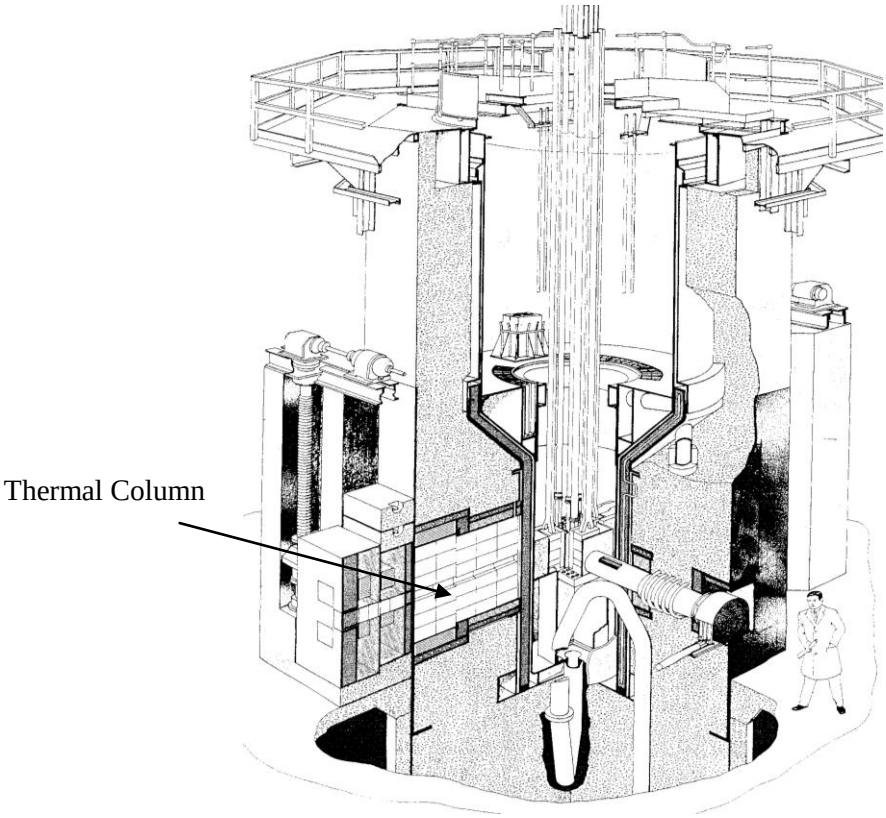


Figure 1: Drawing of MERLIN FRJ-1

Figure 2: graphite block from thermal column

Figure 3: saw-cutted graphite blocks for sample-taking

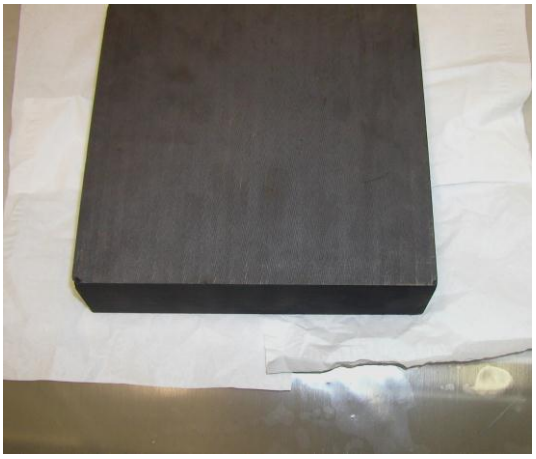


Figure 4: saw-cutted graphite block for sample-taking

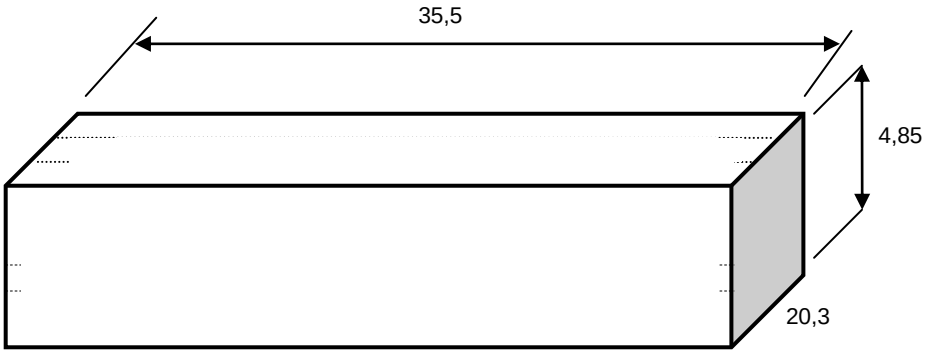


Figure 5: drawing of one saw-cutted graphite block for sample-taking with measures

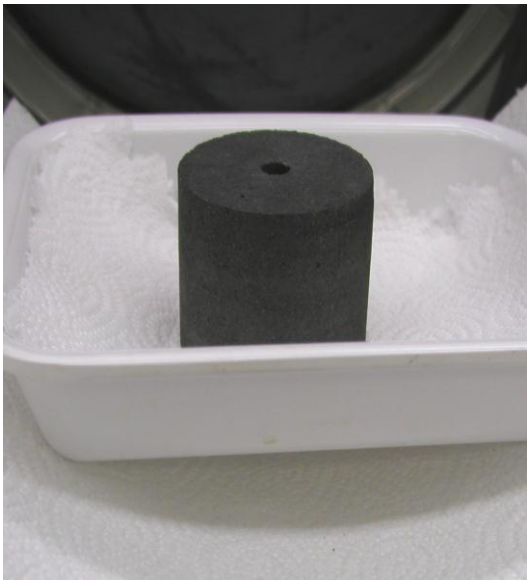


Figure 6: drilled core sample

The samples were taken by core drilling, diameter 5,8 cm, height 4,8 to 5,0 cm. In the middle of the samples and the reference sample is a hole of 0,9 cm from drilling.
The drilled samples were cleaned 15 minutes in an ultrasonic bath with acetone to get the dust from drilling out of the pores.

For the analysed samples with an average weight of 208,4 g the following activities for the 4th of April 2010 were calculated. The results are shown in table 1:

	Bq
H-3	7,0E+05
C-14	7,9E+04
Fe-55	1,9E+04
Co-60	8,0E+02
Ba-133	2,2E+03
Eu-152	1,0E+05
Eu-154	2,8E+03
total	9,08E+05

Table 1: Nuclide inventory

2.2 Apparatus for Water Uptake:

In the following apparatus for water uptake three graphite core drilling samples were tested:

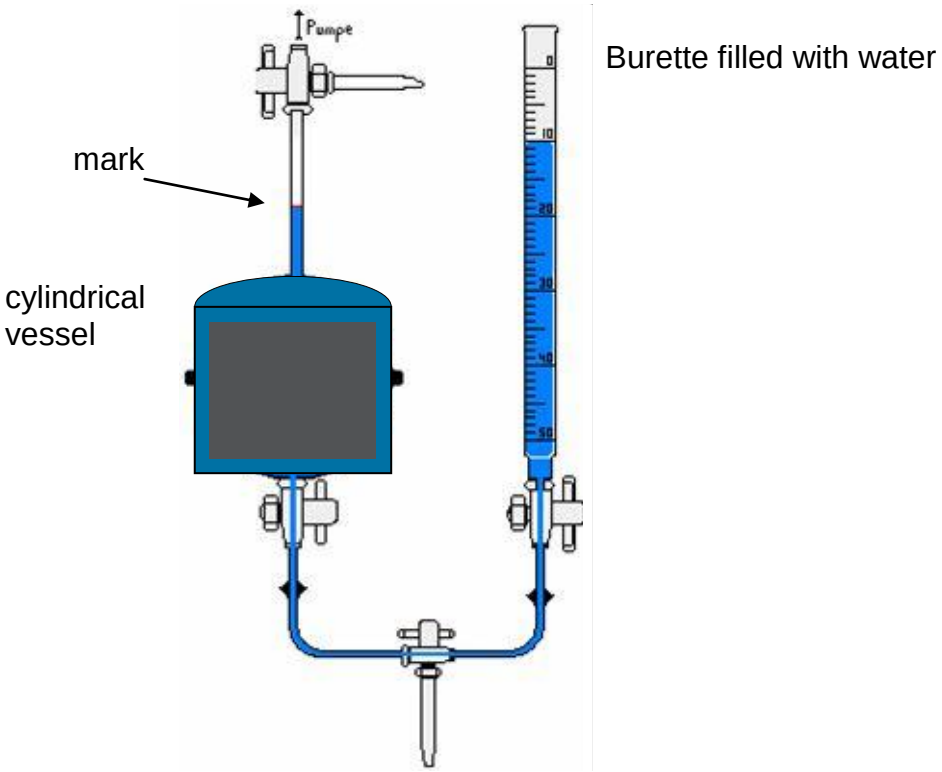


Figure 7: Drawing of the water uptake apparatus

First the dead volume was determined with a reference piece from stainless steel. The dead volume says how much water run into the apparatus without water uptake from pores.

The samples were put for 24 hours into the cylindrical vessel. Water from the burette was filled under vacuum (400 mbar) into the vessel. The burette was filled two to tree times for filling the vessel to the mark. After the vessel was filled, it was aerated. Ambient pressure in the apparatus is necessary to get a stable water balance in the apparatus. After testing the remaining water in the burette was removed, the water in the vessel was collected for further testing.

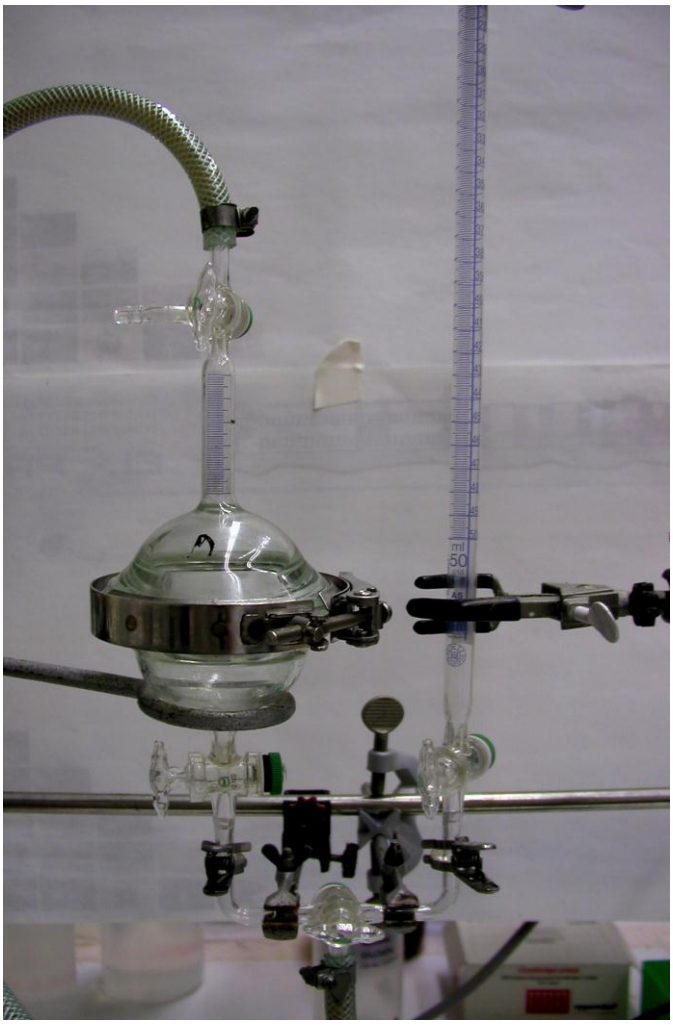


Figure 8: Picture of the water uptake apparatus

2.3 Sample features and results water uptake:

	stainless steel reference	sample 1	sample 2	sample 3
height [cm]	4,80	4,87	4,93	5,05
diameter [cm]	5,75	5,83	5,80	5,79
weight [g]	465,2	203,54	208,70	213,06
water uptake [ml] incl. dead volume	102,8	110,1	105,4	101,01
water uptake during test [ml]	0	0,8	3,6	4,3

Table 2: Sample features

Table 2 shows the measured sample features

Calculation of cylinder volume:

cylindrical volume: $2 \pi r^2 h$

cylindrical volume of samples = external cylinder volume - volume of drill hole

reference: 121,59 cm^3

sample1: 126,91 cm^3

sample 2: 127,12 cm^3

sample 3: 129,75 cm^3

The reference sample volume is smaller than the graphite samples, so that there are to consider only from external volume difference 5,32 ml water ingression more than sample 1, 5,53 ml water ingression more than sample 2 and 8,16 more than sample 3.

Calculation of external surfaces:

cylindrical surface = 2 π r * (h+r)

cylindrical surface of samples = surface of drill hole + external cylinder surface

reference: 153,49 cm²

sample1: 157,63 cm²

sample 2: 158,43 cm²

sample 3: 160,07 cm²

Results:

	sample 1	sample 2	sample 3
total water uptake [ml]	13,4	11,7	10,3
total water uptake per external surface [ml/cm²]	0,085	0,074	0,064
total water uptake per volume [ml/cm³]	0,105	0,092	0,079

Table 3: Calculated results

Table 3 shows the results calculated from sample features.
Total water uptake means water uptake before and during 24 hours testing minus dead volume, considering smaller volume of reference piece.

The water in the apparatus was analysed with γ- measurement after the test. After 24 hours testing there was no radioactivity found in the water.

2.4 Sample features and results brine 2-($MgCl_2$-rich brine) and Mont-Terri salt solution uptake

The pre- test with water for 48 hours show that tests have to run much longer. New tests were established. There were no other radioactive samples available in the needed size than Merlin samples with very low inventory where no leaching of radionuclides was detected. So at AVR virgin graphite and virgin carbon brick samples further uptake was measured Therefore the new measured uptake was not connected with radiochemical measurements The mobility of

radionuclides under repository conditions prior to water-, $MgCl_2$-rich brine and salt solution-ingress is an important concern for final disposal. In Germany $MgCl_2$-rich brine and Mont-Terri-salt solution are model solutions for leaching tests so these liquids were chosen for further tests.

Over 172 days after several times the penetrated volume was noted. Tables 4 and 6 show the sample properties, figures 9 and 10 show the results of uptake in dependence of measurement time

	stainless steel reference	MERLIN Sample	AVR-Virgin	Carbon Brick Virgin
height [cm]	4.80	4.09	5.00	5,61
diameter [cm]	5.75	4.83	4.50	4.85
weight [g]	465.2	208.7	170,2	154.4
Volume [cm³]	121,59	127,12	158,96	129,8
total water uptake [ml] after 174 days	0.0	4,2	2,0	5,6
total water uptake per external surface [ml/cm²]	0.0	0,027	0,019	0,045

Table 4: Sample properties for brine- 2 ($MgCl_2$-rich brine) measurements

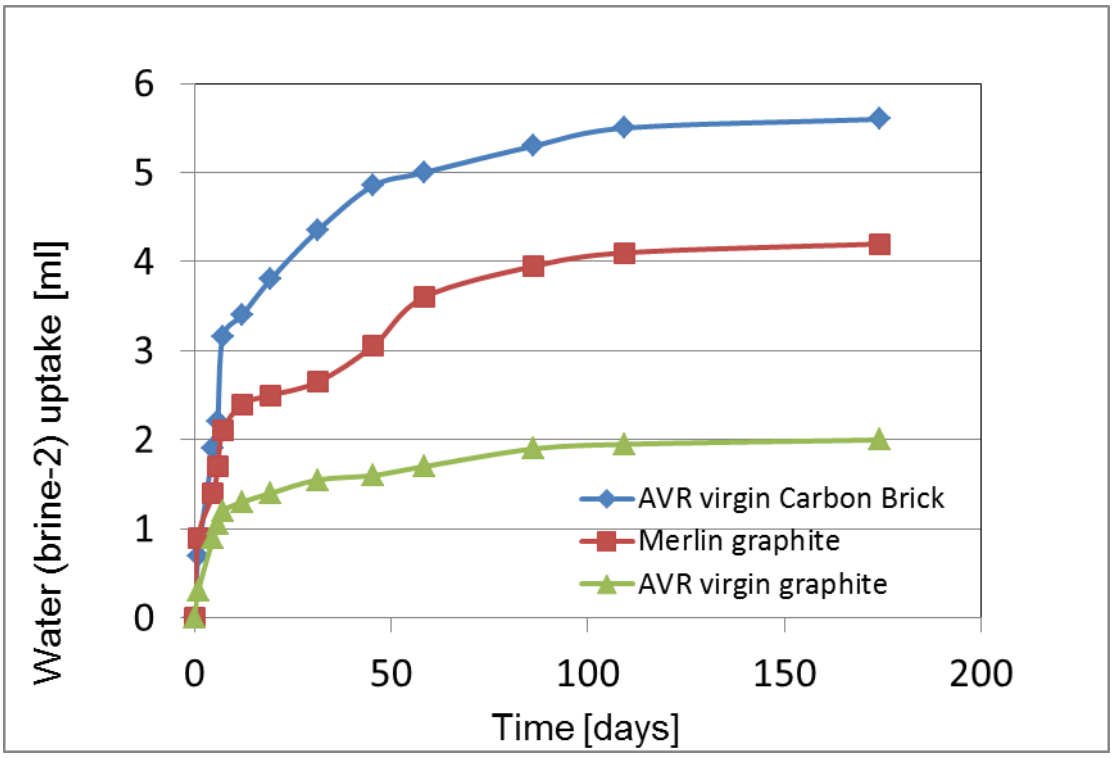


Figure 9: Results of brine 2 ($MgCl_2$-rich brine) measurements

	stainless steel reference	MERLIN Sample	AVR-Virgin	Carbon Brick Virgin
height [cm]	4.80	4.12	4,95	5,52
diameter [cm]	5.75	4.85	4.54	4.73
weight [g]	465.2	209.9	171,3	155.1
Volume [cm³]	121,59	76,12	80,13	97,0
total water uptake [ml] after 174 days	0.0	3,7	1,7	5,2
total water uptake per external surface [ml/cm²]	0.0	0,0032	0,0014	0,0037

Table 5: Sample properties for Mont-Terri salt-solution-measurements

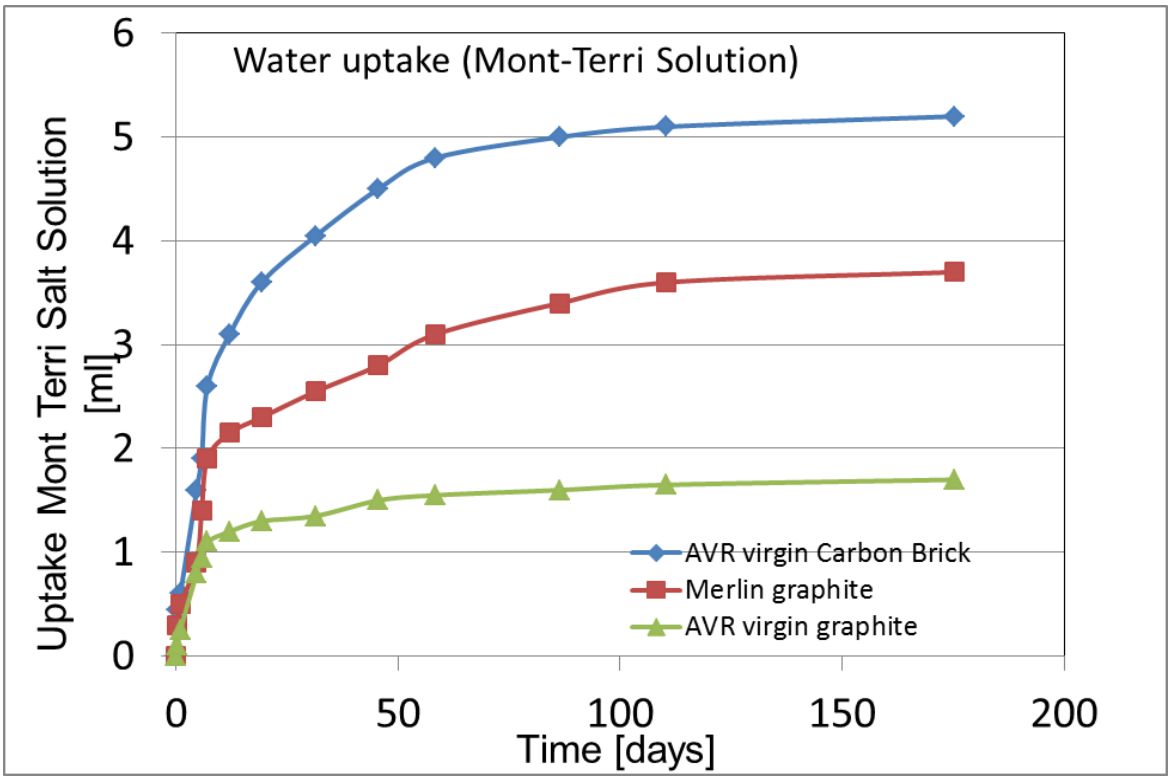


Figure 10: Results of Mont-Terri-salt solution measurements

3 Water Uptake of AVR fuel pebble matrix

3.1 Experimental Set-up

A sketch of the experimental set of the water uptake apparatus is shown in Figure 7. The cylindric vessel is replaced by a spherical flask, which consists of two half spheres for pebble analysing. The half spheres are fixed together by a metal clamp. The valve above the spherical vessel allows the evacuation. Three test series have been performed with different pressures, which could be adjusted by a valve at the vacuum pump.

1. 400 mbar / 0.1 M NaCl
2. 40 mbar / water
3. 2-4 mbar / 0.1 M NaCl

When a stable pressure is adjusted the valve 2 below the flask is opened and the flask is filled with 0.1 M sodium chloride solution or water up to the specified mark. The volume of the solution is measured with the burette. Then the vacuum pump is shut down to establish atmosphere pressure in the flask. The valve 2 is opened again and the flask is filled up again with solution to marked level. The used volume up to this step had been noticed after 1 minute as initial volume V_i. The further water penetration into the pore system of the graphite pebble is compensated again in defined time periods. These additional volumes were noticed as V_t.

3.2 Chloride distribution in the graphite pebble

The pebbles are dried in a furnace at 110°C after the ingression experiments and 10 layers of five 2 mm wide graphite stripes are chipped from the graphite by a lathe. The depth of the layers is shown in Figure 11

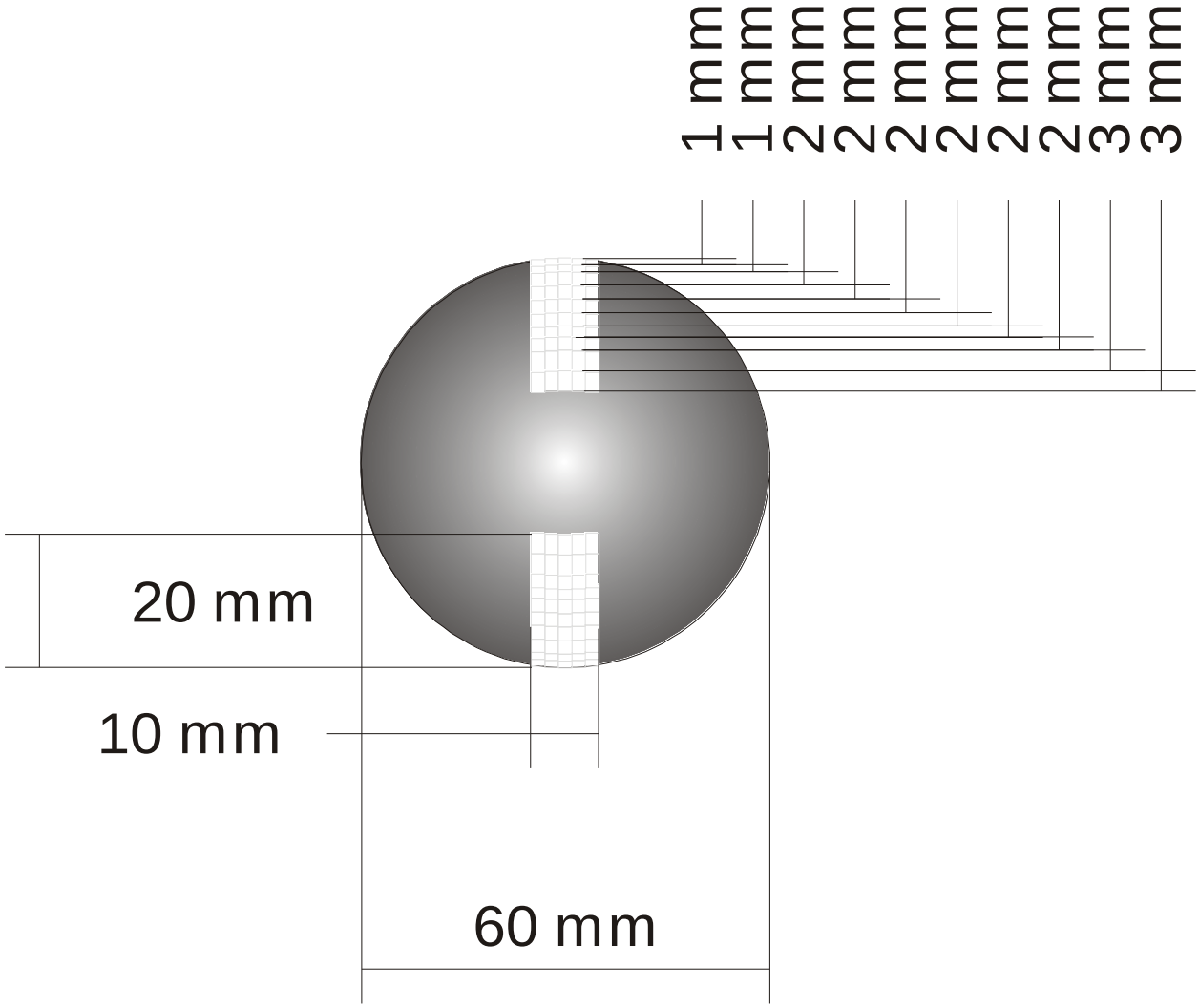


Figure 11: Lathing scheme of the pebble

The graphite chips obtained from the 5 stripes of one layer are mixed and leached in boiling water for 5 hours. The chloride concentration is determined in the filtrate by UV/VIS spectrometry.

3.3 Description of the graphite pebbles

The graphite pebbles had been obtained from an experimental facility, which tested the moving behaviour in an HTR core. Two different types of graphite pebbles had been used in these former experiments, A3-3 graphite and the advanced graphite matrix A3-27. No information was available which type of graphite pebbles was in the obtained transport drum. Following parameters had been determined:

Diameter: 5.95 ± 0.1cm
Mass: 194,5 ± 0,25g
Furthermore the oxidation rate has been measured by thermo-gravimetry to determine the graphite type. Figure 12 shows the measuredoxidation rate to determine the graphite type.

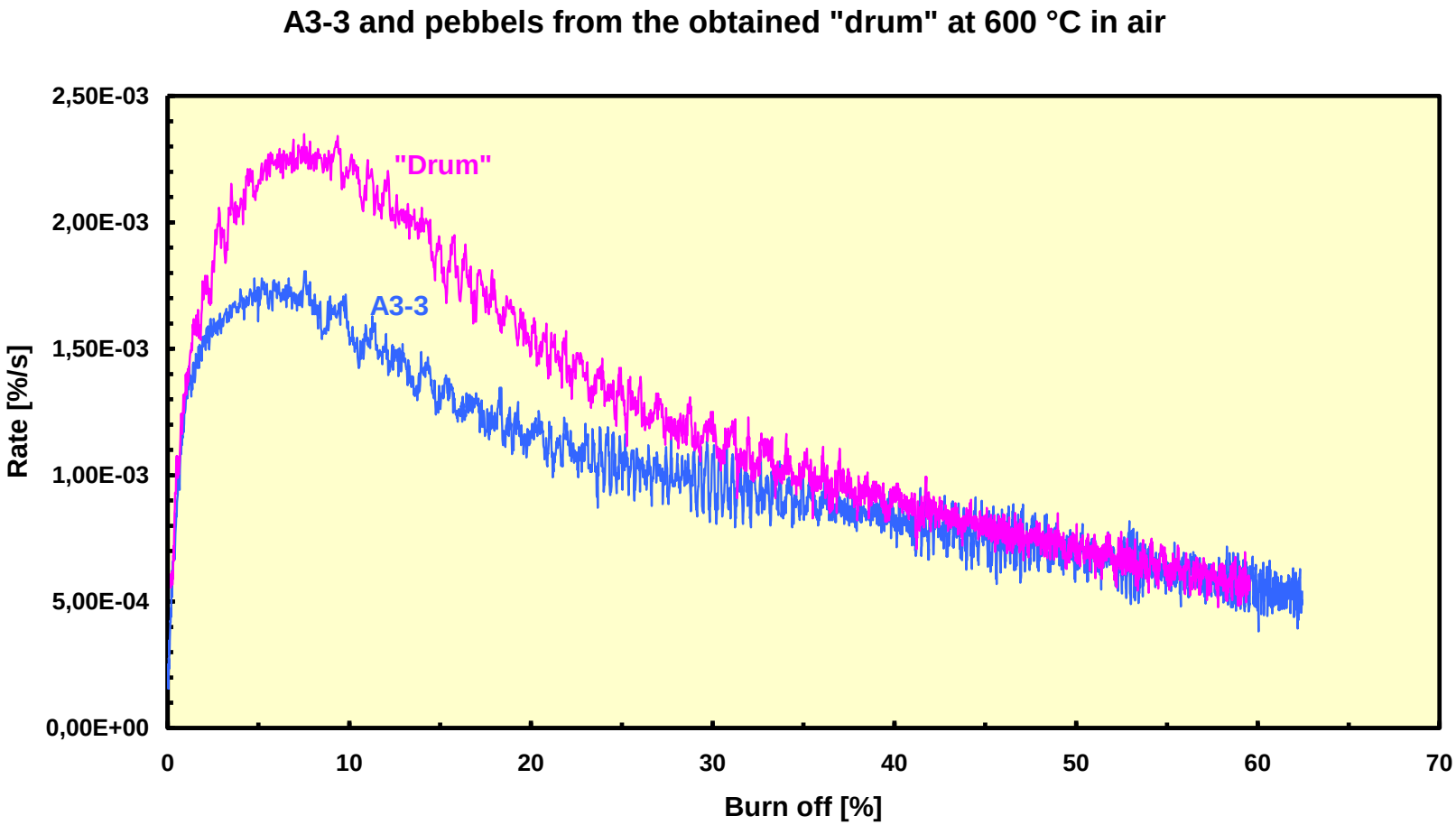


Figure 12: Determiantion of oxidation rates

The shape of the measured oxidation rate curve is similar to well-defined A3-3 graphite. The different height is probably related to up-take of metal impurities and other damages during the moving experiments. The obtained results indicate that the used graphite type is A3-3, which is also more probable because the used amount of A3-3 pebbles was at least two orders of magnitude larger than pebbles made of A3-27.
A first experimental series shows a large scattering of the amount of water in the graphite pores. This could be related to dust on the surface and in the larger pores. Therefore the pebbles were cleaned for further experiments in an ultrasonic bath with ethanol. After cleaning they were dried in a furnace at 90°C. About 80 mg of graphite dust is removed by this cleaning procedure.

3.4 Results of ingression experiments

The first series of ingression experiments were performed with non-cleaned graphite pebbles. The take up of water is nearly completed within 2 hours with a mean water amount of 9.5 ml. A additional take-up of 1 ml solution can be achieved after 16 hours. Picture 13 shows the water uptake results.

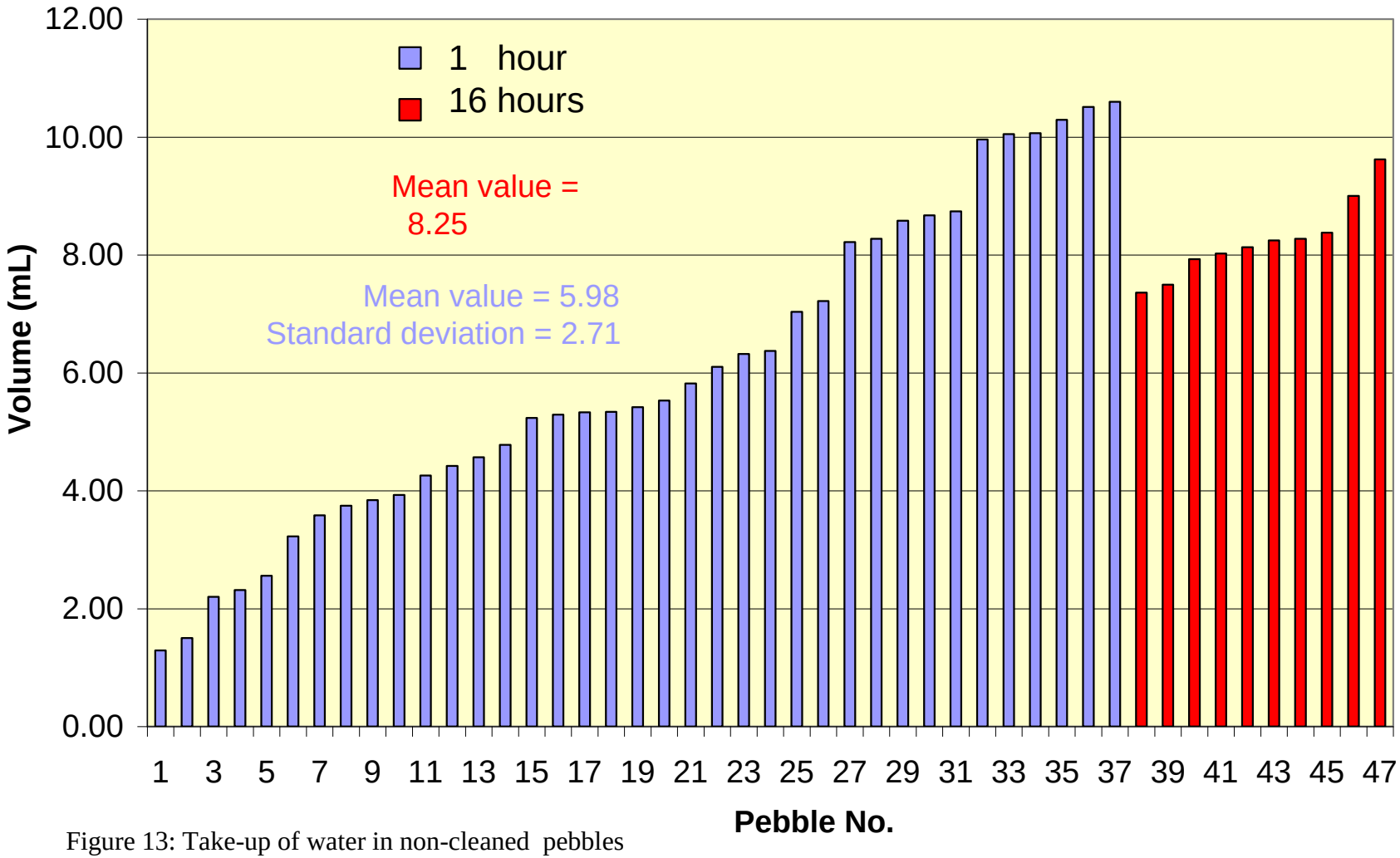


Figure 13: Take-up of water in non-cleaned pebbles

The experiments with non-cleaned graphite show a wide scattered take-up of sodium chloride solution from 1.3 to 10.3 ml after one hour with an mean value of 6 ml. Figure 14 shows the distribution of the number of pebbles related to the water up-take in steps of 0.5 ml.

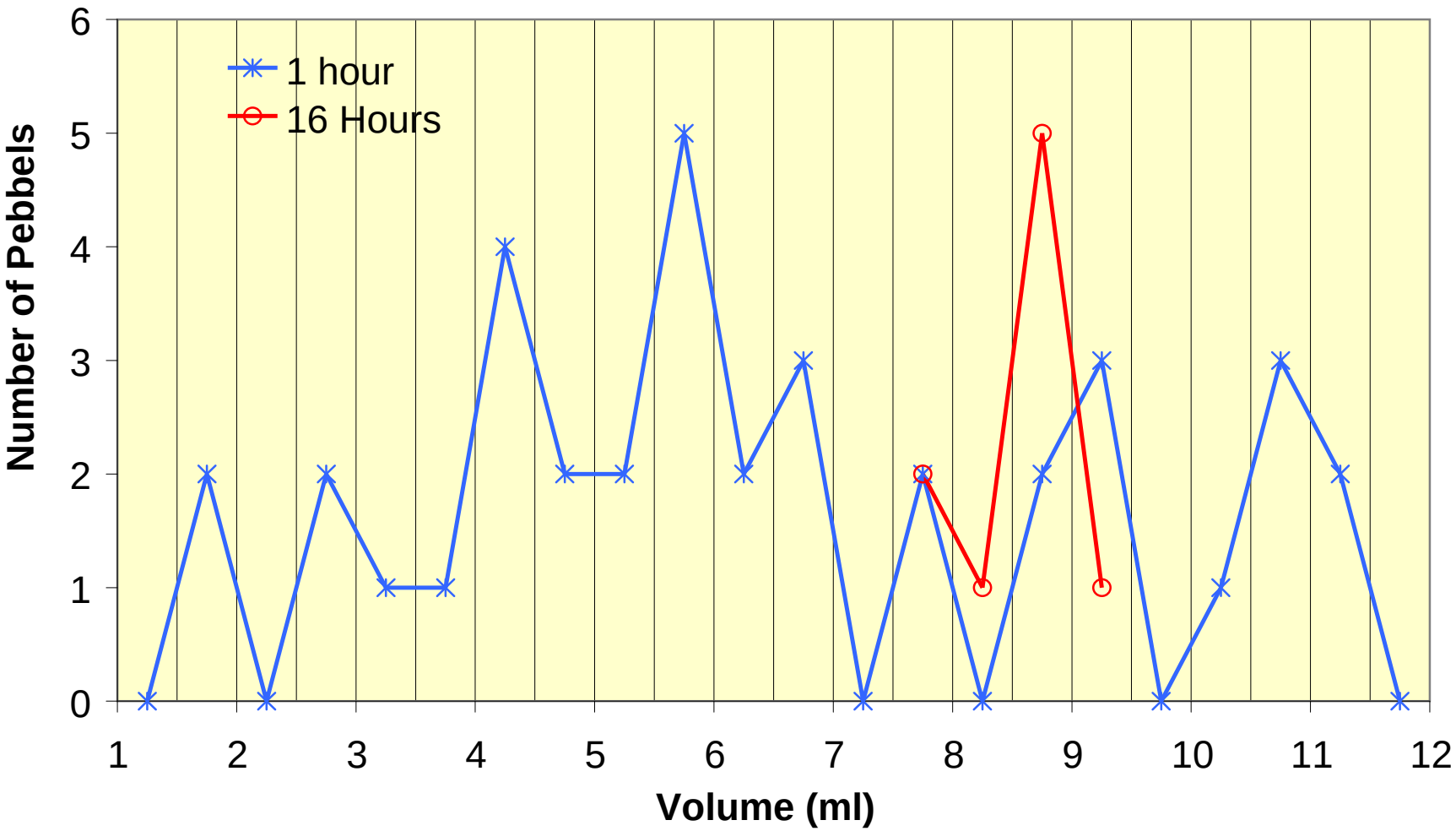


Figure 14: Distribution of the water take-up in non-cleaned pebbles at 2 mbar

No clear defined maximum could be obtained from this figure. These changes after an ingression time of 16 hours. The mean value of the water up-take is increased to 8.25 ml and the distribution curve shows a clear maximum in this area. This wide scattering of the water up-take could be explained by the history of the pebbles. The pebbles had been used for experiments to study their movement in a HTR reactor core. Graphite was rubbed off from the surface of the pebbles during these experiments and fine graphite dust has clogged some surface pores. Therefore further experiments were performed with cleaned graphite pebbles. The Figures 15, 16 and 17 show the results of the water ingression into cleaned pebbles at 2, 40 and 400 mbar. The scattering of the water up-take is reduced in these experiments series in comparison to the non-cleaned pebbles.

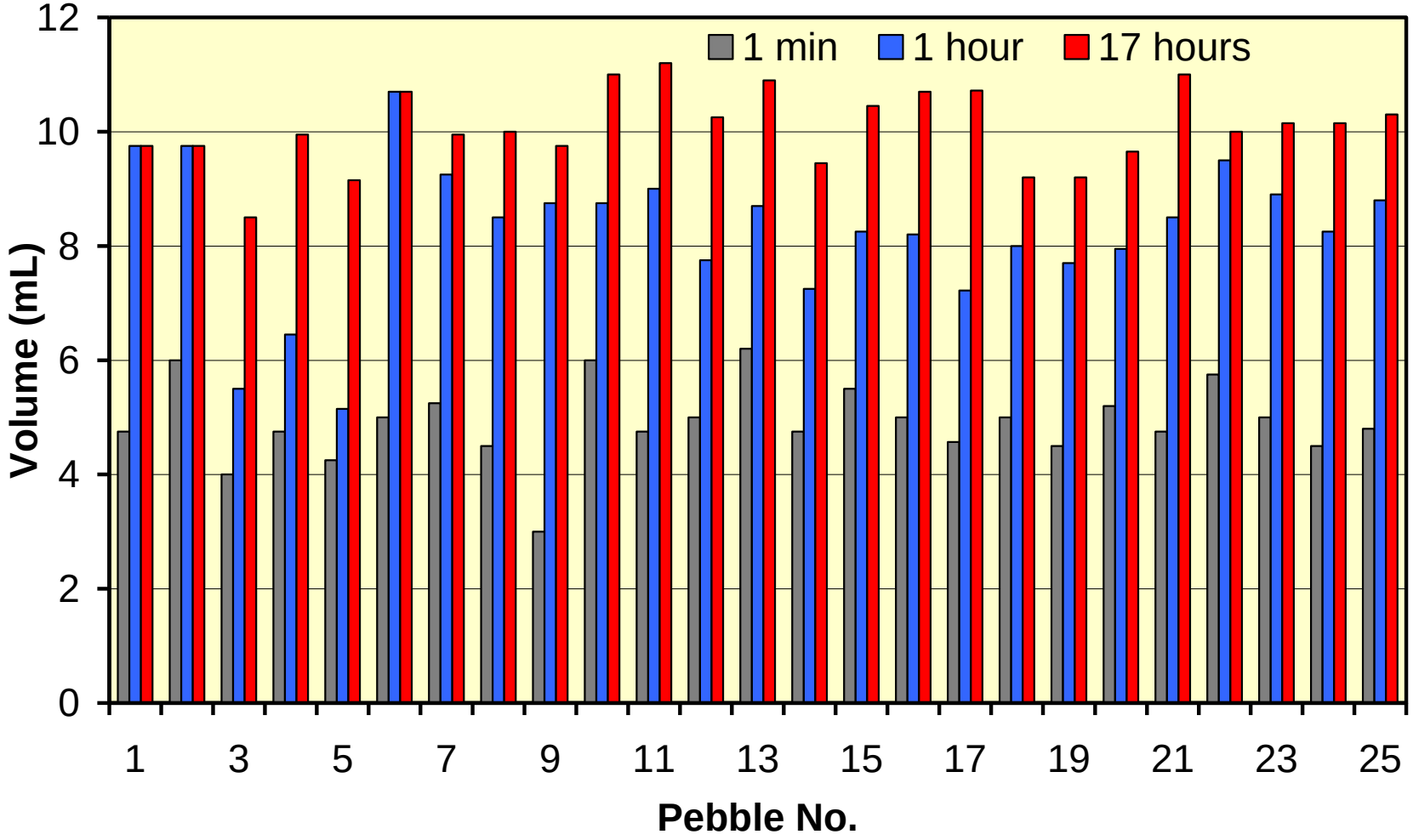


Figure 15 Take-up of water in cleaned graphite pebbles at 2 mbar

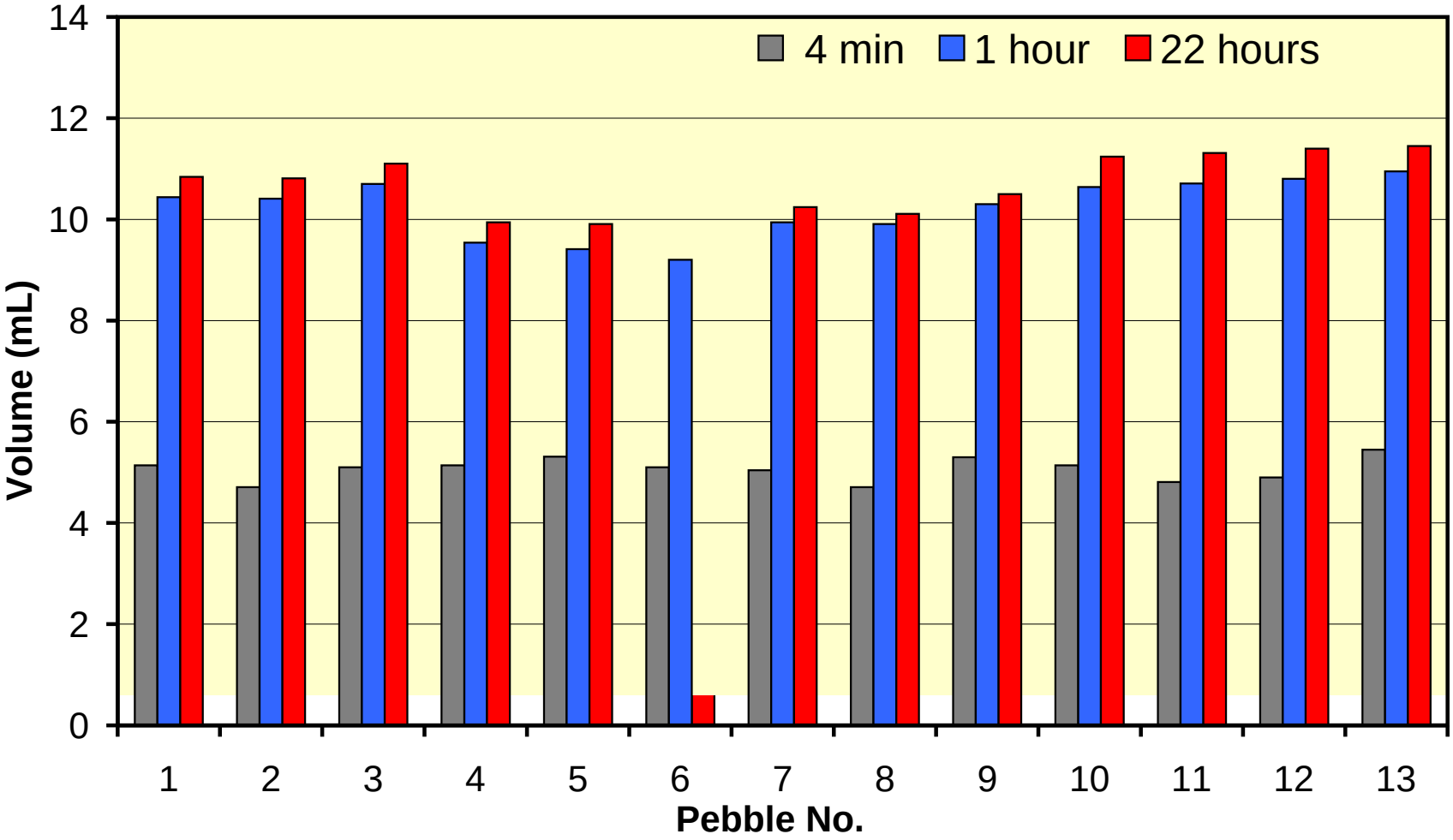


Figure 16: Take-up of water in cleaned graphite pebbles at 40 mbar

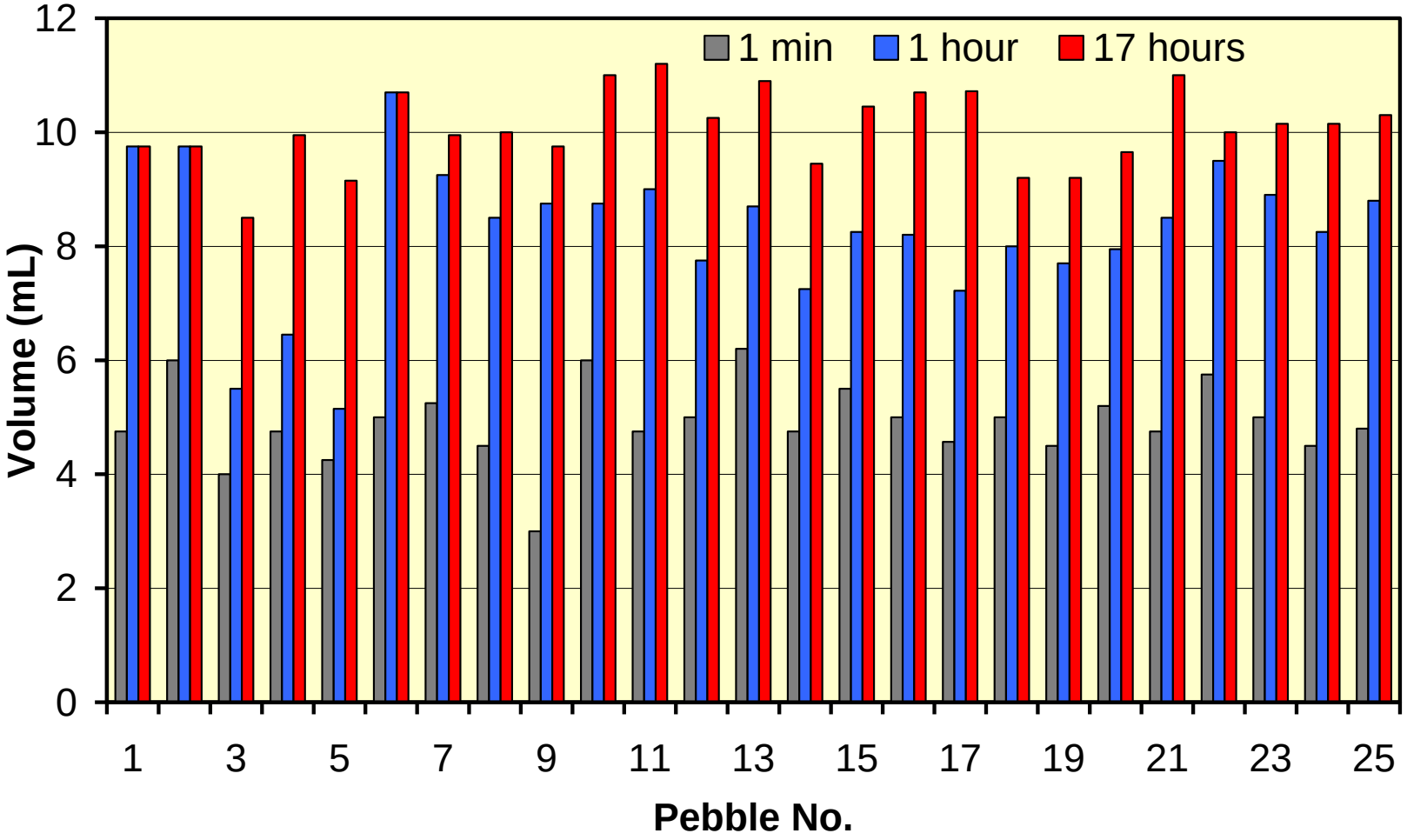


Figure 17: Take-up of water in cleaned graphite pebbles at 400 mbar

Table 6 shows the mean values of the ingression experiments at 2 mbar, 40 mbar and at 400 mbar. The mean values of the water take-up are compatible at the different pressures. However the standard deviations at 40 and 400 mbar are smaller in comparison to the series at 2 mbar. This can also be seen in the distribution of the number of pebbles related to the water up-take. The experimental series at 2 mbar shows only a significant maximum at the instantaneous water take-up after 1 minute. The experimental series at 400 mbar shows significant maxima at all times whereas the amount of penetrated water becomes more scattered in the experimental series at 40 mbar.

Pressure	2 mbar		40 mbar		400 mbar	
Time	∅	Std.dev.	∅	Std.dev.	∅	Std.dev.
(min)	(mL)		(mL)		(mL)	
1			1.7	0.2		
5	4.9	0.7	5.1	0.2	6.1	0.6
10	6.3	1.2	6.4	0.2	7.3	0.5
30	7.5	1.3	9.3	0.3	8.3	0.5
60	8.3	1.3	10.2	0.6	8.9	0.5
120	8.9	1.2	-	-	9.6	0.4
180	9.1	1.2	10.5	0.6	9.6	0.4
1020	10.1	0.7	-	-	10.3	0.4
1320	-	-	10.7	0.6	-	-

Table 6: Take-up of water at different pressures

3.5 Determination of the water distribution in the pebble

Figure 18 shows the distribution of chloride in a pebble after drying, which was taken up with 11.4 ml of a 0.1 M NaCl solution. The graphite contained about 0.23 mg Cl^- mg/g graphite with a standard deviation of 0.03 except the surface layer. Dependence from the depth could not be observed.

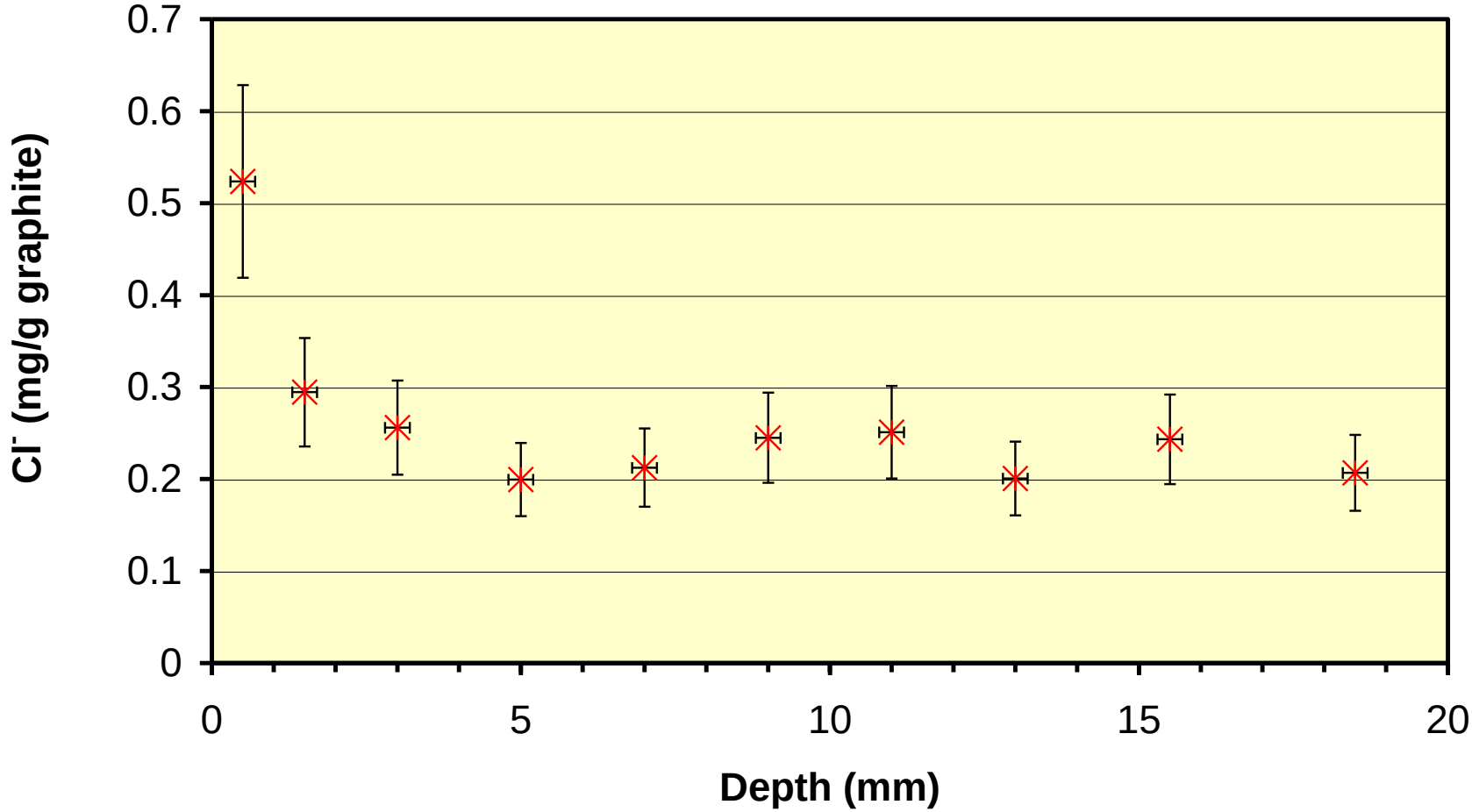


Figure 1: Chloride distribution in a pebble after saturation with 0.1 M NaCl solution

3.6 Conclusions from A3-3 Matrix Investigations

The obtained results show, that a pure A3-3 graphite pebble will take up on an average about 10 to 11 ml of water. The uncleaned pebbles show a wide scatter take-up of solution after one hour and an average water take-up of about 8 to 9 ml after 16 hours. This is about 3 ml less than the average water take-up of cleaned pebbles. This can be explained by clogging of pores with fine graphite particles. Such an effect can be expected also for spent HTR fuel pebbles. However the clogging is causing larger deviations and smaller amounts of water take-up. This would results in a larger number of examinations to obtain a reliable statistic. Furthermore the larger volumes of penetrated solution are conservative for further considerations. Therefore further investigations will be performed only with cleaned graphite.
However the pore volume of cleaned pebbles still varies in a wide range and the take-up of water differs from 9 ± 12 ml. This scattering of the water up-take increases, when up-take reaches the maximum filling volume.

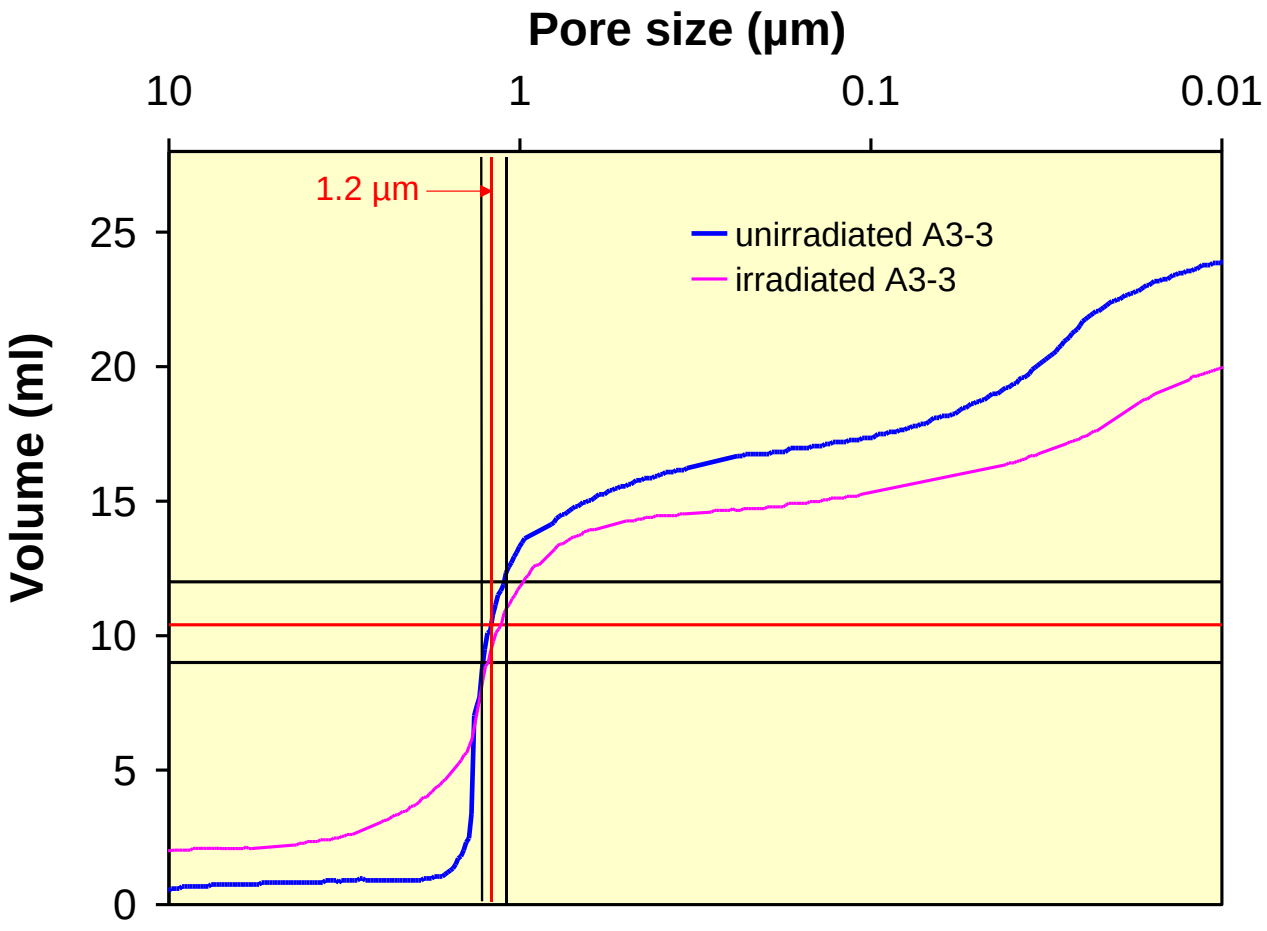


Figure 2: Pore size to volume correlation

This average volume of 10.5 ml indicates that the pores down to size of about 1 µm are filled with water. However the pore size distribution was measure with Hg porosimetry, which correlated the pore size with the pressure necessary to fill the pores. But this method can lead to an opening of closed pores and especially the larger volume of pores < 1 µm. Therefore it is not clear whether the pores > 1µm were filled with water or the open pore volume is smaller than indicated by the blue curve in Figure 19.

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

4.1 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. 20a and Fig. 20b.

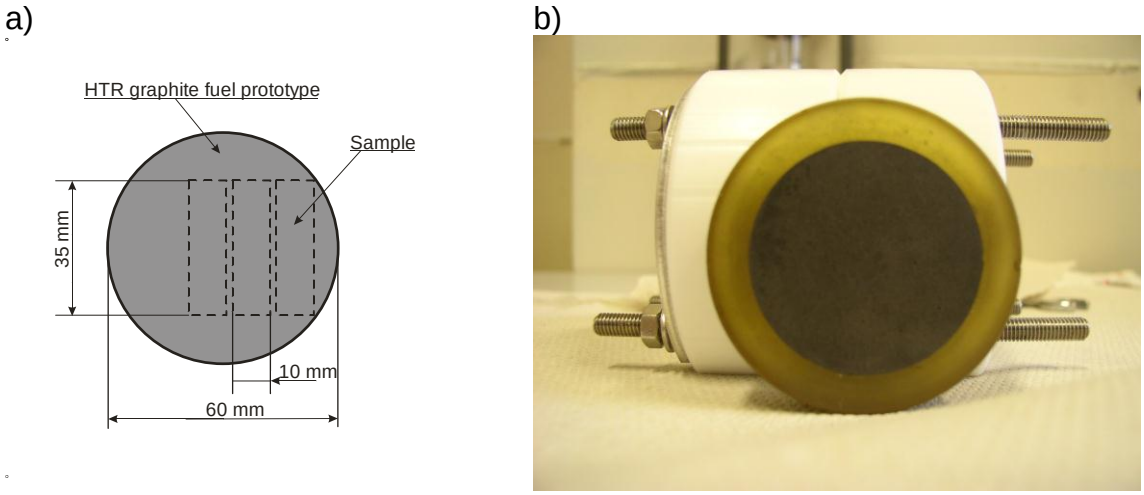


Figure 20 : Scheme of the making of D1, D2, D3 samples (a) and photo of sample (b)

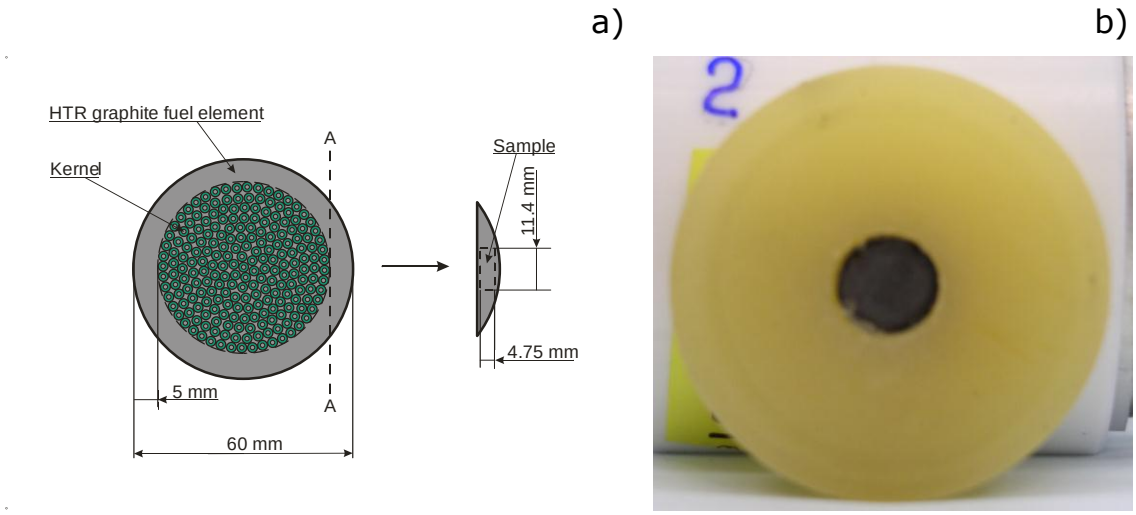


Figure 23. Scheme of the making of AVR-2 samples (a) and photo of sample (b)

The apparatus used for the diffusion experiments is shown in Figure 22

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.

a) b)

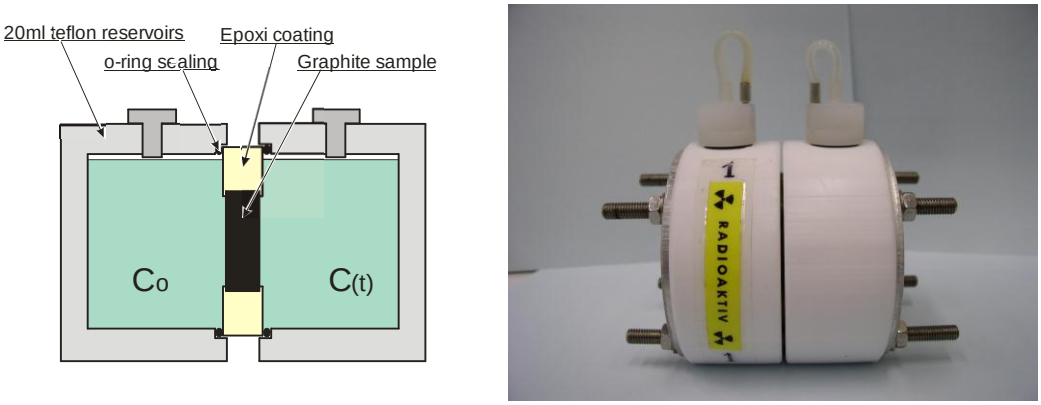


Figure 22: The apparatus for the diffusion experiments

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.

4.2 Results and discussion

4.2.1 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 5.

a) b)

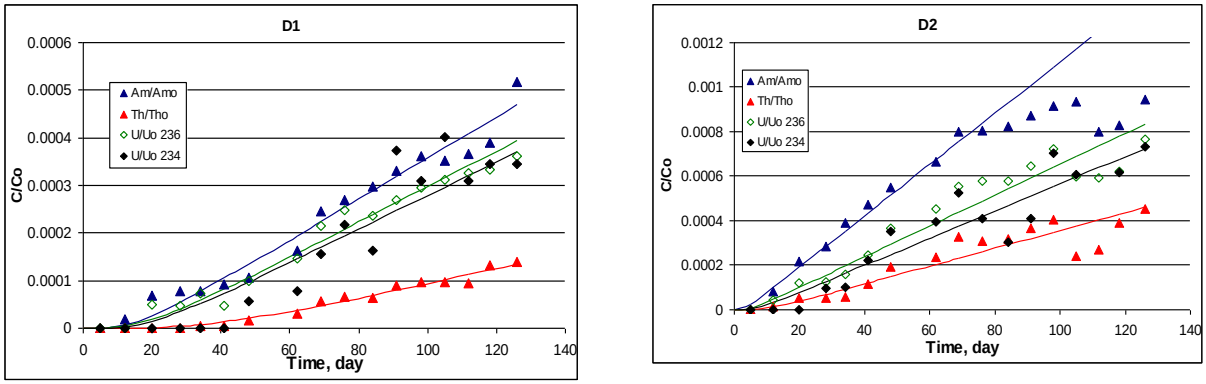


Figure 3: Through-diffusion of ^{241}Am, ^{230}Th, ^{234}U, ^{236}U tracers in diffusion cells D1 (a) and D2 (b).

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 7: effective diffusion coefficient and capacity factor

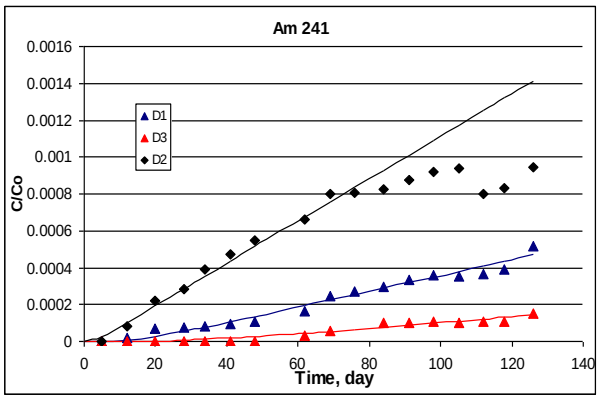
Moreover, the ratio of De^{Am241}/De^{Th230} equals ~ 2.7 for both cells. For the uranium tracers, the values of *De* are nearly equal and, as usual, $De^{U234} \sim 0.9De^{U236}$. For all radioactive tracers, the diffusion coefficients for the diffusion cell D2 are on average in 2.2 times higher then *De* for the diffusion cell D1, it is shown in Table 6.

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

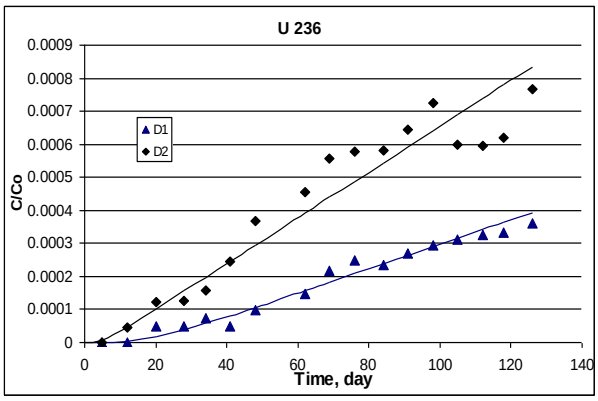
Table 7 : diffusion coefficients for diffusion cell D2

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 21a.

a)



b)



c) d)

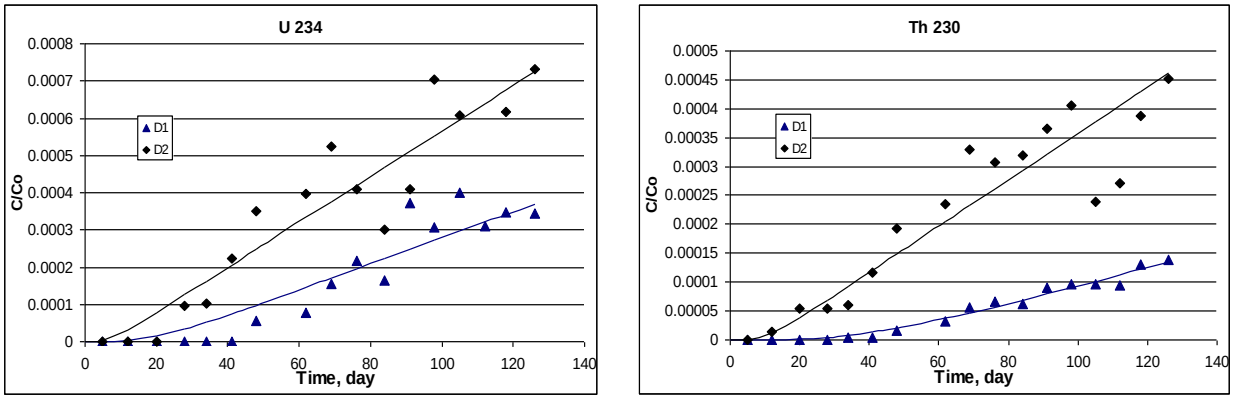


Figure 4: Through-diffusion of ^{241}Am (a), ^{236}U (b), ^{234}U (c) and ^{230}Th (d) tracers in diffusion cells D1, D2 and D3 (measured data (points) and fitted curves (solid lines)).

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:

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

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

As shown in Figure 24, 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*):

$$De = \frac{AVL}{S} \quad (4)$$

$$\alpha = -\frac{6BV}{LS} \quad (5)$$

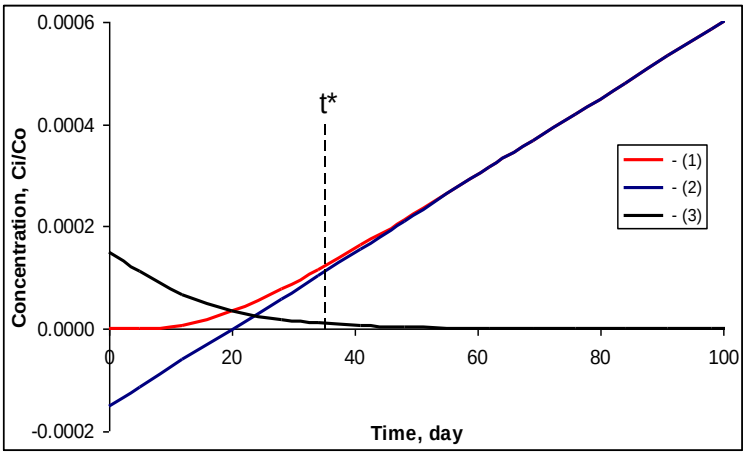


Figure 5: Through-diffusion curve.

Therefore, at the times longer than 60 – 80 days, the dependence *C/Co = f(t)* was assumed as linear (dotted line in Figure 25), 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 8, 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 8: effective diffusion coefficient for the diffusion cell D2 at the times later on 60 – 80 days

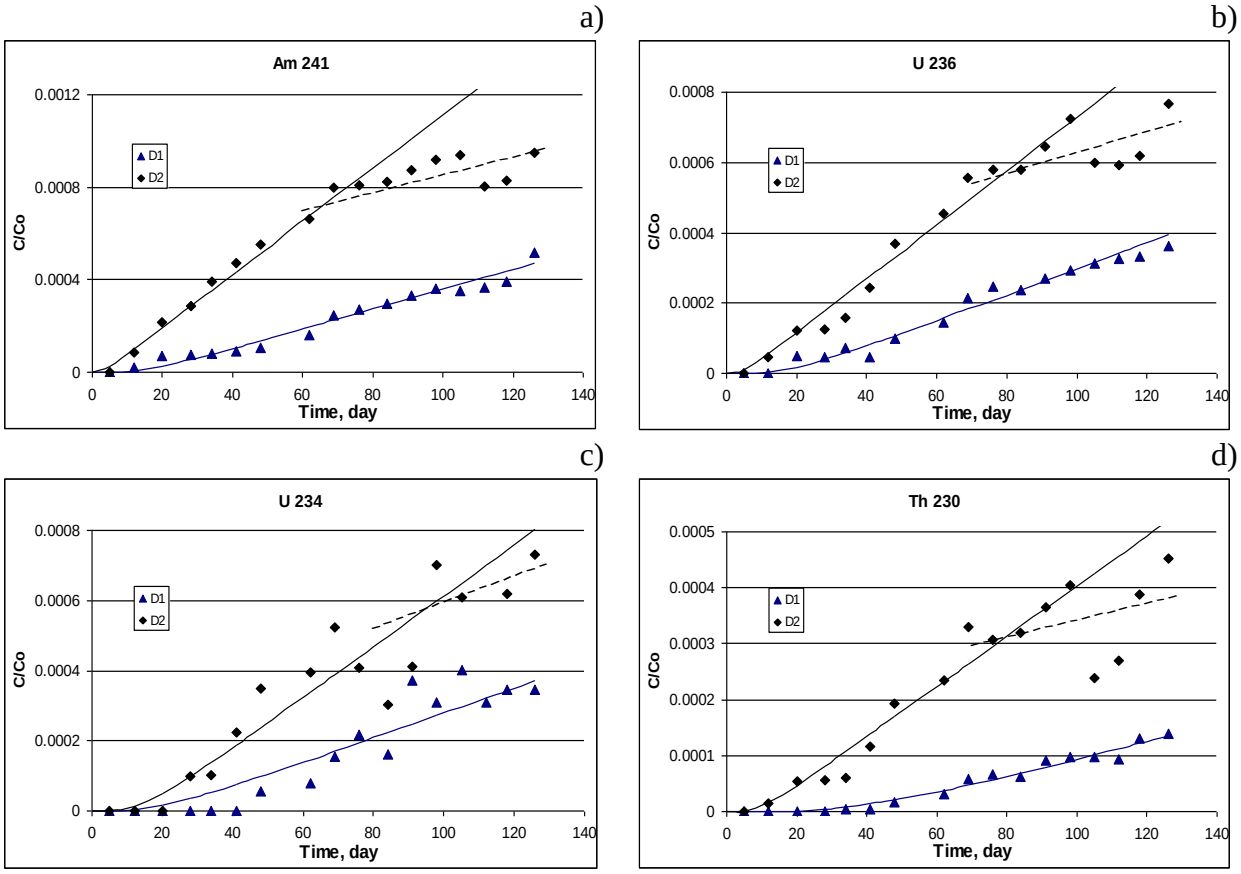


Figure 26: Through-diffusion of ^{241}Am (a), ^{236}U (b), ^{234}U (c) and ^{230}Th (d) tracers in diffusion cells D1 and D2 (measured data (points) and fitted curves (solid lines) linear assumption (dotted lines)).

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 10. 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 10: analysis of radioactive tracers concentration in left and right containers for the diffusion cell D2

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

4.2.2.1 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. 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, Figure 24. 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%

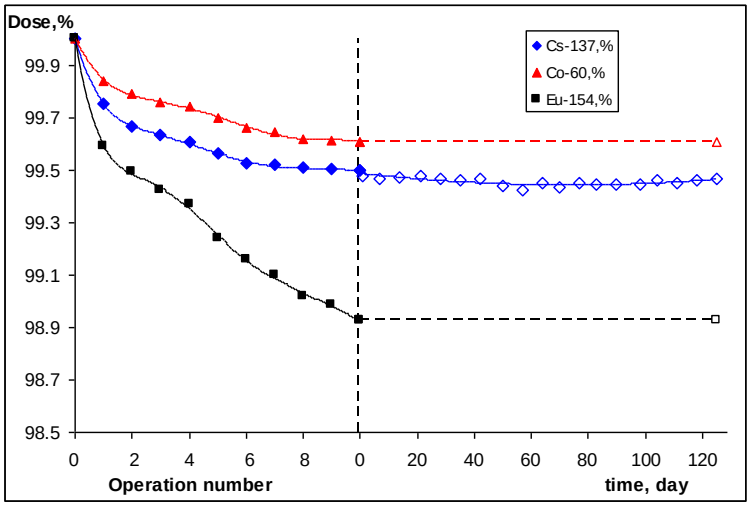


Figure 67: ^{60}Co, ^{137}Cs and ^{154}Eu radionuclide contents in AVR-2 sample depending on the wash diffusion experiment.

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 11:

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

Table11: radioactive tracers equal detection level

4.2.2.2 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 25. 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}Eu = 14.3% > ^{60}Co = 7.9% ^{137} > Cs = 6.8%

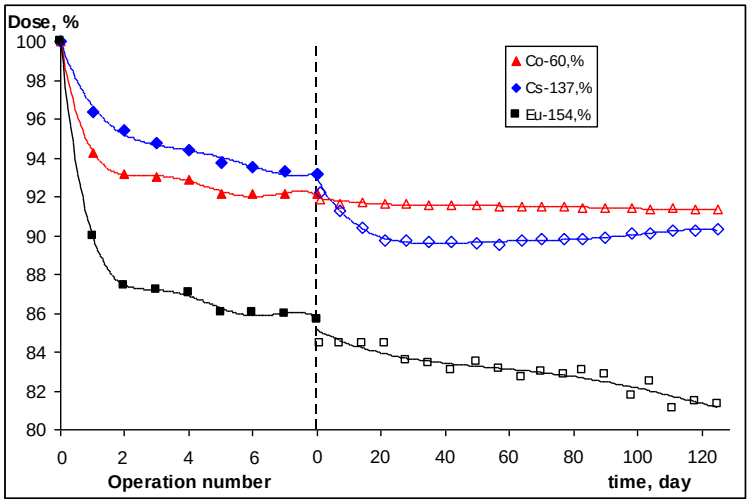


Figure 7: ^{60}Co, ^{137}Cs and ^{154}Eu radionuclide contents in AVR-1 sample depending on the washed operation number and time of diffusion experiment.

During the following diffusion experiment, the 60Co 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, shown at Figure 28. After the diffusion experiment during 125 days, the order of

the 154Eu, ^{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 10.

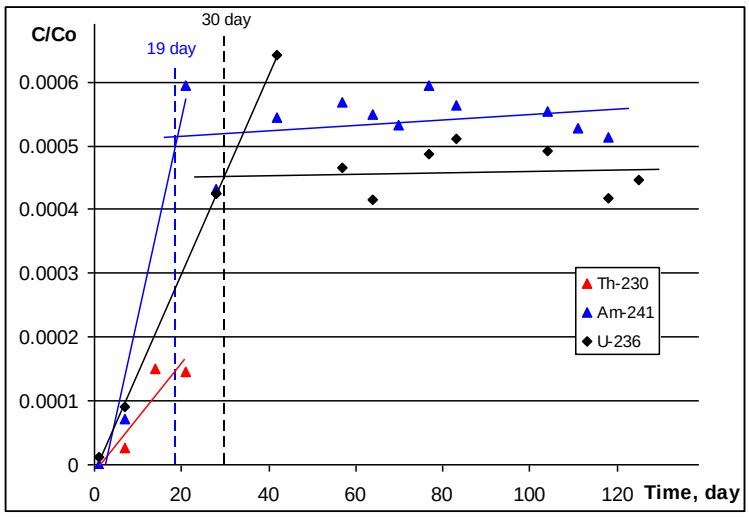


Figure 8: Through-diffusion of ^{241}Am, ^{236}U (b) and ^{230}Th (d) tracers in diffusion cells AVR-1.

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 12: 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}Am > ^{236}U > ^{230}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 13.

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

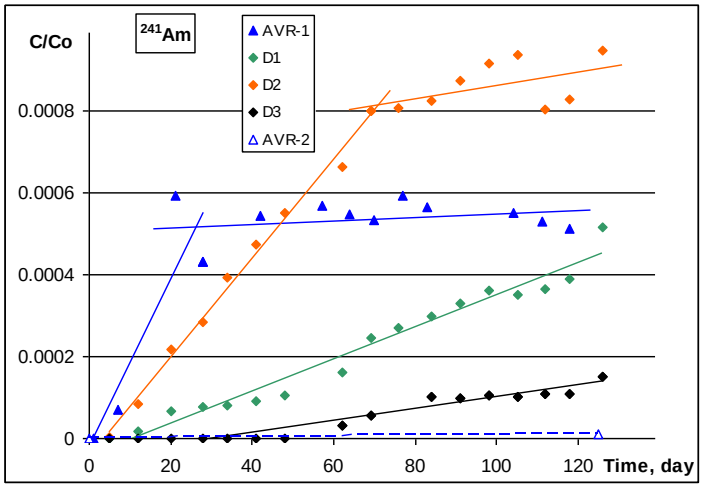
Table 13: relation between the effective diffusion coefficients De^{Th230}/De^{Am241} and De^{U236}/De^{Am241}

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:

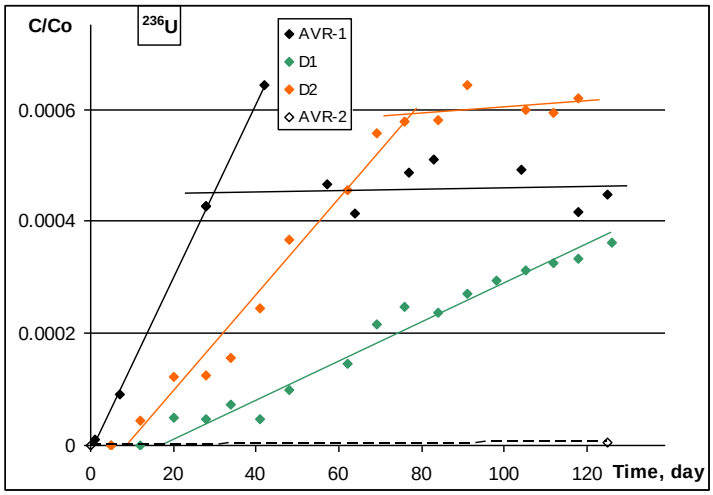
AVR-1 > D2 > D1 > D3 > AVR-2

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

a)



b)



c)

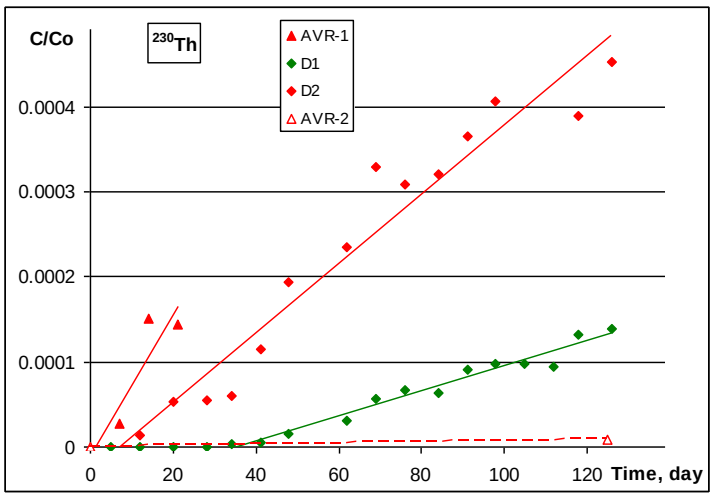


Figure 9: Through-diffusion of ^{241}Am (a), ^{236}U (b) and ^{230}Th (c) tracers for all investigated samples.

4.3 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 14.

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 14: 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:

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

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

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

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6 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