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STUDY OF THE RELATION BETWEEN TREATMENT AND DISPOSAL ON THE PERFORMANCE OF RBMK-1500 GRAPHITE IN CRYSTALLINE ROCK

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STUDY OF THE RELATION BETWEEN TREATMENT AND DISPOSAL ON THE PERFORMANCE OF RBMK-1500 GRAPHITE IN CRYSTALLINE ROCK

Executive summary

The main task for LEI was to study the relation between treatment and disposal on the performance of RBMK-1500 graphite in crystalline rock. The source term was based on the analysis performed by LEI within the WP3. Different treatment options may lead to removal of more labile part of C-14 inventory during the process. Due to lack of empirical data the illustrative rates were selected in this study to represent possible differences on non-treated/treated graphite for the analysis of C-14 migration from the graphite disposed of and to evaluate the impact of these differences on the radionuclide flux to geosphere. The importance of waste leaching rate was analyzed within the context of different performance of engineered barrier considering 3 different cases each supplemented by a number of variants. Modeling of the radionuclide migration within the near field was performed using AMBER code (UK). Model for the radionuclide transport through the geological formations was developed with TOUGH2 (USA) computer code. Modeling was made for two options, using the reference near field model (for non-encapsulated waste) and alternative near field model (considering possible encapsulation). Modeling results were discussed and summarized.

Revisions

Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
00	dd/mm/yyyy	Issue	Name, Organisation *Signature*	Name, Organisation *Signature*	Name, Organisation *Signature*	Name, Organisation *Signature*
01	23/05/2012	First issue	Poskas LEI	Poskas LEI		Grambow Subatech
02	25/03/2013	Final issue	Poskas LEI	Poskas LEI		Grambow Subatech

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1 Introduction

LEI team is involved in the **Task 6.4: Assessment of waste performance** of WP6. The main task for LEI is to study the relation between treatment and disposal on the performance of RBMK-1500 graphite in crystalline rock. This will contribute to the analysis of the performance of waste disposal concept 1) from the point of view of a near field/waste package model and 2) from the point of view of an overall performance evaluation.

For the evaluation of the performance of repository containing irradiated graphite the radionuclide transport models have to be developed and the migration analysis has to be performed. Usually the analysis of the radionuclide transport in the near field, far field and biosphere is performed by the means of mentioned models.

2 Methodology

In order to perform this task the work was organized in several stages as summarized in Fig. 1.

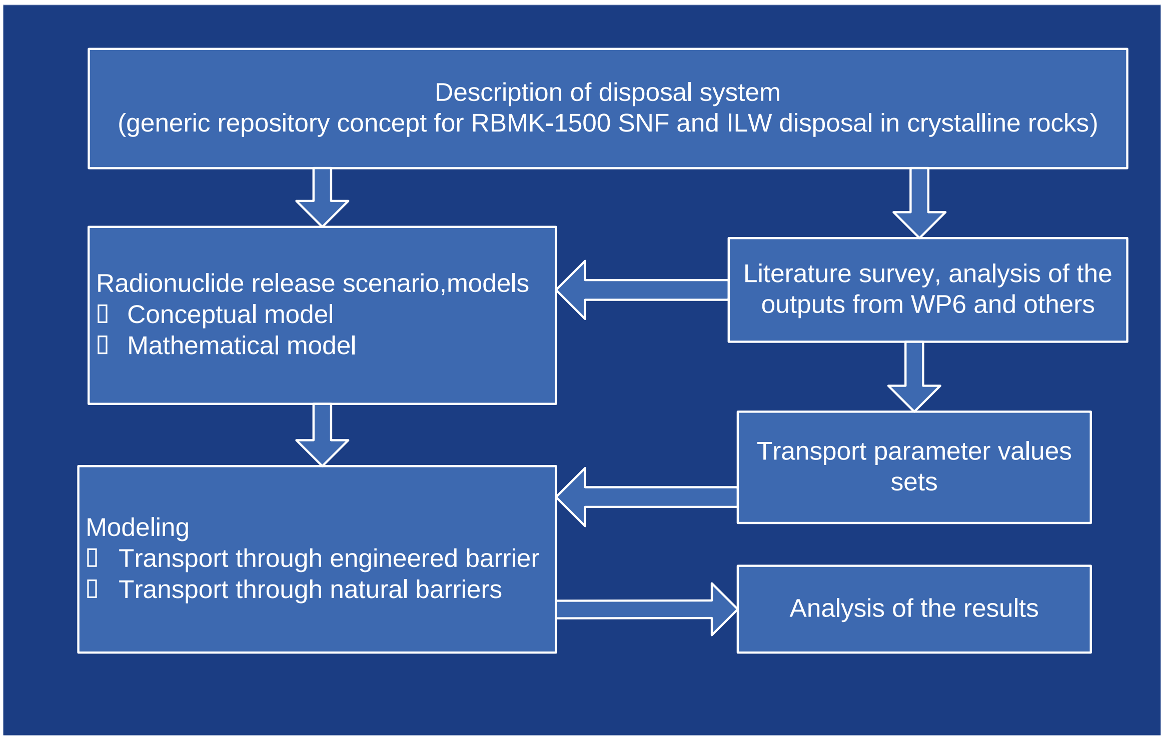

Fig. 1 Stages of the LEI work within task 6.4

For the development of the conceptual and mathematical model for the assessment of the radionuclide release from the irradiated graphite and subsequent transport from the geological repository the information on the repository concept, engineered and natural components and their properties is required as well as data on the various physical/chemical processes determining the release of the radioactive nuclides and further dispersion in the surrounding environment. Thus the following sections summarizes the available information based on the Lithuanian plans on the radioactive waste disposal and the current status in this field; and the available outcomes from the other participants of the WP6 were considered and are mentioned shortly as well.

3 Development of radionuclide transport models from the RBMK-1500 graphite disposed of in the crystalline rocks

3.1 Repository concept

There is no final decision on the long-lived intermediate level waste (ILW) disposal option or disposal container in Lithuania. Such ILW (including spent graphite) are planned to be stored in the steel containers at interim storage facility until the final decision will be made [1]. According to proposed generic repository concept of RBMK-1500 spent nuclear fuel (SNF) disposal in the crystalline rocks in Lithuania, the long-lived intermediate level waste (ILW) could be disposed at the same repository at certain distance from SNF emplacement tunnels [2]. The ILW emplacement tunnels could be app. 16 × 16 m in cross-section. Within this study the cementitious grout (NRVB backfill) was assumed as proposed in NIREX concept (United Kingdom) to be fill void regions within the tunnels after the emplacement of the ILW.

3.1.1 RBMK-1500 graphite

The origin of graphite waste is the reactor core elements from Lithuanian nuclear power plant at Ignalina site. Two units of Ignalina NPP were equipped with RBMK type reactors containing a graphite as a moderator and reflector. This means that after the dismantling of both reactor cores app. 3800 tones of graphite blocks and sleeves will be accumulated [1]. During the operation of NPP this material has been exposed to the neutron fluxes and the radioactivity has been induced.

For the subsequent safe and effective management and disposal of this type of radioactive material detailed investigations of its radiological characteristics are being carried out. The results of the research performed by LEI within the WP3 forms a basis for the radionuclide transport analysis from RBMK-1500 type spent graphite (source activity).

The graphite blocks are of GR-280 type and the graphite sleeves are of GRP-2-125 type. The activity depends on the neutron flux (location in the reactor core), operating power history, initial concentration of impurities, amount of cooling gases, etc. Based on the results of WP3 [3] and taking into account maximal initial nitrogen impurities in graphite matrix and nitrogen present in all (open and closed) graphite pores, the maximum activity of GR-280 type graphite (blocks) was used in radionuclide migration modeling (activity of blocks is higher than of graphite sleeves). The amount of graphite waste coming from the Ignalina NPP Unit 1 ad Unit 2 has been taken into account. Material properties used in the modelling are summarized in the Table 1.

Table 1. Material properties of RBMK-1500 reactor graphite components

Graphite type	**Bulk density (kg/m^3)**	**Porosity (open + closed) (%)**	**Mass (tones)**
GR-250 (blocks)	1744 [4]	22.8 (17 % open pores and 6 % closed pores) [4]	~3800
GRP-2-125 (rings/sleeves)	1850	16 (14 % open pores and 2 % closed pores) [5]	

3.1.2 Waste package

According to proposed generic repository concept of RBMK-1500 spent nuclear fuel (SNF) disposal in the crystalline rocks in Lithuania a metal containers could be used for the disposal of ILW. Report [6] presents the information on the UK plans for packaging irradiated graphite. At this stage of investigation the dimension of the waste package has not been developed in Lithuania yet, thus for this modelling study the Generic Specifications [7] developed in UK were analyzed and the information on the waste package was used in terms of geometrical data. The height of the metal container is 2.2 m, the length is 4.013 m, the width is 2.238 m (Fig. 2) based on [7, 8]. For the possible waste grouting a cementitious material (encapsulant) was assumed. Its thickness inside the container was considered to be 0.15 m. Total internal volume of the container of these dimensions is 18.9 m^3 based on [8]. Taking into account the packing efficiency of 0.67 [8] and the amount of RBMK-1500 graphite from both units of Ignalina NPP the total amount of containers to be required is app. 173 with total internal volume to be occupied by waste 12.7 m^3.

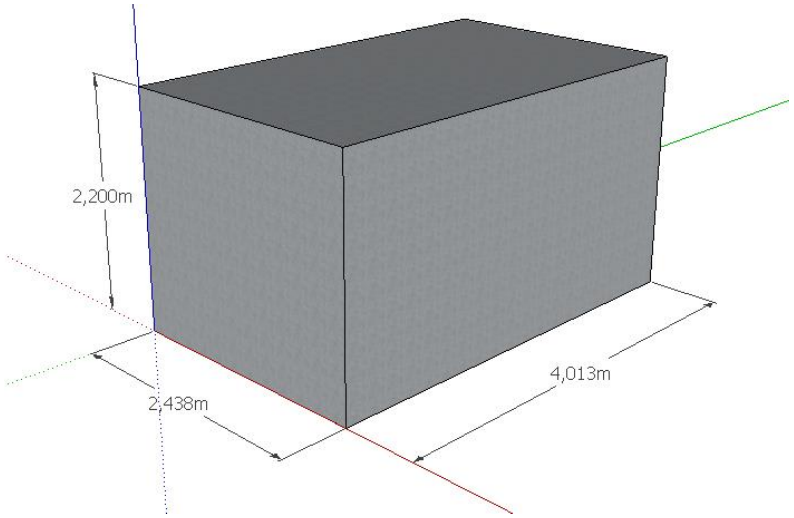

Fig. 2 Steel container considered for the RBMK-1500 graphite waste in this analysis

3.1.3 Disposal tunnel

The disposal tunnels will be constructed in the hard host rock (crystalline rocks). The orientation of the tunnels would be based on the stress analysis in the field and preferential groundwater movement. The tunnels will be oriented in such way that it would lead the radionuclides released to be transported along the disposal tunnel. Containers stack is of app. 10 m height and of 13 m width. Waste containers would be emplaced in these tunnels with a spacing of 0.9 m. Within the tunnel of 16 m x 16 m dimensions 4 containers in horizontal and vertical directions could be stacked. The upper part of the tunnel has to be sufficient for the container handling equipment (Fig. 3).

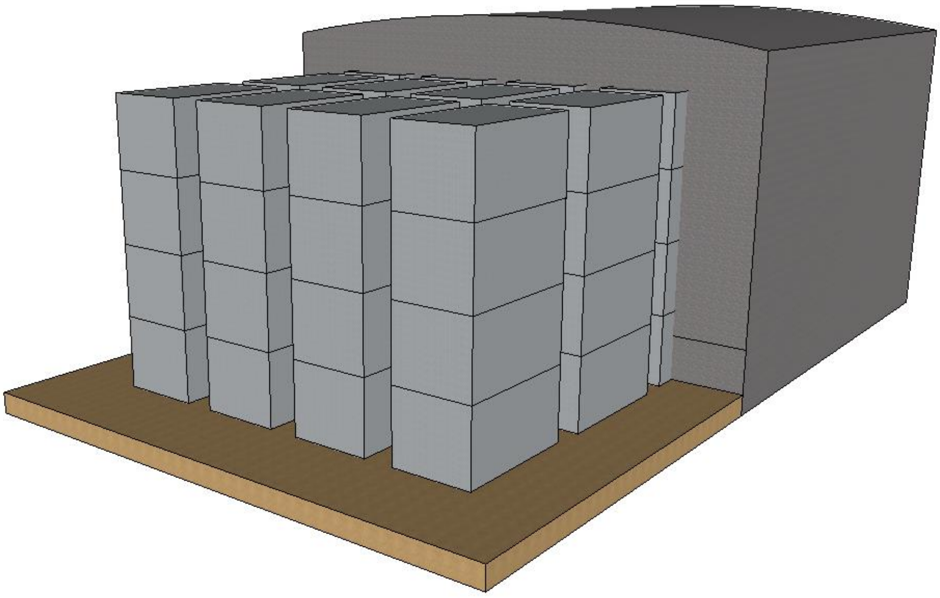

Fig. 3 Graphite waste disposal tunnel considered for the modelling

Taking into account the approximate number of the containers required and scheme of possible emplacement the length of disposal tunnel for the RBMK-1500 graphite disposal would be app. 50-60 m (stacked waste). After the waste emplacement tunnels will be backfilled with selected material. Based on the proposed generic repository in Lithuania the cementitious backfill (NIREX Reference Vault Backfill NRVB) could be used for the filling disposal tunnels after the waste emplacement. This material would stabilize the waste stacks within the vaults and, most importantly, would chemically condition the waste packages and any inflowing groundwater. NRVB is composed of a mixture of ordinary Portland cement (OPC), limestone flour, hydrated lime and water [10]. As indicated in [10] this produces:

- a very porous cement that promotes homogeneity and allows gas migration;
- high alkalinity for long-lived chemical conditioning;
- low bleed and high fluidity for good void filling;
- absence of organic additives to avoid the formation of metal-organic complexes (which would increase the solubility of some radionuclides); and,
- relatively low strength (compared to most cements) to aid retrievability of the waste packages, if required.

In this study cement as an encapsulant is considered in the analysis (for the alternative near field model development). Properties of backfill and encapsulant material are summarized in Table 2.

Table 2. Material properties of backfill and encapsulant material used in the modelling

Material	Bulk density (kg/m^3) [8]	Darsi veolocity (m/s) [8]	Porosity (m^3/m^3) [8]
Backfill (cementitious material)	1730	$6 \cdot 10^{-10}$	0.55
Encapsulant (cement)	2100	$1 \cdot 10^{-11}$	0.125

3.2 Geosphere

In case of RBMK-1500 graphite disposal in the crystalline rocks in Lithuania the far field will be constituted by the crystalline rocks and the cover of sedimentary rocks. Hydrogeological cross-section of Lithuania is presented in Fig. 4 .

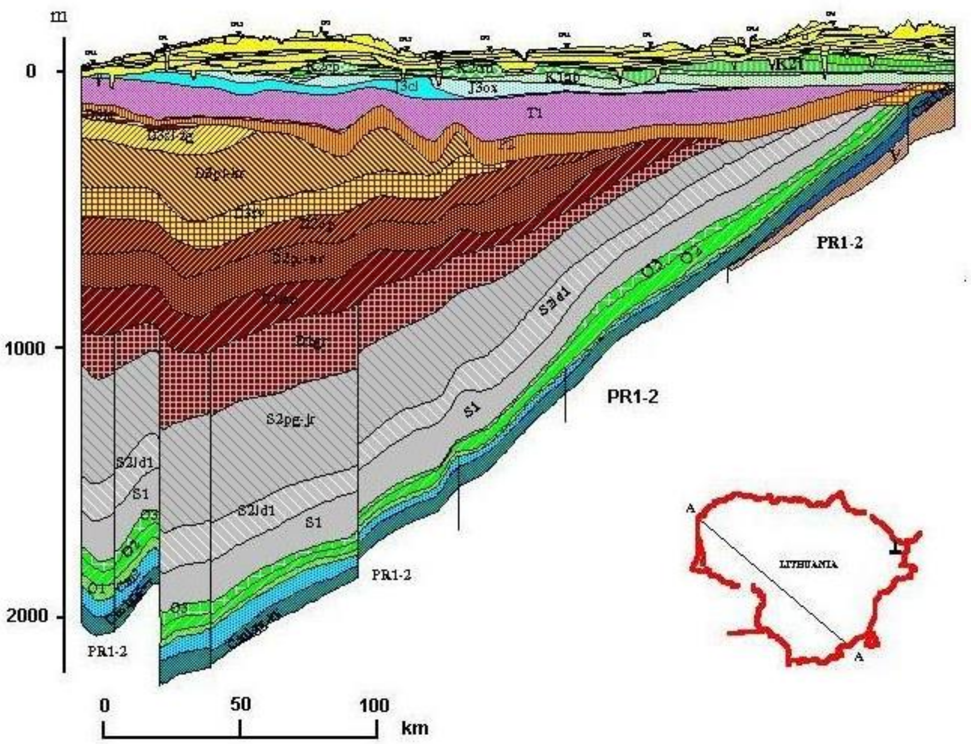

Fig. 4 Hydrogeological cross-section of Lithuania; crystalline basement is marked as “PR1-2” (Authors: L. Kilda, S. Šliaupa, J. Lazauskienė)

As there are no outcrops of the crystalline basement in Lithuania it will be difficult to find crystalline rocks at the depth smaller than 200–300 m. In the southern Lithuania crystalline rocks occur at the depths ranging from 210 m to 700 m, while in most of Lithuania territory the depth of the basement exceeds 700 m, reaching 2300 m in the west [2]. The crystalline rocks underlie the sedimentary rocks of different hydrogeological properties (forming aquifers and aquitards).

3.3 Radionuclide release from repository containing irradiated graphite

^{14}C is a key activation product within the graphite. The activation products in the graphite, such as ^{36}Cl, ^{59}Ni etc. present in the graphite waste as a result of impurities’ activation existing in the graphite structure. ^{14}C may be formed due to the activation of raw graphite material – carbon, impurities activation in the graphite matrix and within the pore space, and due to the activation of reactor coolant gases (such as N_2). Additionally, graphite may be contaminated due to the incidents during reactor operation, etc. Thus the inventory of radionuclide depends on the impurity concentration (defined by graphite manufacturing), operating conditions within reactor core, and subsequent storage and treatment technologies applied after reactor dismantling.

For the radionuclide release by leaching, the waste form and pore space within it has to come into the contact with water. Water penetration (saturation, pore space filling with water) depends on the graphite structure, porosity, pore size, etc., which tend to be changed during the irradiation in the reactor core. Water impregnation (pore space filling with water) in irradiated graphite is the first process that leads to the release of radionuclides contained in the graphite. If this phase is slow and low, it can control both the kinetics and the release rate of radionuclides in solution. Based on the experimentally obtained results (within WP6 of this project) with non irradiated and radiated graphite from G2 and St Laurent A2 UNGG reactors [11], the effect of irradiation on the water impregnation of graphite, which tends to increase the impregnation kinetics and the saturation rate of the graphite, was revealed. It suggests the assumption that the pores of irradiated graphite potentially will be saturated with groundwater for the subsequent release.

Carbon release

Radionuclides absorbed onto the material surfaces and pores and associated with impurities existing in the pores are able to dissolve in the groundwater flowing through the repository. The other part of inventory is incorporated in the crystal lattice and is much less available, if at all [8]. Performed leaching tests with graphite samples from different French reactors showed small release of ^{14}C whatever the sample origin, but the initial activities are far more important than ^{36}Cl. Leaching rates depend neither on the temperature nor on the leaching medium [12]. Reported ^{14}C dissolution rate was derived from G2 experiments is 3.10^{-7} $g \cdot m^{-2} \cdot j^{-1}$ [12]. In the [8] the empirically derived ^{14}C leaching rate from the graphite is used ($1.83 \cdot 10^{-5}$ 1/yr). The release rate at which the radionuclide is being release by leaching in groundwater depends on the waste matrix itself and whether or not the waste form has been pre-treated [9]. Some fractional release rates are indicated in the [9] for non treated waste and three rates for treated waste in different form. These rates are indicated as being illustrative ones rather than the demonstrative due to lack of empirical data [9], but could reflect a range of possible effectiveness of the waste form (Table 3). The main difference in the rates possible representing different state of graphite waste (treated, non treated) is expected to be related to the fast initial release within short time period. During treatment process this more labile fraction of the inventory is expected to be removed and the leaching rate would be lower and possibly constant.

Table 3. Possible ^{14}C leaching rate from graphite of different form [9]

Pre-treated?	Waste form	Release rate (1/yr)	
		First 10 yr	**10-10^6 yr**
N	Granular (i.e. something intermediate between block and powder)	$1\cdot10^{-1}$	$1\cdot10^{-2}$
N	Block	$1\cdot10^{-1}$	$1\cdot10^{-3}$
Y	Pulverised	$1\cdot10^{-1}$	
Y	Granular (i.e. something intermediate between block and powder)	$1\cdot10^{-2}$	
Y	Block	$1\cdot10^{-3}$	

^{14}C can also be released from the graphite as radio-labelled gases [8]. Gas phase of ^{14}C may be available for release at rates greater than would normally be experienced through the liquid (groundwater) pathway [13]. If a significant amount of gaseous ^{14}C activity is released through the air pathway that release will deplete the inventory of ^{14}C available for release through the groundwater pathway. This scenario could have an important impact on the source term [13].

Based on the experimental results mentioned in [8], only small release of ^{14}C was measured from a sample of crushed graphite immersed in the aqueous alkaline solution. The further work carried out with ^{14}C release from the intact and crushed graphite showed that the majority of ^{14}C remains in graphite., but there is some release of ^{14}C (probably carbonate) into the aqueous phase and a small early release (thought to be methane) into the gas phase. The fraction of the ^{14}C released from the graphite that was methane (organic form) is probably less than 0.01. In [14] the results of radionuclide immobilization in irradiated RBMK graphite blocks are reported. Leaching tests were performed for RBMK graphite GR-280 samples cured in fixative “atomik”. Leaching tests indicated no radionuclides release into the distilled water within 2 years period. Leaching tests were carried out for several samples without fixative however the radioactivity of radionuclides Cs-137, Cs-134, Co-60 and total activity of nuclides released in solution was measured only. Results of research on ^{14}C, Cl-36 leaching rates from RBMK graphite were not available during this study.

Chlorine release

As indicated in [15] the experimental tests were carried out with graphite samples from UNGG G2 reactor. The results showed that the leaching rate of ^{36}Cl depends on the position of the sample studied in the reactor at nuclear power plant and particularly the temperature of the graphite during reactor operation. The release kinetics of the labile fraction of ^{36}Cl in solution can be described by a diffusion process through the graphite porosity. Two chemical forms of chlorine can be found in solution: chlorides for the most part and chlorites. Concerning ^{36}Cl it was reported in [12] that its release fraction in aqueous solution range from 10 to 90 % depending on the thermal operating conditions. The strong influence of the sample dimensions (mass and V/S ratio) on ^{36}Cl leaching rate was indicated as well. Based on the results obtained it is suggested that two forms of ^{36}Cl may be present within graphite: a labile fraction which is weakly bound to graphite, and a chemical form is less labile or whose localization within graphite porosity is less. Water penetration into graphite, chlorine solubilisation, chlorine leaching kinetics are the issues which might control and restrain the release of ^{36}Cl and thus need to be investigated. Thus in case of underwater dismantling the large part of chlorine may be trapped into the ion exchange resins. In case of dismantling in dry conditions the chlorine inventory release will occur as the groundwater penetrate the waste package. In the reference [8] a fractional release rate of $6.3 \cdot 10^{-2}$ 1/yr for ^{36}Cl is accepted for the radionuclide migration analysis from the geological repository.

The solubility of the particular species of radionuclide (such as ^{36}Cl) under certain chemical conditions could results in the formation of some insoluble form and subsequently controls its release to the environment. Thus in case of fast release of chlorine from graphite the proper consideration of such phenomena could provide a benefit for the reduction of its radioactivity flux to the environment.

Retention of radionuclides

In the report [17] the retention properties of particular cement paste (constitutive of concrete that could be used for i-carbonaceous waste packages) was investigated. ^{36}Cl retention properties depend on the alteration state of the cement paste and also on saturation effect generated by stable chloride ions provided by several sources in a repository (the groundwater, the concrete itself).

As it is reported in [17] wet chemistry measurements show that distribution ratios (R_d) values slowly increase during the first 20 days of contact time, and then, whatever the case, R_d reaches a steady mean value. R_d values are relatively low. The maximum R_d value (35 ml/g) was measured for the degraded state and at a low chloride concentration (4.8×10^{-5} mol/l). The results obtained showed that retention of ^{36}Cl strongly depends on the stable chloride concentration in solution. At high chloride concentration, the saturation effect is observed (non-linear sorption isotherm). Thus the importance of knowledge and quantification of all main sources of stable chloride in order to be able to carry out performance assessment was highlighted.

In [18] the compiled database of the analyzed data on diffusivity, sorption, porosity, density of various materials considered to be used in the repository for low and intermediate level waste in Sweden (SFR-1) is presented. As it is reported in [4], the cement phases which dominate sorption are CSH phases which are common to all types of cement. Thus, identical K_d values are provided for sorption on different cements/concrete, so that differences in radionuclide transport through different cementitious materials will be governed only by differences in physical parameters (tortuosity, constrictivity, porosity; these parameters expressed through diffusivity) for each cement/concrete. Here the sorption coefficient of inorganic ^{14}C on concrete/cementitious materials is reported to be 0.5 m^3/kg (best estimate) (fresh concrete) and 0.01 (pessimistic value). For degraded concrete 0.01 m^3/kg is reported as the best estimate with K_d=0 m^3/kg [18]. The sorption of ^{14}C in NIREX cementitious backfill (NRVB backfill) is reported to be 0.2 m^3/kg [8]. For this study the value reported for NRVB was used (and K_d=0 m^3/kg) as it is in line with data reported in [18]

Radionuclide transport

The transport of radionuclides released from the waste matrix will be determined by the hydrogeological conditions in the surrounding environment and could be dominated by advective of diffusive transfer (or both) accompanied with the interaction with material. Cementitious backfill is projected to be very porous cement that promotes homogeneity and allows gas migration. In this material advective flow is expected to be dominated transport mechanism when the radionuclides are transferred from the waste package.

Flow and transport in a fractured medium, such as crystalline rock, mostly takes place in the fractures of the rock mass. The permeability of the rock blocks in between fractures is sufficiently small to give an advective flow within the rock blocks that is negligible in most situations. The fractures and joints only make up a very small fraction of the total volume (flow porosity do not exceeds 2 % and usually it is only parts of the percent) and thus the flow capacity of the rock can be very limited. Furthermore, in rocks with a sparse network of fractures the probability of fractures intersecting is low and as a consequence, the hydraulic conductivity of such rocks is generally small. However, limited number of continuous flow paths extending over long distances may exist in the rock. Since the volume of the fractures where flow takes place is limited, this often gives rise to relatively high velocities in the continuous flow paths available, typically in the order of meters per year. The fraction of the rock volume that is occupied by such channels is defined as flow porosity. During transport most solute species interact with the fracture surfaces through physical and chemical processes. These processes can cause retardation, i.e. delay the release of radionuclides from the geosphere, or retention of radionuclides from being released from the geosphere. If the contaminant can penetrate into the rock matrix by diffusion (matrix diffusion) they gain access to the very large internal surfaces in the pore space of the rock. Chemical processes such as sorption and precipitation can immobilize the solutes causing a large long-term retention of radionuclides in the geosphere. The retention capacity of the rock is thus large enough to cause a significant reduction in radionuclide concentration as well as retardation of radionuclides in time. For radionuclides with a half-life shorter than, or comparable to, the transport time through the rock, the retardation of the release is large enough for radioactive decay to result in a complete retention.

The possibility of ^{14}C occurrence and transport out of repository in gaseous form has already been under discussion widely. It has been pointed out the possibility of faster its transport through a geosphere to the biosphere and subsequent radiological impact due to its release inside the buildings, uptake in plants and resulting human exposure.

As indicated in [19] unlike many other radionuclides, ^{14}C (e.g., as carbon dioxide or the bicarbonate ion) can be highly mobile in many geological environments and its mobility is strongly correlated with the media through its complex aspects such as chemistry and hydrogeology. pH is one of the key variables that largely control the speciation and mobility of carbon in aqueous environments and the general major element chemistry of the pore water may also play important role in controlling the ^{14}C mobility within the media. The amount of ^{14}C that will be released will be also highly dependent on the flow regime in which the wastes reside.

For the analysis of ^{14}C labelled gases transport from the repository it is essential to know the amount of ^{14}C released in gaseous form and its release rate. Based on [9] there is no evidence to suggest that more than 1 % of the ^{14}C inventory would be released in a gaseous form. Furthermore, given the stable, refractory nature of graphite, it is totally implausible that a large percentage of the mass of the graphite would be released as gas on any reasonable timescale [9]. It seems plausible that up to 1 % of the ^{14}C inventory in the graphite might be released as either carbon dioxide or methane. It is anticipated that $^{14}CO_2$ would be less mobile than $^{14}CH_4$ in any cementitious materials used for any engineered barriers system (EBS) structures as CO_2 is likely to form carbonates and therefore be significantly retarded.

Besides the ^{14}C in gaseous form from graphite, there are other sources in the repository. The sources of ^{14}C labelled gases are the degradation of organic materials and from metal corrosion [20] if the intermediate level waste is going to be disposed together with graphite. Thus in order to evaluate the total amount of gases produced in the repository all sources have to be accounted properly.

3.4 Modelling of radionuclide release through the EBS

Based on these data and RBMK-1500 graphite radionuclide inventory [3], the models had been developed and numerical modelling of ^{14}C radionuclide transport in the near field was performed. The computer code AMBER [22] has been used for the modelling.

3.4.1 Conceptual model

Main premises assumed for the modelling are as follows:

- No credit is taken for the integrity of metallic container (container do not provide an additional barrier for the transport and is totally corroded soon after the repository closure);
- Radionuclide are released from the waste matrix in inorganic form and interact with the groundwater percolating the waste matrix;
- Dissolved radionuclides are transported by diffusion and advection (in the encapsulant material) and mainly by advection through the backfill of disposal tunnel up to the fracture intersecting the disposal tunnel;
- Chemical conditions within the encapsulant and backfill may limit the amount of dissolved radionuclides if the concentration in porewater exceeds the solubility limit;
- The interaction mechanisms of dissolved radiocarbon with encapsulant and backfill material are accounted by the linear sorption coefficient (or omitted sorption as analysis variant);
- During the transport of radionuclides through the EBS the radioactive decay occurs and is taken into the consideration.

As there is no site specific data on fracture spacing in Lithuanian hard rocks yet, fracture spacing assumed to be 50 m based on [8]. This will constitute the average length of radionuclide transport in the disposal tunnel prior its release to geosphere by the flowing water in the fracture (Fig. 5). Due to repository construction in the hard rock and the emplacement of engineered material in the disposal tunnel the flow enhancement could take place there. The flow increment factor of 30 has been taken into account base on [8] when evaluating the advective flow through the disposal tunnel.

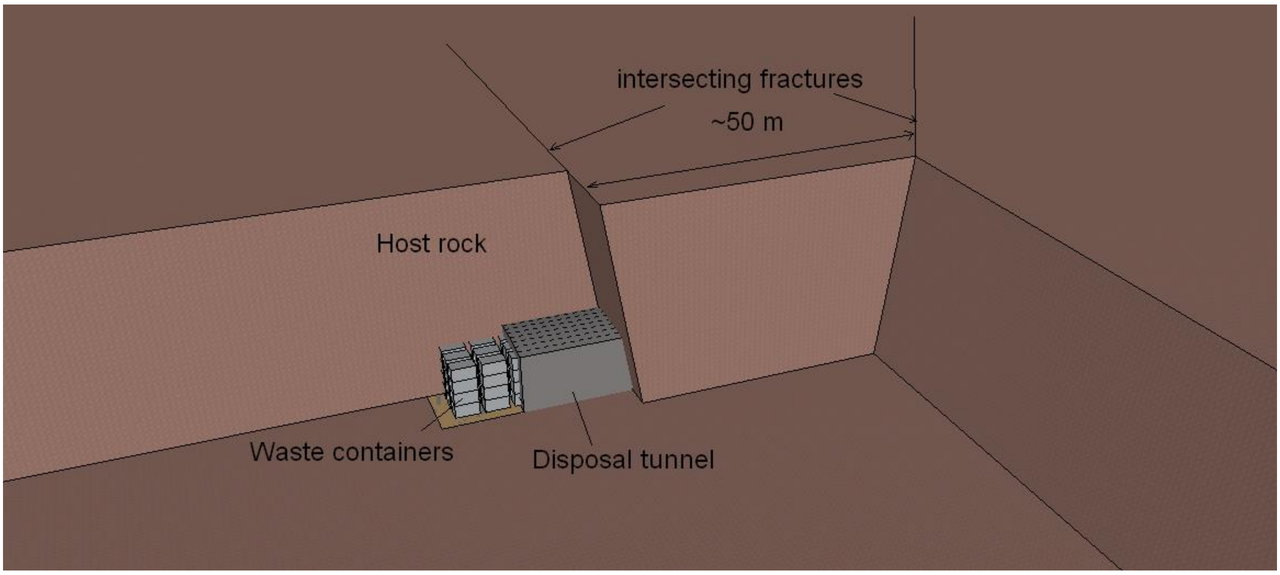

Fig. 5 Scheme of disposal tunnel and intersecting fractures

Radionuclide transport analysis is performed for the average container located in the disposal tunnel. Total flux of radioactivity to the geosphere will be based on the release from the average container and the number of containers required.

The main processes being considered in the near field release model are radionuclide release from the waste matrix (constant, time dependant), its solubility in the porewater of surrounding engineered barriers (encapsulant, backfill), advective and diffusive transport through the encapsulant, advective transport in backfill and sorption on different materials in the near field. No credit has been taken for the waste packages, i.e. it is assumed that the metal canister was corroded immediately. However according to [8] the container of such as dimensions (4 m box) could be collapsed after 240 000 years assuming the collapse as 80 % is corroded by uniform corrosion and corrosion rate of $1 \cdot 10^{-8}$ m/yr. In case of unbreached container the radionuclides migration takes place through the container vent only and is diffusion driven mainly. The presence of container as a physical barrier would result in a significant delay of groundwater flow through all the waste and the highest release significantly later.

Modelling was made for two basic cases, using the **reference near field model** (for non-encapsulated waste) and **alternative near field model** (considering possible encapsulation). As a result the radionuclide flux to the groundwater in the crystalline rocks was evaluated. For more detailed analysis of leaching mechanisms (rates) the analysis of ^{14}C release through the repository engineered barriers was divided into several cases considering the differences in the data on its leaching rates from the spent graphite:

Reference near field model (not encapsulated waste)	**Alternative near field model (encapsulated waste)**
Case 1: K_d=0, no solubility limitation	**Case 1: K_d=0, no solubility limitation**
Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D	Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D
Case 2: K_d=0, solubility limit=0.01 moles/m^3	**Case 2: K_d=0, solubility limit=0.01 moles/m^3**
Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D	Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D
Case 3: K_d=0. 2 m^3/kg, solubility limit=0.01 moles/m^3	**Case 3: K_d=0. 2 m^3/kg, solubility limit=0.01 moles/m^3**
Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D	Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite: A1, B1, B2, C1, C2, C3, D

During the modeling the data on leaching of RBMK type graphite were not available, thus a range of possible range of leaching rates were examined Leaching rates used in the simulations are of more indicative nature rather than the precise ones. Summarized data for different cases are presented in Table 4.

Table 4. Summarized data on possible ^{14}C leaching rates (assumed for the analysis)

Case 1: K_d =0, no solubility limitation		
Variants based on fractional leaching rate (1/yr) of ^{14}C from graphite:		
Variant A	$1.83\cdot10^{-5}$	Experimentally measured rate [9]
Variant B1	<10 yr - 0.1 >10 yr - 0.01	Corresponds to not treated waste, higher leaching rate [9]
Variant B2	<10 yr - 0.1 >10 yr - 0.001	Corresponds to not treated waste, lower leaching rate
Variant C1	0.1	Corresponds to treated waste, the highest leaching rate [9]
Variant C2,	0.01	Corresponds to treated waste, lower leaching rate [9]
Variant C3	0.001	Corresponds to treated waste, the lowest leaching rate [9]
Variant D	instant release from waste	Barrier function of solid waste matrix disregarded
Case 2: K_d=0, solubility limit=0.01 moles/m^3		
Variant A	$1.83\cdot10^{-5}$	Experimentally measured rate [9]
Variant B1	<10 yr - 0.1 >10 yr - 0.01	Corresponds to not treated waste, higher leaching rate [9]

Variant B2	<10 yr - 0.1 >10 yr - 0.001	Corresponds to not treated waste, lower leaching rate
Variant C1	0.1	Corresponds to treated waste, the highest leaching rate [9]
Variant C2,	0.01	Corresponds to treated waste, lower leaching rate [9]
Variant C3	0.001	Corresponds to treated waste, the lowest leaching rate [9]
Variant D	instant release from waste	Barrier function of solid waste matrix disregarded
Case 3: K_d =0.2 m3/kg, solubility limit=0.01 moles/m^3		
Variant A	$1.83\cdot10^{-5}$	Experimentally measured rate [9]
Variant B1	<10 yr - 0.1 >10 yr - 0.01	Corresponds to not treated waste, higher leaching rate [9]
Variant B2	<10 yr - 0.1 >10 yr - 0.001	Corresponds to not treated waste, lower leaching rate
Variant C1	0.1	Corresponds to treated waste, the highest leaching rate [9]
Variant C2,	0.01	Corresponds to treated waste, lower leaching rate [9]
Variant C3	0.001	Corresponds to treated waste, the lowest leaching rate [9]
Variant D	instant release from waste	Barrier function of solid waste matrix disregarded

In variants B1 and B2 leaching rates used corresponds to the possible release from the non treated graphite waste, in variants C1, C2, C3 leaching rates used corresponds to the possible release from the treated graphite waste. Variant D represent conservative assumption of 100 % available ^{14}C inventory to be released from the container.

In order to evaluate the importance of the radionuclide interaction with the backfill, the flux of radionuclide to the geosphere was assessed considering the sorption and disregarding this process (K_d=0 m^3/kg). Thus the flux estimated in this case will serve as a bounding value for radionuclide release from the EBS. The effective diffusion coefficient ($1\cdot10^{-11}$ m^2/s) reported for NIREX Reference Vault Backfill were assumed. However the lower coefficients of ^{14}C (of the order of 10^{-15}-10^{-13} m^2/s) diffusivity in cement are mentioned in [21], while in [18] for porous concrete range of $1\cdot10^{-10}$-$6\cdot10^{-10}$ m^2/s is reported. During the study the data on solubility limit determined by the geochemical conditions in the backfill saturated with the groundwater from Lithuanian crystalline rocks were not available. The solubility limit was assumed to be equal to that determined for UK conditions (0.01 mol/m^3) [8]. The impact of solubility process was analyzed with a modeling a case of radionuclide migration without taking into consideration the solubility limitation.

AMBER was used for modelling the radionuclide migration through the engineered barriers of the repository. The disposal system is divided into the compartments and the mass balance equations are being solved numerically by AMBER code:

$$\frac{dI_i^n}{dt} = -\left[\lambda_r^n + \sum_j TC_{ij}\right] I_i^n + \lambda_r^{n+1} I_i^{n+1} + \sum_j TC_{ji} I_j^n \,;$$

where I_i^n – amount of radionuclide *n* in *i*th compartment (moles), TC$_{ij}$ – transfer coefficient from compartment *i* to compartment *j* (1/yr), λ_r^n – radioactive decay constant of radionuclide *n* (1/yr), λ_r^{n+1}– radioactive decay constant of parent radionuclide (1/yr). Advective flow and diffusive transport are handled by the transfer coefficients.

3.5 Modelling of radionuclide migration in the geosphere

Porous medium (continuum approach) was applied for modelling the groundwater flow and contaminant migration in the larger scale model, comprising the first fractured natural barrier and the subsequent ones (crystalline and sedimentary rocks). In this case, the analysis was performed for the radionuclides released from the crystalline rocks and being transported upward to the groundwater discharge area. The conceptual model of the repository is based on the conservative assumption that the repository would be located in the area where the groundwater flow is upward, thus the distance to the surface is shorter than could be expected during the designing of real repository. Model was developed taking into account hydrogeological data of geological formations (thickness, hydraulic conductivity, porosity, etc., Table 5) in southern Lithuania [23] (Fig. 6).

Table 5. Hydrogeological data of natural barriers [23]

Layer no.	Depth (m)	Hydraulic conductivity (m/s)	Porosity (m^3/m^3)	Description
1	0–100	$1.2\cdot10^{-5}$	0.25	glacial loam
2	100–200	$2.3\cdot10^{-5}$	0.35	sand
3	200–250	$5.8\cdot10^{-5}$	0.5	anhydrite
4	250–270	$1\cdot10^{-6}$	0.5	limestone
5	270–290	$5.8\cdot10^{-5}$	0.05	sandstone

Layer no.	Depth (m)	Hydraulic conductivity (m/s)	Porosity (m^3/m^3)	Description
6	290–310	$1.16\cdot10^{-10}$	0.13	clay
7	310–410	$6\cdot10^{-6}$	0.05	sandstone
8	410–420	$1.16\cdot10^{-7}$	0.01	weathered crystalline rocks
9	420–520	$1.16\cdot10^{-13}$	0.0038	monolithic crystalline rocks

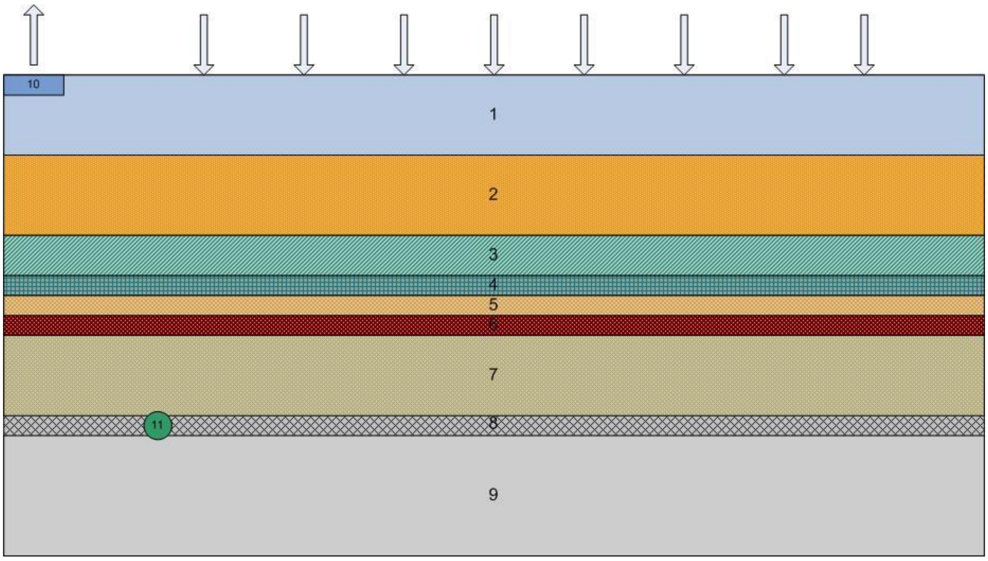

Fig. 6 Far field model (1 – glacial loam, 2 – sand, 3 – anhydrite, 4 – limestone, 5 – sandstone, 6 – clay, 7 – sandstone, 8 – weathered crystalline rocks, 9 – monolithic crystalline rocks, 10 – groundwater discharge area, 11 – radionuclide flux from the crystalline rock)

The results of ^{14}C release through the EBS in case of instant release from the graphite (not encapsulated waste, variant D) and with no retention in the backfill were used as an input data taking into the consideration the number of disposal containers.

Applying the continuum models, the groundwater flow in porous medium could be determined by solving the equation for the groundwater pressure variation within the system being modelled:

$$\frac{\partial}{\partial t}\phi S\rho = div\left[k\frac{k_r}{\mu}\rho\nabla(P+\rho gz)\right];$$

where ϕ – rock porosity (m^3/m^3); S – rock storage coefficient (1/m); ρ – groundwater density (kg/m^3); k – absolute permeability (m^2); k_r – relative permeability (-); μ – dynamic viscosity (kg/m·s); P – groundwater pressure ($kg/m\cdot s^2$).

After the assessment of groundwater flow rate, the contaminant distribution within the modelled system could be assessed by solving the equation:

$$\frac{\partial C^n}{\partial t} = D_M \nabla^2 C^n + v \nabla C^n - \lambda_r^n C^n + \lambda_r^{n+1} C^{n+1};$$

where C=C(x, y, z, t) – radionuclide concentration (kg/m^3); D_M – hydrodynamic dispersion coefficient (m^2/s); v – pore flow rate, determined dividing the Darsi flow rate by rock porosity (m/s), λ_r^{n+1}, λ_r^n – decay rates of parent and daughter nuclides (s^{-1}).

4 Results and discussion

4.1 Migration through the engineered barriers

4.1.1 Reference near field model

Radionuclide transport modelling was performed for the time period of up to 1 million years after the repository closure. Such a long time frame is typically considered analyzing the performance of deep geological repository. As it is presented in the following figures this time period is enough to assess the peak annual release which directly corresponds to the maximal exposure dose.

Case 1: K_d=0 m^3/kg, no solubility limitation

The ^{14}C flux to the geosphere is expressed in terms of Bq/yr per Bq of ^{14}C disposed in one container (fractional flux) and is presented in Fig. 7 . No encapsulation of waste is considered in this case.

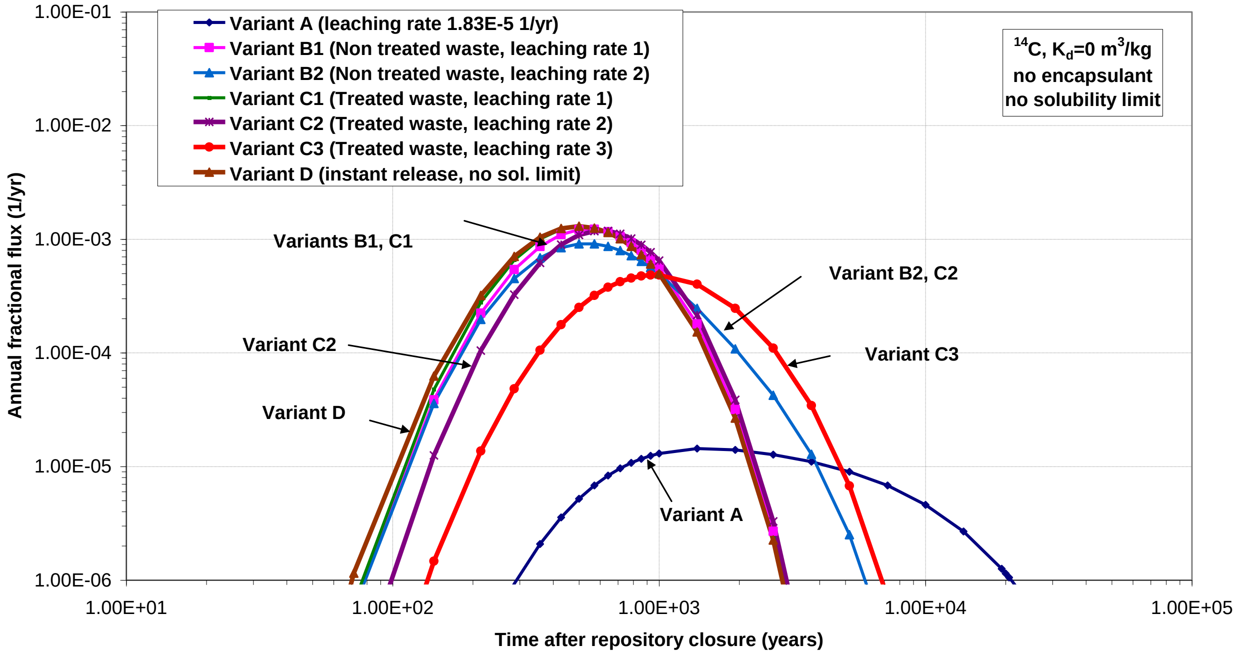

Fig. 7 Release of ^{14}C to the geosphere from one disposal container with non encapsulated waste

Modeling results of ^{14}C transport in case of no interaction with surrounding engineered barrier showed that the differences in leaching rate do not lead to the same differences in the peak flux to the geosphere. The peak fractional flux of ^{14}C dissolved in groundwater varies within app. 2 order of magnitude due to different leaching rates from the graphite waste. While the difference in the leaching rate could be up to app. 5 orders of magnitude (variants A and D (instant release)) (Table 4). As it could be expected the lowest peak flux is observed for variant A, due to the lowest leaching rate and lowest amount of radiocarbon released to the backfill. The main difference in the leaching rate for non-treated and treated waste form is related to the higher release rate for the first 10 years and is based on the experimental observations (more intensive release firstly and constant congruent release later). During the treatment procedures this fraction which releases more rapidly is going to be removed thus the leaching rate profile is proposed not to be time dependent. Based on the results in Fig. 7 , it could be seen that the radionuclides released from the graphite by the rates of the order of 10^{-2} – 10^{-1} 1/yr (variants B1, B2, C1, C2) were released to the geosphere at similar rate as in case of instant release. This suggests that radionuclide flux depend on the hydrodynamic conditions determining the groundwater flow and transport conditions in the backfill and not on the radionuclide flux coming from waste matrix determined by different leaching rates.

If the radionuclides were released at lower rates (or the order of 10^{-3}, 10^{-5} 1/yr for the inventory of this study) the flux to the geosphere becomes more dependent on the leaching rate.

While assessing of graphite suitability for geological disposal in wide range of systems the approach for barrier performance analysis was proposed in [25]. As presented in [25] a transfer rate could be derived that give rise to impacts around the regulatory guidance levels (17 μSv/yr) was determined for radionuclide inventory associated to UK graphite and using generic biosphere model. For UK inventory the transfer rate metric of $7 \cdot 10^{-6}$ 1/yr was determined for ^{14}C transport in liquid form. In case of no sorption and no solubility limitation the peak fractional flux (10^{-3} 1/yr) from the backfill with RBMK-1500 graphite disposed of is higher than this metric.

The scope of the radionuclide flux attenuation (how many time the flux to geosphere is lower if leaching at certain rate occurs vs. instant release from graphite matrix) is expressed as ratio of maximal fluxes $\left(\frac{Q_{\max}^{IRF}}{Q_{\max}^{Case\ X}} \right)$ and is named flux attenuation factor in the Fig. 8 . This factor indicates how many times the maximal flux determined in case of instant release would be decreased if the particular leaching rate would be demonstrated. The higher factor (>1) corresponds to the higher effectiveness of leaching rate of that order. The time of peak fractional release is observed after the shortest time past closure in case of instant release (variant D) ($\sim 5 \cdot 10^{2}$ yr). For the evaluation of the prolongation of time of maximal flux to geosphere when leaching at certain rate occurs vs. instant release from graphite matrix, the peak flux times were compared and are expressed as factor of peak flux time increase $\left(\frac{T(Q_{\max}^{Case\ X})}{T(Q_{\max}^{IRF})} \right)$. As this ratio is >1, the peak flux will be observed later, after the longer time period past closure. The comparison of maximal fluxes and its observation times is summarized in Fig. 8 .

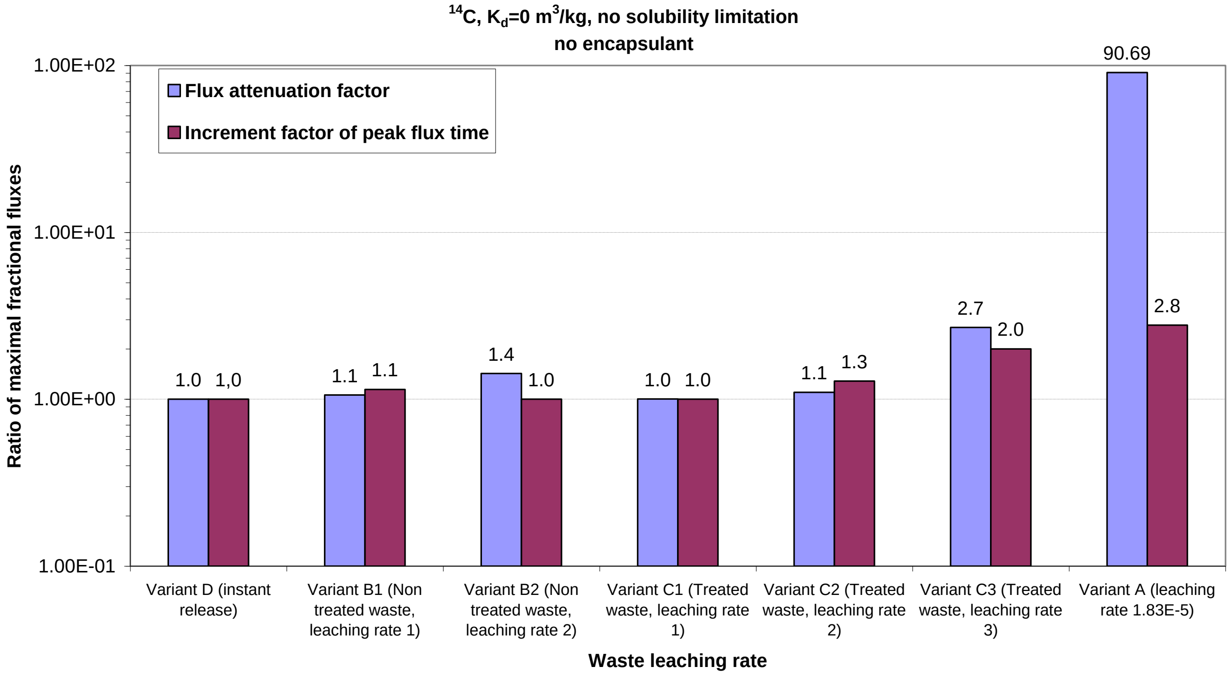

Fig. 8 Variations of peak fractional flux and time of maximal flux within the cases being analyzed

As it could be seen in Fig. 8 the most effective decrease of maximal flux is demonstrated if leaching rate is of the order of 10^{-5} 1/yr with particular radiocarbon inventory being analyzed. In this case the maximal flux to the geosphere would be decreased by a factor of ~91. If the leaching rate is of the order of 10^{-3} 1/yr, the fractional flux would be lower by a factor of ~3 (variant C3). If the flux coming from the inventory leaching at the rate 10^{-3} 1/yr follows the higher release within the short time period (variant B2) it contribute to maximal flux, thus flux attenuation factor becomes lower (1.4). Results for the inventory leaching at the rate of 10^{-2} 1/yr (variants B1, C2) are similar with time to peak occurring delayed for the later case. Based on the modeling results the reasoning of leaching rate of the order 10^{-1} 1/yr does not indicate any significant benefit. Time of peak flux is prolonged mostly in case of low leaching rate (variant A). In other cases it slightly increases (B2, C2, C3), while for the rest cases it is the same as in case of instant release.

Case 2: K_d=0 m^3/kg, solubility limit=0.01 moles/m^3

The results of ^{14}C flux to the geosphere out of EBS disregarding the sorption in the backfill material, but taking into consideration of limited solubility are presented in Fig. 9 .

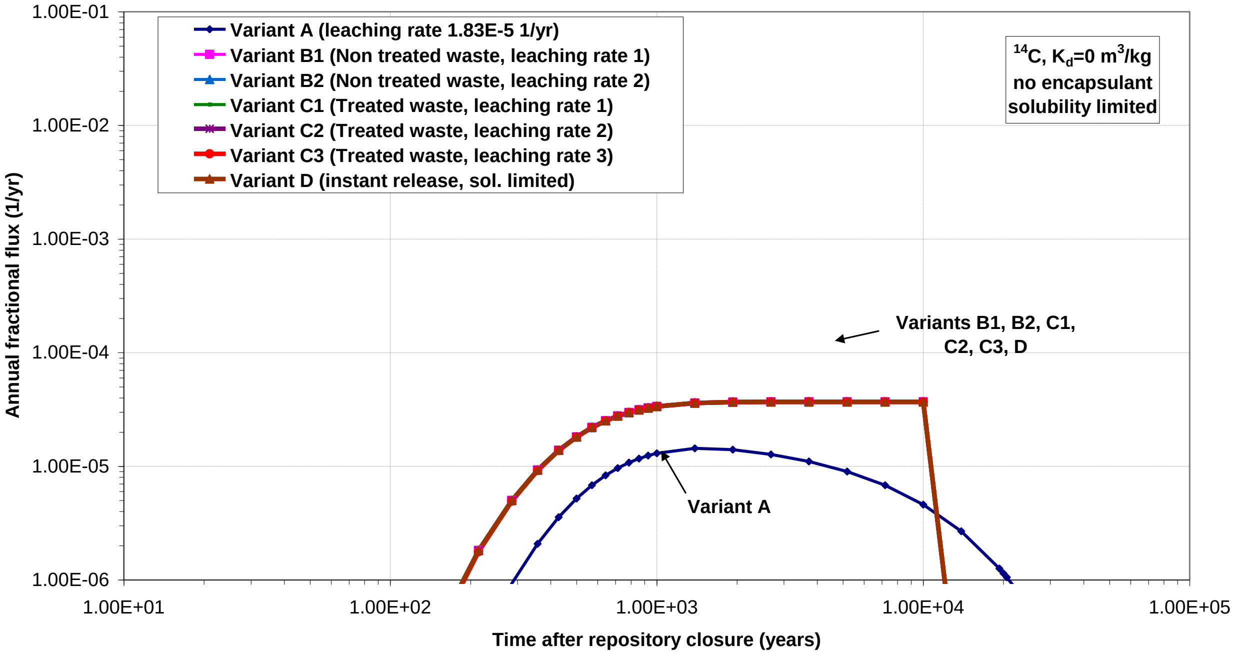

Fig. 9 Release of ^{14}C to the geosphere from one disposal container with non encapsulated waste

If reasonable estimates are available for the geochemical conditions in the near field and possible limitation of ^{14}C dissolution in the porewater of cementitious material (solubility limit of 0.01 moles/m^3 assumed), this would lead to the decrease of peak flux from the near field by app.one order of magnitude and diminished impact of different leaching rates on the flux to the geosphere. This is indicated by the plateau of the flux profile. For the variants analyzed the amount of ^{14}C released from RBMK graphite by the rate of 10^{-3}–10^{-1} 1/yr or released instantly is higher than maximum allowable concentration of ^{14}C dissolved in water. In case of ^{14}C released by the rate of 10^{-5} 1/yr all inventory released from the waste is allowed to dissolve. Whether or not the solubility limitation would have an impact on the flux to geosphere, highly dependent on the maximum concentration which is determined by the prevailing geochemical conditions and the flux from the waste, which is determined by the inventory and leaching rate. 10 times lower inventory and 10^{-4} 1/yr leaching rate would result in the similar flux to backfill as in variant A, thus subsequently the similar results on the geosphere flux. The comparison of ^{14}C peak flux to the results of Case 1 showed that due to limited solubility (at the limit of 0.01 mol/m^3) the peak flux decreased more than one order of magnitude besides the diminished differences if leaching rates. By comparing of peak fractional flux to transfer rate metric it could be seen that for the particular inventory and particular water flow and transport condition, the metric is still exceeded.

The comparison of maximal fractional fluxes and its observation times is done to that of observed in case of instant release and is summarized in Fig. 10.

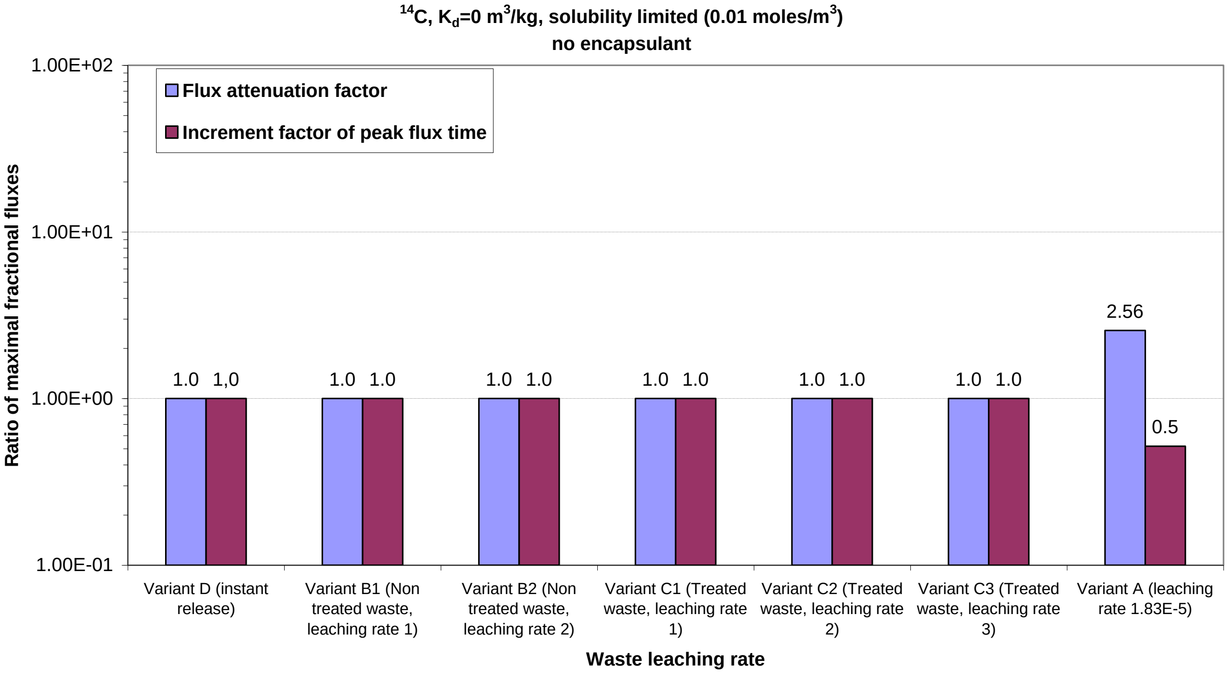

Fig. 10 Variations of peak fractional ^{14}C flux and its observation time within analyzed cases

As could be seen in Fig. 10 the impact of solubility limit is obvious. Decreased of peak flux was observed for variant A only with its earlier time to peak.

Case 3: K_d=0.2 m^3/kg, solubility limit=0.01 moles/m^3

In this case the credit has been taken to chemical interaction in terms of sorption and solubility limitation as the backfill from cementitious material suppose to play role of chemical barrier primarily. Fig. 11 presents the comparison of the radionuclide flux to the backfill and to the geosphere and could represent the substantial role of backfill in radionuclide retardation in such a repository configuration. The attenuation of the inorganic ^{14}C flux coming from the waste take places over the orders of magnitude.

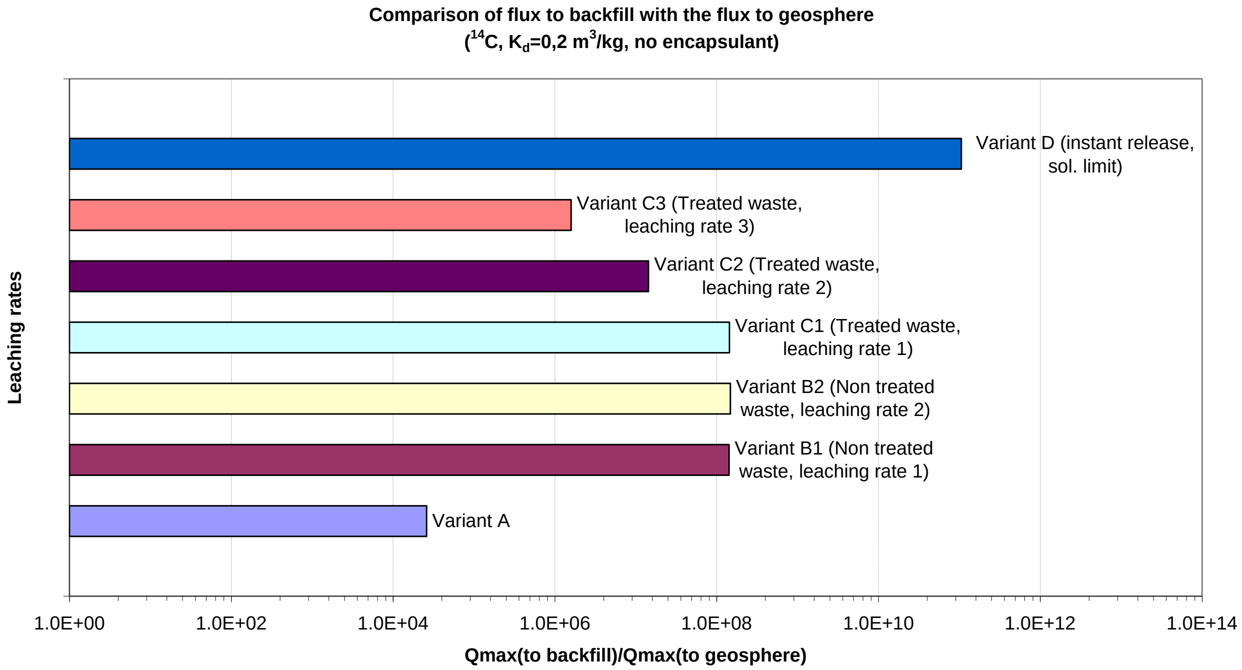

Fig. 11 Comparison of ^{14}C flux to the backfill and to the geosphere under from RBMK-1500 inventory released at different waste leaching rates

Due to transport and interaction with a tunnel backfill material the ^{14}C flux to the geosphere was decreased and the maximal fractional flux for different leaching conditions is in the range of 10^{-12} - 10^{-11}1/yr (Fig. 12.) in comparison to 10^{-5}–10^{-3}1/yr without consideration of these two processes.

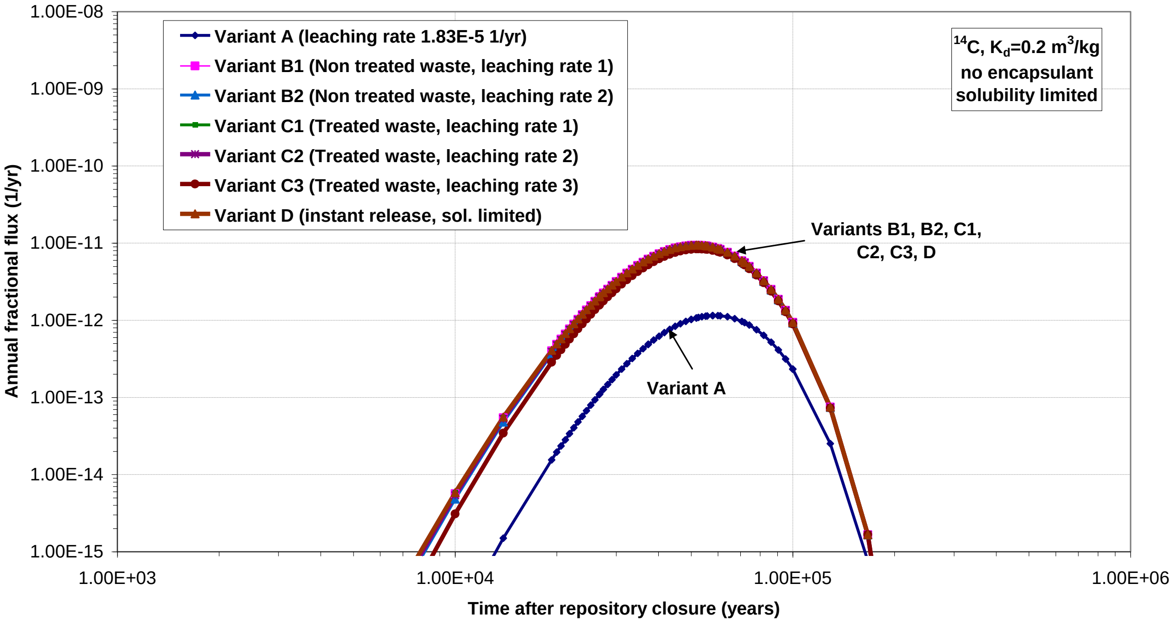

Fig. 12. Release of ^{14}C to the geosphere from one disposal container with non encapsulated waste

That means that the engineered barriers as a system provide effective measures to retard the radionuclide within them and indicates the role of backfill material on the radionuclide retardation and decreased importance of the leaching rate profile.

In case of ^{14}C interaction with backfill, encapsulant material which could be expressed in term of sorption, a significantly decreased flux was observed in comparison to Case 1 (without consideration of chemical interaction, Fig. 7). As it could be seen there is no plateau in the profile indicating the concentration in the pore water is not longer limited as a large part of leached inventory could be removed from liquid phase by undergoing sorption. The comparison of maximal fractional fluxes and its observation times was performed to that of observed in case of instant release and is summarized in Fig. 13 . Most effective decrease of maximal flux was demonstrated if the current inventory is being leached at a rate of 10^{-5} 1/yr (flux lower by factor 8).

Modelled peak fractional flux was significantly below the transfer rate metric ($7 \cdot 10^{-6}$ 1/yr for UK inventory) applicable for the near field barrier indicating that in this case the disposability of RBMK-1500 graphite in such system of near field barriers could be supported.

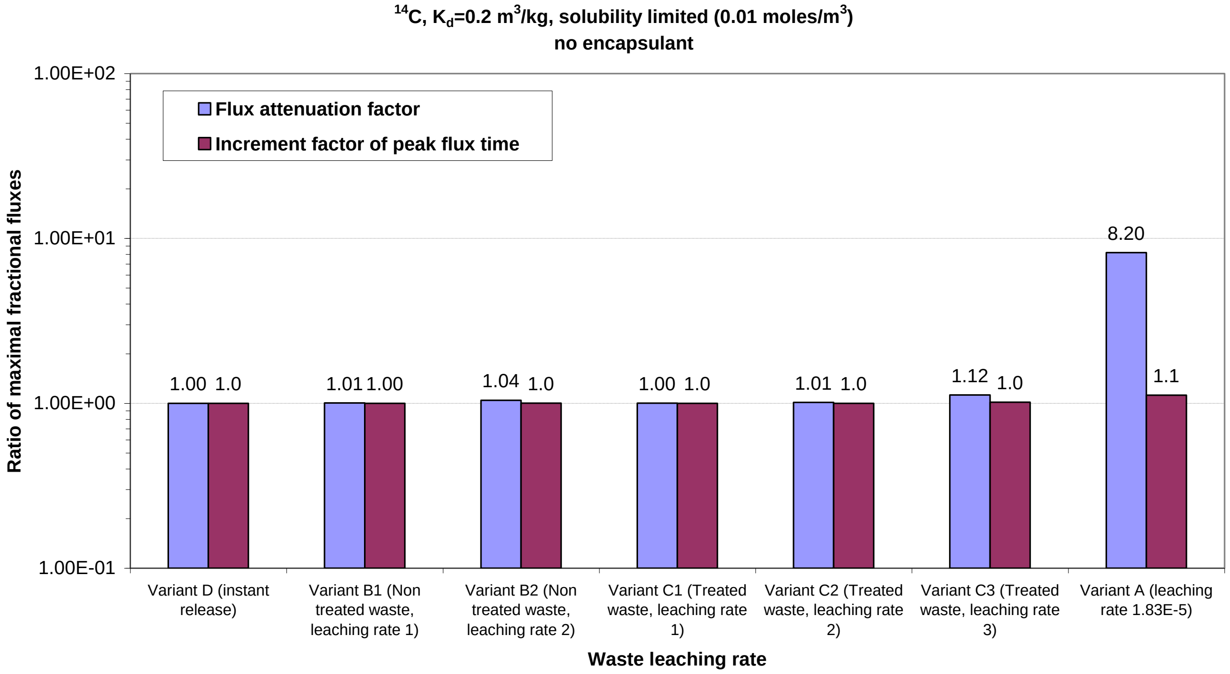

Fig. 13 Variations of peak fractional ^{14}C flux and its observation time within the cases being analyzed

The modelling results presented and their comparison indicated the decreased importance of differences of leaching rate if the chemical interaction in the repository backfill is demonstrated. In general, the impact of the options (treatment vs no treatment of graphite) on the radionuclide flux to geosphere is not straightforward. It depends on the leaching rate which is demonstrated (expected) for the particular option, particular inventory and prevailing transport conditions.

4.1.2 Alternative near field model (encapsulated waste)

The alternative near field model has been developed in order to represent the effect of possible waste encapsulation in the cementitious material. The impact of the variations of leaching rate on the annual fractional flux is similar to that of for non encapsulated waste.

Case 1: K_d=0 m^3/kg, no solubility limitation

If no sorption and no solubility limitation occur in the encapsulant (cement) as well as in the backfill material (cementitious material also), the estimated radionuclide flux dependencies are very close to that of determined in the reference near field model (without waste encapsulation) (Fig. 14).

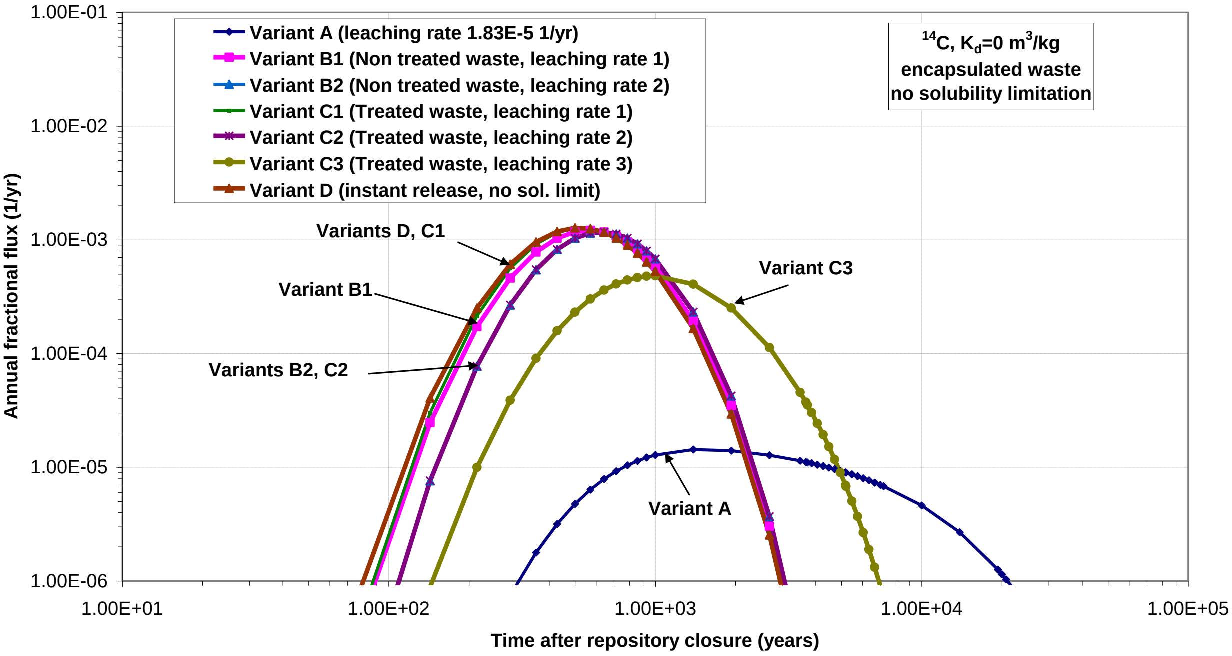

Fig. 14 Release of ^{14}C to the geosphere from one disposal container with encapsulated waste

The diffusive and advective transport through the encapsulant material does not result in a significant decrease of radionuclide flux to geosphere and its delay. The comparison of maximal fractional fluxes and its observation times is done to that of observed in case of instant release and is summarized in Fig. 15.

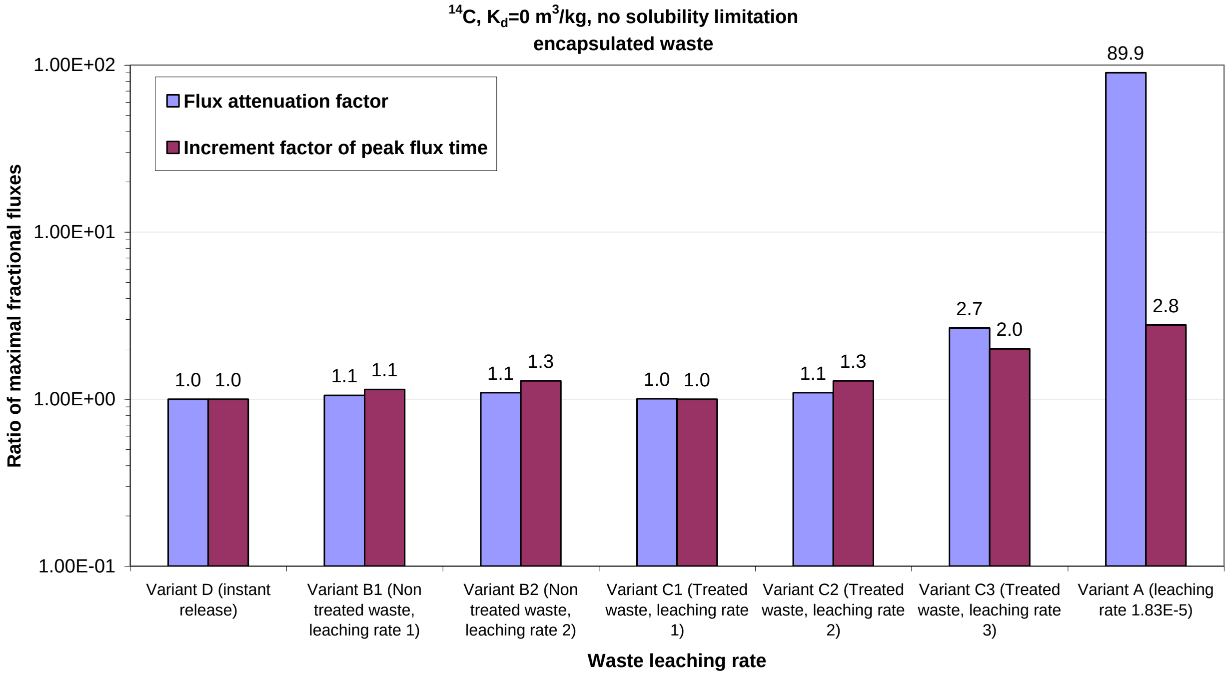

Fig. 15 Variations of peak fractional ^{14}C flux and its observation time within the cases being analyzed

The impact of the variations of leaching rate on the annual fractional flux is similar to that of determined for non encapsulated waste.

Case 2: K_d=0 m^3/kg, solubility limit=0.01 moles/m^3

It was observed that peak flux to geosphere remains the same although the release from the waste is instant or at the rates of the order of 10^{-3}-10^{0} 1/yr (Fig. 16). The profile of the flux out of the backfill indicates the impact of solubility limitation. In case of encapsulated waste the solubility limitation takes place in the material closest to the waste (encapsulant). The peak flux is lower in comparison to non encapsulated waste as well as the time to peak is slightly increased due to the presence of encapsulant. The maximal fractional flux in case of instant release (variant D) is observed ~$6\cdot10^{3}$ years after the repository closure.

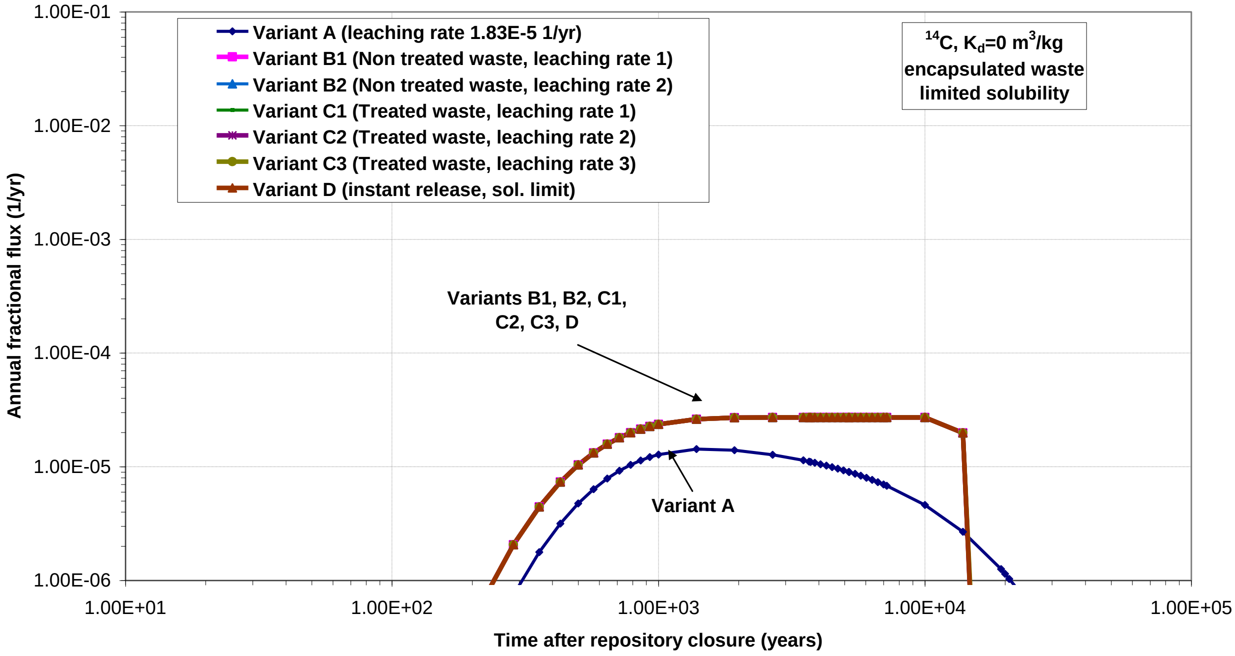

Fig. 16 Release of ^{14}C to the geosphere from one disposal container with encapsulated waste

The impact of lower annual flux from the waste in comparison to instantly released inventory is presented in Fig. 17 .

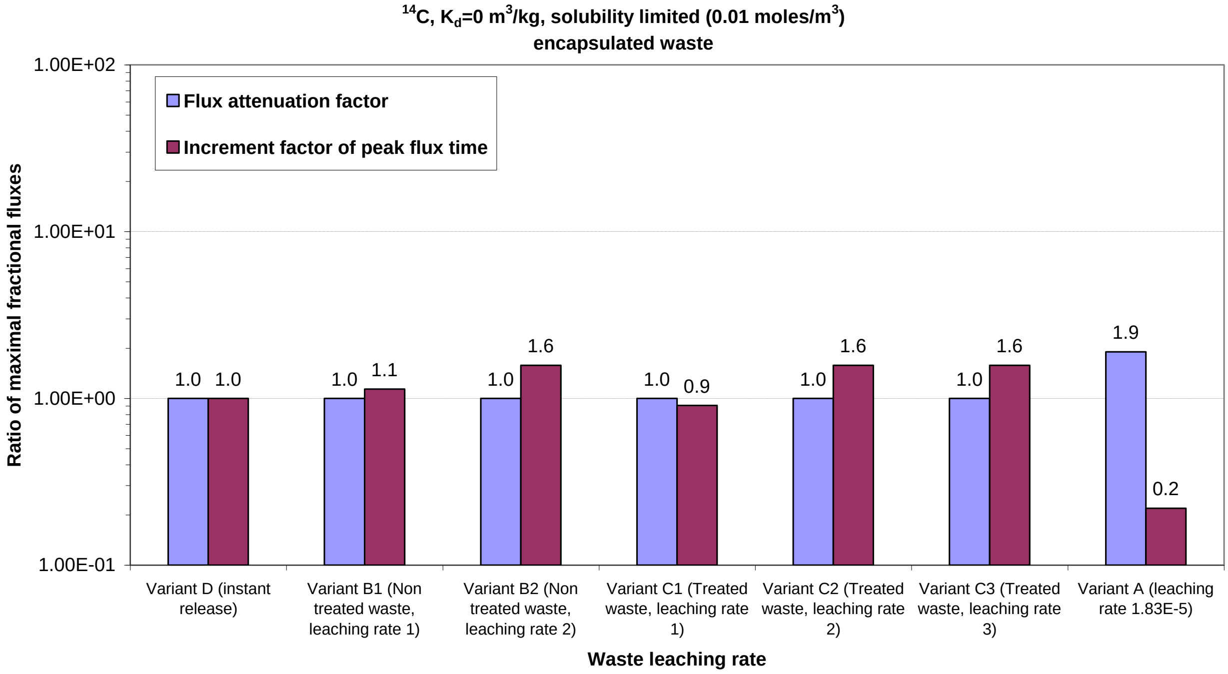

Fig. 17 Variations of peak fractional ^{14}C flux and time of peak release within the cases being analyzed

Case 3: K_d=0.2 m^3/kg, solubility limit=0.01 moles/m^3

The variations of peak fractional flux's time are diminished as for non encapsulated waste (Fig. 12.). However the presence of the additional chemical barrier (encapsulant) resulted in lower peak flux app. by one order of magnitude (Fig. 18).

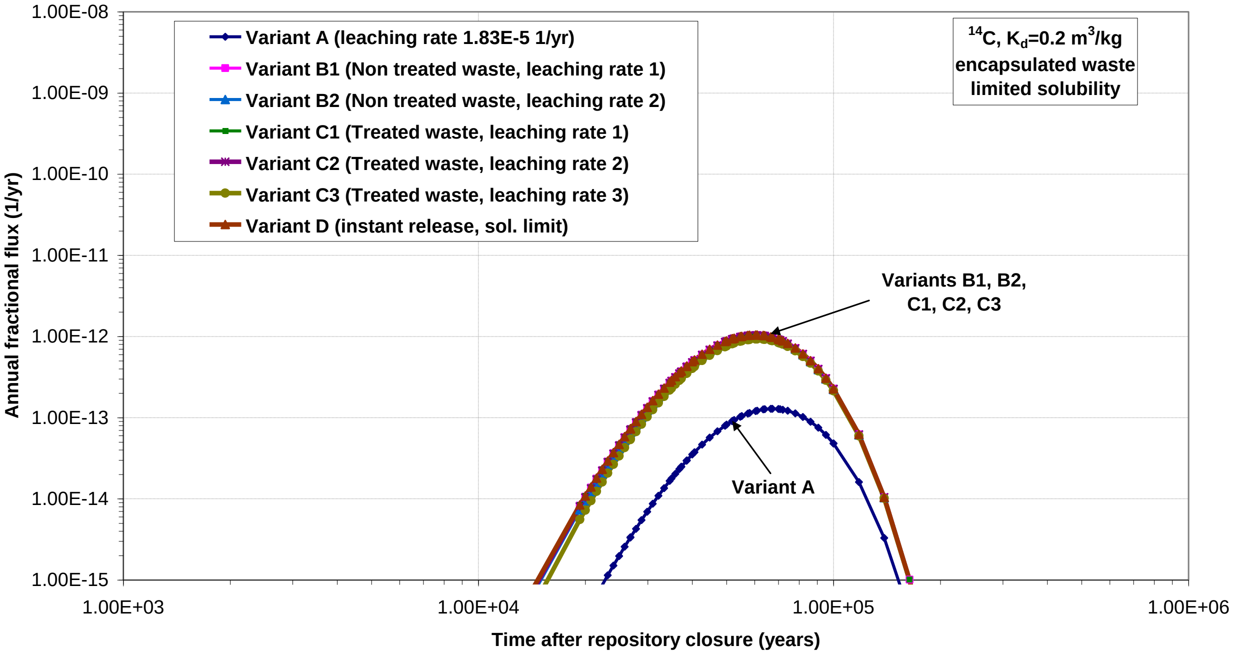

Fig. 18 Release of ^{14}C to the geosphere from one disposal container with encapsulated waste

Estimated maximal fractional flux from RBMK graphite to the geosphere depends on the leaching rate and varies within the range of 10^{-13}-$1\cdot10^{-12}$ 1/yr. The comparison of maximal fractional fluxes and its observation times is done to that of observed in case of instant release and is summarized in Fig. 19 .

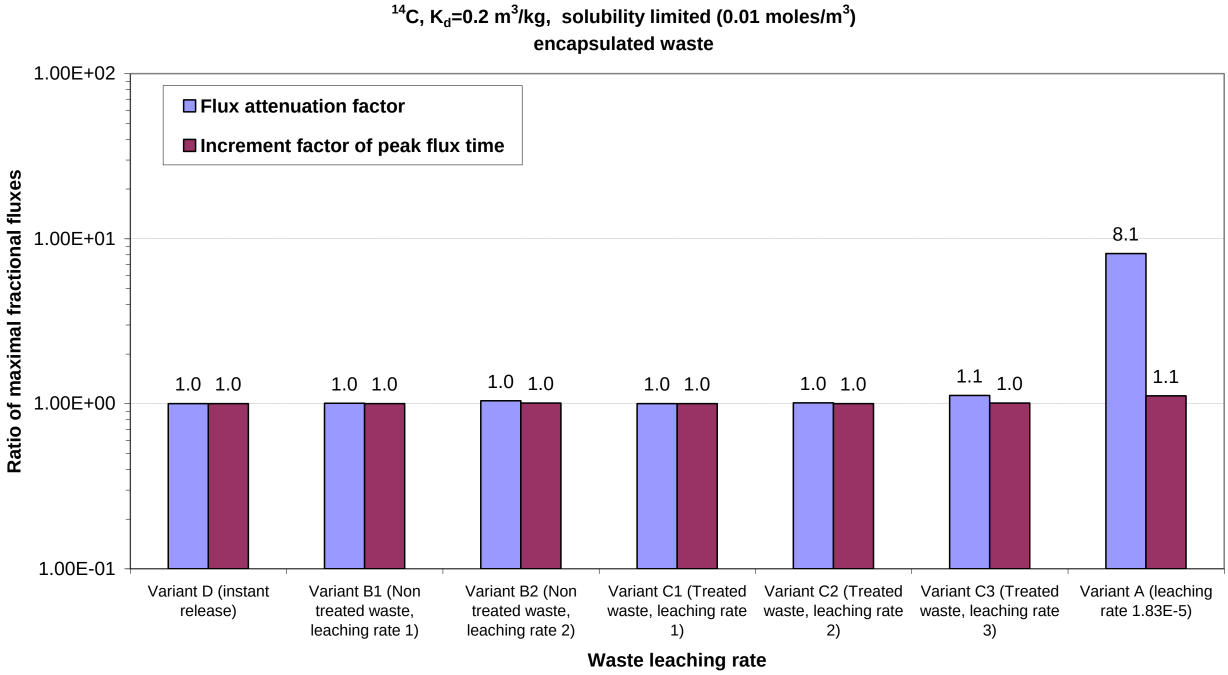

Fig. 19 Variations of peak fractional flux and its observation time within the cases being analyzed

As it could be seen in Fig. 19 , the most effective decrease of maximal flux is demonstrated if leaching rate is the order of 10^{-5} 1/yr. In this case the maximal flux to the geosphere would be decreased by a factor of ~8. The reasoning of leaching rate of the order 1-10^{-3} 1/yr does not provide any significant benefit for the analyzed graphite, as the maximal fractional flux is almost the same. Time of peak flux is prolonged in case of low leaching rate (variant A) only (by a factor of 1.1), while in the rest cases it is almost the same as in case of instant release.

Comparison of the modelling results for not encapsulated and encapsulated waste

As it has been mentioned above if no credit has been taken for the solubility limitation and sorption in the encapsulant and in the backfill, the estimated maximum of radionuclide flux are very close to that of determined in the reference near field model (without waste encapsulation) (Fig. 20) The diffusive and advective transport through the encapsulant material does not result in a significant decrease of radionuclide flux to geosphere and its delay.

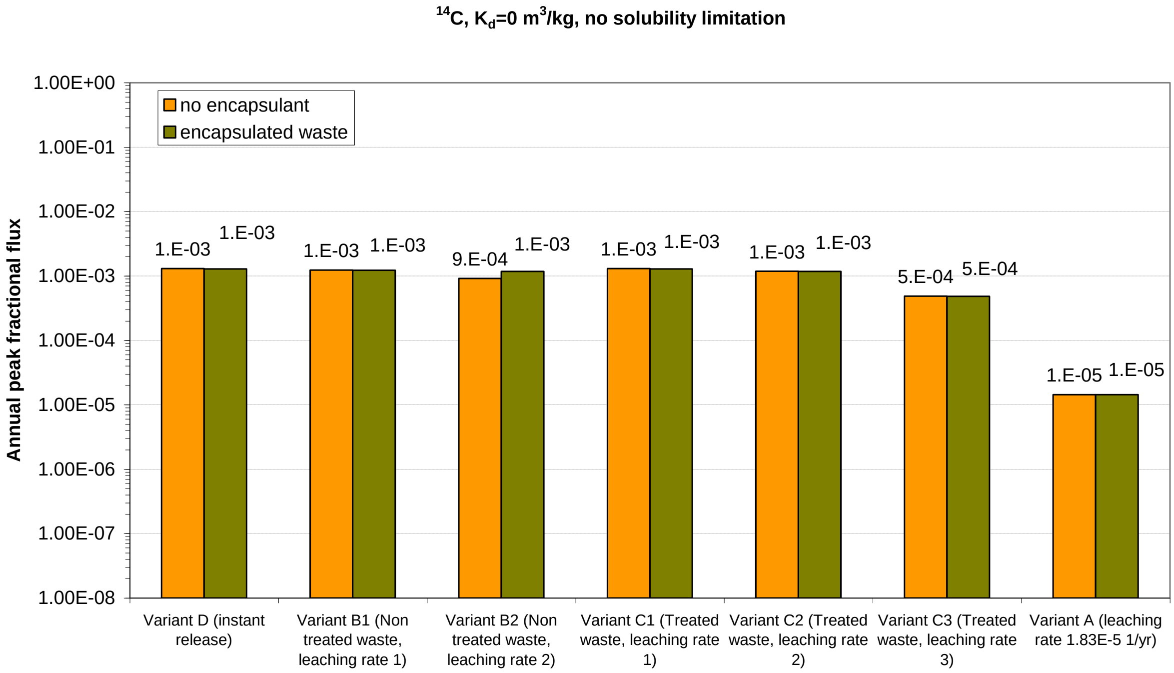

Fig. 20 Comparison of peak release for non encapsulated and encapsulated waste (Case 1)

If the chemical interactions with the material of encapsulant and backfill will result in the conditions for limited solubility of ^{14}C in the porewater and the inventory is being leached by the rate of the order of 10^{-3} - 10^{0} 1/yr (covered by analyzed variants B-D) the encapsulant acts as the additional chemical barrier and results in the decrease of flux by a factor of 1.3 (Fig. 21). If the leaching rate is of the order of 10^{-5} 1/yr, the radionuclide concentration in the porewater of encapsulant do not exceed solubility limit and the encapsulation does not show any benefit for the particular ^{14}C inventory in RBMK-1500 graphite.

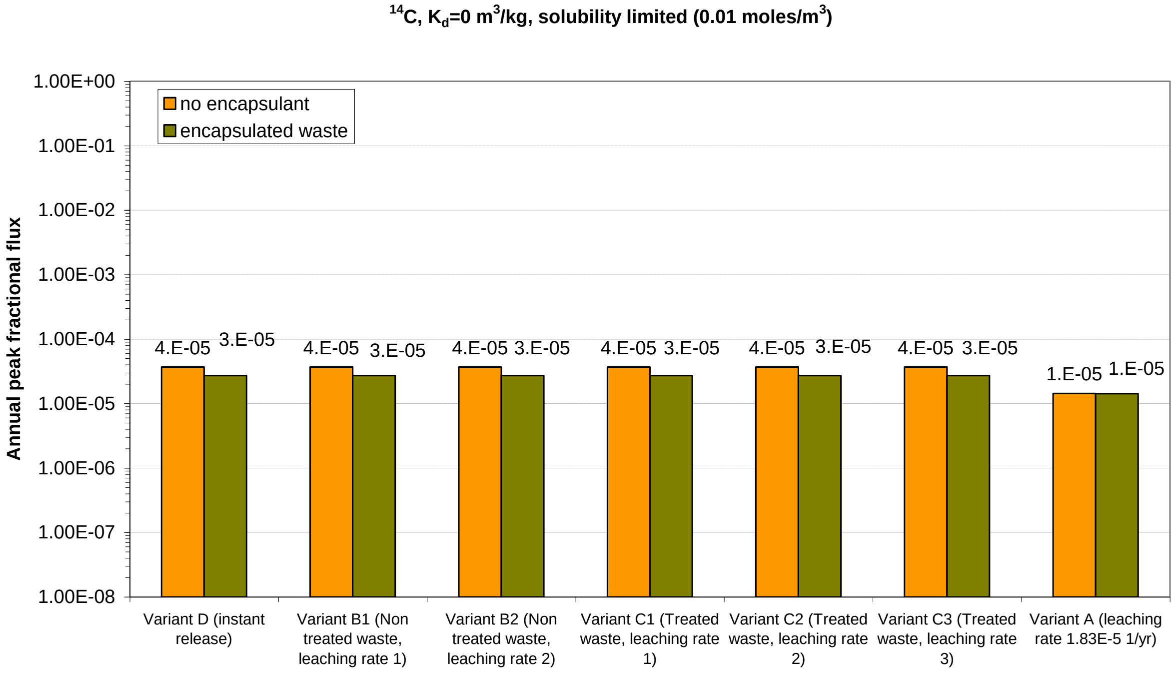

Fig. 21 Comparison of peak release for non encapsulated and encapsulated waste (Case 2)

If the retention occurs also in terms of sorption and limited solubility, the impact of waste encapsulation is more significant and lead to the decrease of peak flux by up to app. one order of magnitude in comparison to not encapsulated waste (Fig. 22).

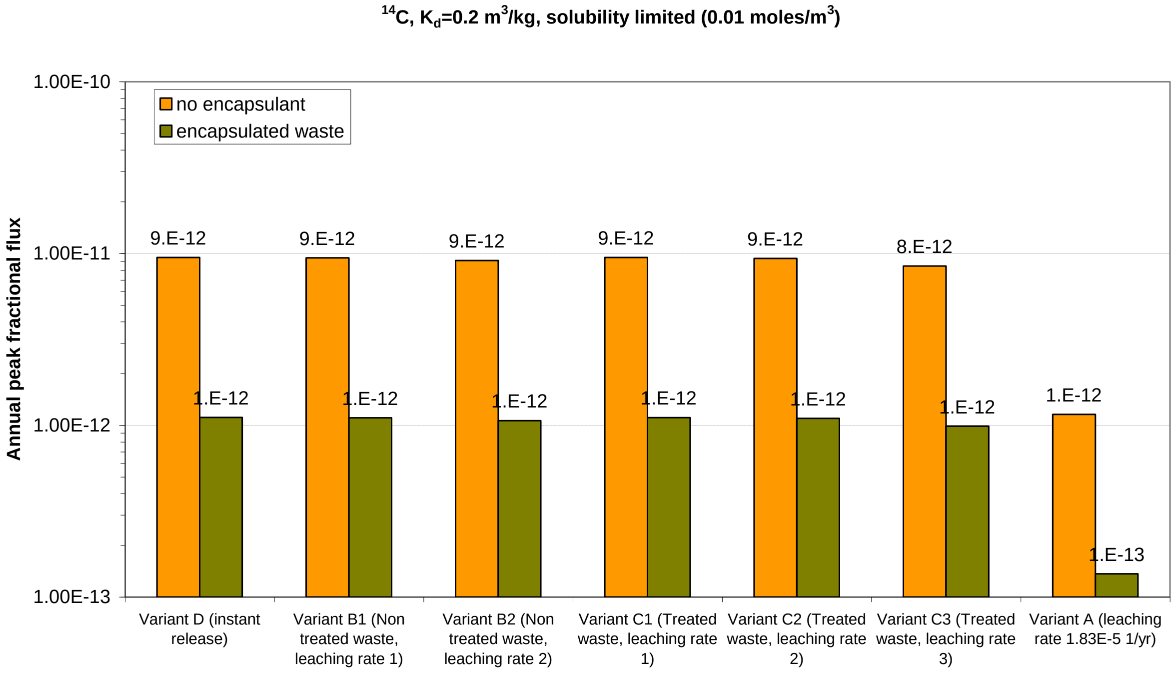

Fig. 22. Comparison of peak release for non encapsulated and encapsulated waste (Case 3)

4.1.3 Release of ^{36}Cl

Chlorine release through the EBS from the waste matrix and considering no interaction with the backfill as well no solubility limitation was modelled too. The leaching rate of $6.3\cdot10^{-2}$ 1/yr and instant release were assumed. The modelling results are presented in Fig. 23 .

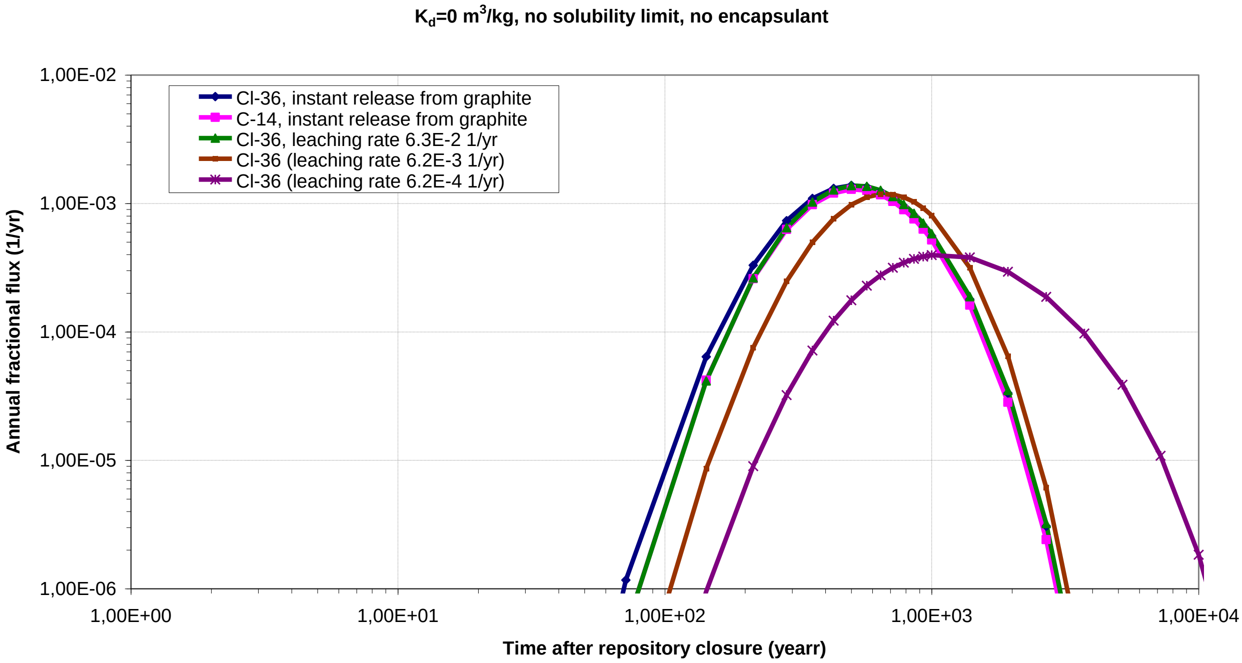

Fig. 23 Release of ^{36}Cl to the geosphere from one disposal container with non encapsulated waste

The modelling results have showed that the release profile and maximum of fractional ^{36}Cl flux to the geosphere do not differ in case of instant release and the leaching rate of $6.2\cdot10^{-2}$ 1/yr. As it could be expected the annual fractional flux is similar to that of ^{14}C if no retention in the EBS is considered. The radionuclide instantly released from the waste matrix (no credit for container corrosion) and is transported to the geological environment in a fast manner.

The variant calculations were performed assuming leaching rate of $6.2\cdot10^{-2}$ decreased by 1-2 orders of magnitude and are presented in the Fig. 23 The results have showed that only in case of leaching of the order of $1\cdot10^{-4}$ 1/yr the flux to the geosphere becomes lower (by a factor ~3.4). During ^{36}Cl migration its concentration in the porewater is not affected by interaction with surface in terms of sorption or formation of precipitates (due to limited solubility). Due to large half-life of ^{36}Cl, decay does not have a significant impact on the flux to the geosphere. Thus in case of fast release of ^{36}Cl the dominating role for the repository safety has to be guaranteed by the geological barriers or the EBS of lower permeability resulting in slower migration.

4.1.4 Summary

The impact of the options (treatment vs no treatment of graphite) on the radionuclide flux to geosphere is not straightforward. It depends on the leaching rate which is demonstrated (expected) for the particular option and the radionuclide inventory. The results presented in the sections 4.1.1-4.1.2 and their analysis indicated that the impact of the differences in the waste leaching rate depends on the performance of engineered barriers (in term of retention). Comparison of the radionuclide flux (^{14}C) released to the backfill and from it indicates the backfill being of key importance as well. Depending on the amount of radionuclides being released from waste and the transport conditions in the backfill the impact of the differences in the leaching rates might be slightly or significantly diminished. If the retention in terms of “sorption” do not occur the peak fractional flux of ^{14}C dissolved in groundwater would vary within app. 2 order of magnitude due to different inventory released from graphite waste, while the difference in the leaching rate could be up to app. 5 orders of magnitude. Decreased importance of variations in leaching rates is indicated if the retention in the repository backfill could be demonstrated with the projection in the long-term perspective.

For ^{14}C inventory in RBMK graphite considered in this study the leaching rate of the order of 10^{-5} 1/yr indicates more significant decrease of peak flux from near field in comparison to flux determined by instantly released inventory. The radionuclide transport analysis has been carried out considering the possible graphite waste encapsulation in the cementitious material. It was observed that the option of waste encapsulation could give a benefit for the radionuclide flux attenuation if the retention in the repository backfill could be demonstrated. In case of sorption of 0.2 m^3/kg the peak fractional flux to the surrounding geosphere could be decreased app. by one order of magnitude with various leaching rates.

The peak fractional flux was compared to the transfer rate metric what could give rise to an impact around the regulatory guidance levels and was defined for UK graphite inventory. The comparison showed that in case of no sorption and/or limited solubility the peak fractional flux exceed the metric. If the near field barrier acts as a chemical barrier the peak fractional flux of ^{14}C is significantly below the metric and supports the RBMK-1500 graphite disposability even without the consideration of the geosphere.

Comprehensive evaluation of treatment vs no treatment demand of RBMK graphite could not be based on the leaching rate only, but needs to be complemented by the examination of a range of flow/transport, chemical conditions expected in the engineered barriers followed by reasonable evaluation of radionuclide inventory within it.

4.2 Release through the natural barriers

Release of ^{14}C in the far field (geosphere) was modelled using the computer code Petrasim [24] with a TOUGH2 incorporated, which is widely applied software for multiphase multicomponent flow modelling. For the radionuclide transport the module EOS7R was applied.

Modelling has been performed under the conservative assumption that all instantly released ^{14}C does not interact with the backfill (no sorption and no solubility limitation) and is transported to the geosphere. No interaction with the geological media assumed also. The simulations were performed assuming that ^{14}C from the repository is transported in liquid phase..

The analysis of radionuclide transport is concentrated on the radionuclide flux to the surface water located in the upper corner of the conceptual model. This “sink” tends to represent some kind of surface water body (river, lake, etc) with a constant water level in it. Fig. 24 represents the annual fractional flux radionuclide ^{14}C from the geosphere (in terms of Bq/yr per Bq of ^{14}C disposed in the repository). For the comparison the annual fractional flux from the near field is presented too.

As it could be seen from the figure, the system of natural barrier even disregarding the sorption in geologic environment contributes to the significant delay of radionuclides and it maximal release rate occurs after app. 40 thousand years past closure. While the peak flux from the near field is observed around 500 years past closure. Comparison of the peak fluxes also shows the significant contribution to the attenuation of radionuclide flux. Maximal fractional flux from the far field is lower by app. 5 order of magnitude. Sorption on the geological barriers will lead to the lower flux than indicated in the Fig. 24 under the same flow conditions. Thus the importance of waste leaching rate depends on the performance of the backfill (in term of sorption and solubility) and natural barrier system (on the scope of its impact on the attenuation of the radionuclide flux).

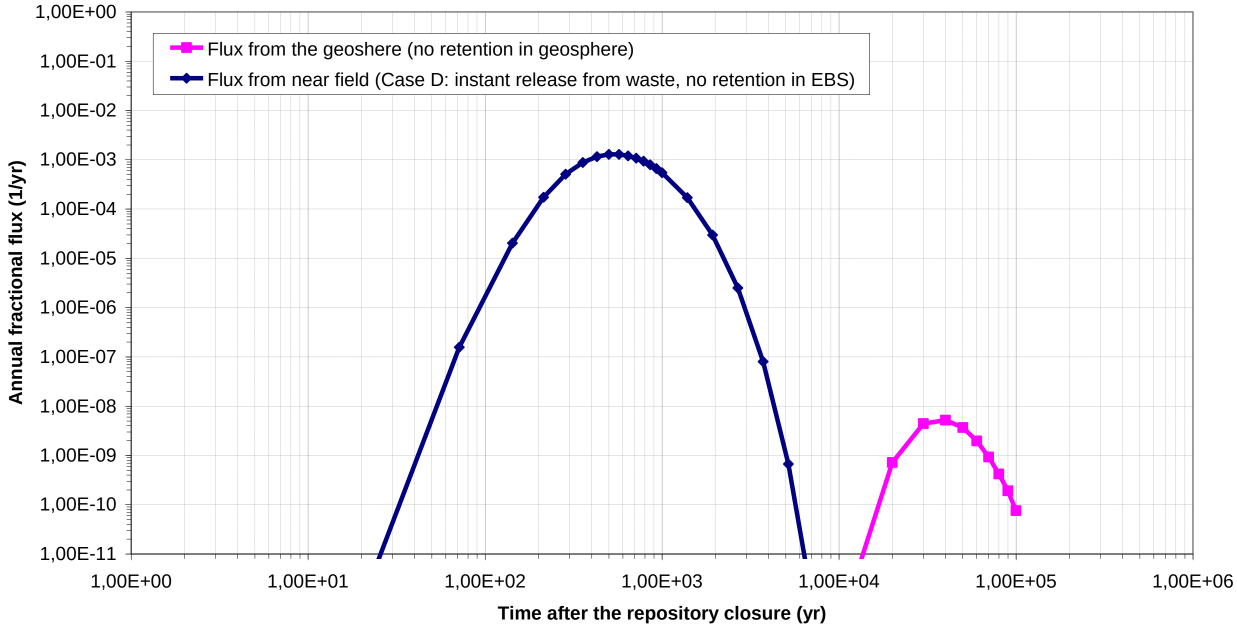

Fig. 24. Release of ^{14}C from the geosphere (instant release from graphite, no sorption in the EBS and no solubility limit)

In case of no retention in the EBS and geosphere and if the flux in the geosphere is not attenuated up to sufficient level, the precise definition of ^{14}C leaching rate from graphite waste becomes an important issue. Based on the comparison of the releases from the near field in case of instant release (variant D) and leaching rate of $1.83 \cdot 10^{-5}$ 1/yr (variant A) the flux from the near field would be lower app. 2 orders of magnitude thus the flux from the geosphere is expected to be decreased additionally at least by the same factor. Comparison of peak fractional flux to transfer rate metric showed that the metric is not exceeded even the near field do not provide enough containment. This would support the conclusion of RBMK-1500 graphite disposability in geological repository in the respect of ^{14}C migration in liquid form.

The distribution of dissolved radionuclide in the environment surrounding the repository is presented in Fig. 24 after 40 thousand years of repository closure.

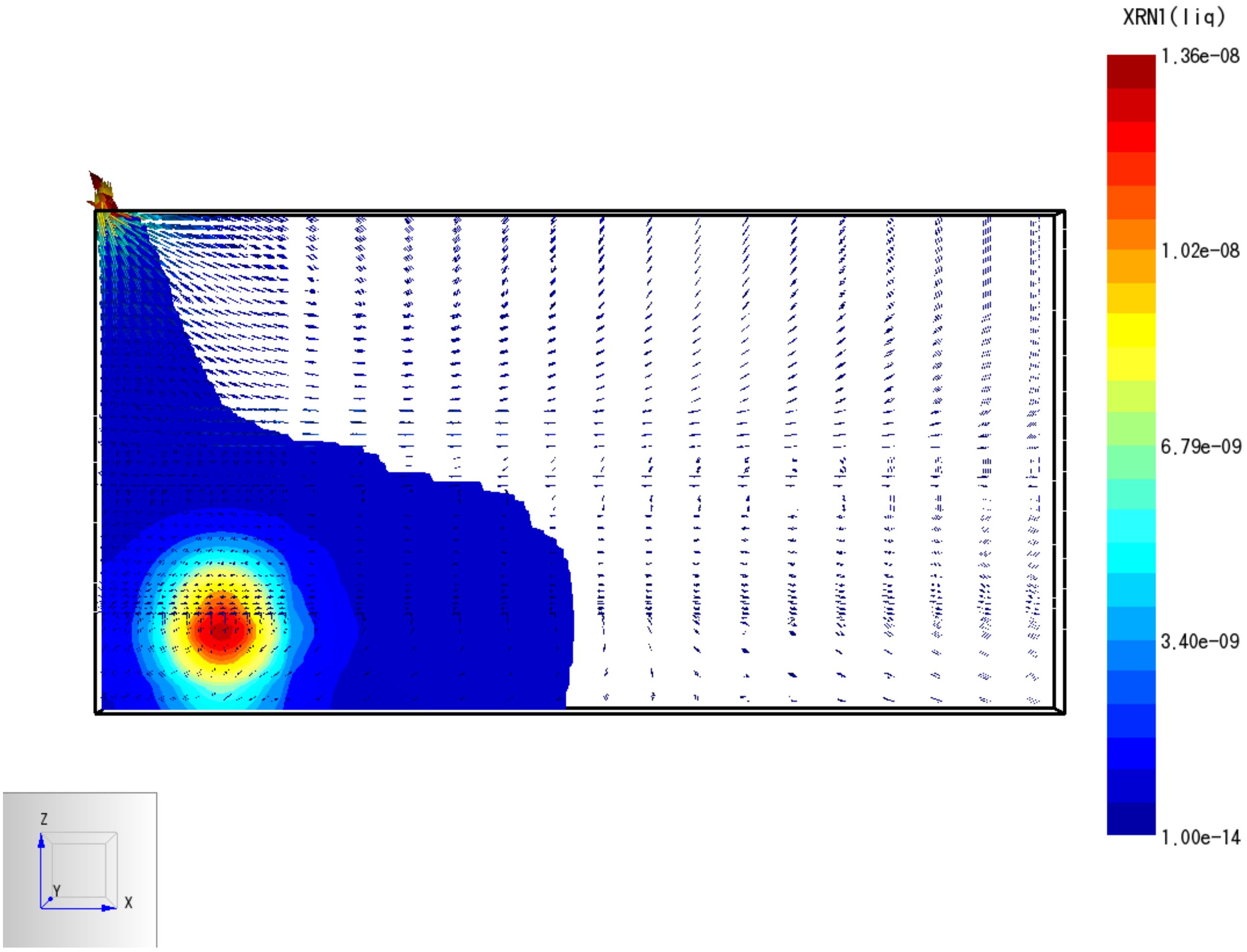

Fig. 25. Distribution of dissolved ^{14}C in the geological environment surrounding the repository (after 40 thousand years of repository closure)

As it could be seen there is radionuclide plum migration in the geosphere and it is distributed more in the direction of groundwater flow towards the surface. The repository location is in the crystalline rocks, which might be fractured and some major fractures (fractured zone) could be at certain distance from the repository. During this modelling study a fractured zone of regional scale has not been considered. Besides the crystalline rocks in Lithuania are underlying with the system of sedimentary rocks (differently to that is in Sweden or Finland), thus the direct pathway by fracture up to the surface is less expected. Such a complex system of the geological barriers of different properties may provide an adequate containment and isolation function and need to be investigated in more detailed manner.

5 Summary and conclusions

The main task for LEI was to study the relation between treatment and disposal on the performance of RBMK-1500 graphite in crystalline rock. In order to perform this task the radionuclide transport models in the near field/ far field environment were developed. The differences in the waste leaching waste (for ^{14}C) corresponding to the different state of the graphite (treated, non treated) were analyzed, the impact on the near field flux was evaluated. The importance of waste leaching rate depends on several aspects: on the performance of the backfill (in term of sorption) and natural barrier system (on the scope of its impact on the attenuation of the radionuclide flux). The impact of the options (treatment vs no treatment of graphite) on the ^{14}C flux to geosphere is not straightforward. While reasoning of option of treatment/not treatment the inventory, leaching rates, barrier performance and transport conditions need to be considered.

The performed evaluation of the ^{14}C distribution using the developed numerical models of the RBMK-1500 irradiated graphite disposed of in the crystalline rocks allows to conclude that:

- The peak fractional flux from the near field would vary within app. 1-2 orders of magnitude due to different leaching rates from the graphite waste (difference in the leaching rate was within app. 5 orders of magnitude);
- The radionuclide transport analysis considering the possible graphite waste encapsulation in the cement material revealed that waste encapsulation could give a benefit for the radionuclide flux attenuation up to app. one order of magnitude;
- For the conservative far field modelling case (instant release from waste, no sorption in the near field and far field, repository location in upward flow conditions), the natural barriers contribute to the delay and decrease of dissolved ^{14}C flux from the hypothetical repository with RBMK-1500 irradiated graphite by at least by 5 orders of magnitude.
- For much lower amount of RBMK-1500 graphite than in UK case the peak fractional flux would be less than the UK inventory based transfer rate metric even with our conservative assumptions on the near field and far field performance. The compliance with the metric ($7 \cdot 10^{-6}$ 1/yr) would support the geological disposability of RBMK-1500 graphite in respect of ^{14}C migration in liquid form.

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