LITHUANIAN ENERGY INSTITUTE

3 Breslaujos str., LT-44403 Kaunas, Lithuania
Tel. +370 (37) 351403 • Fax +370 (37) 351271

ISO 9001
ISO 14001

Nuclear engineering laboratory

Report on
EC 7th Framework Programme for research and technological development project

CARBOWASTE „TREATMENT AND DISPOSAL OF IRRADIATED GRAPHITE AND OTHER CARBONACEOUS WASTE“

EC Grant Agreement No.: FP7 - 211333
2008 04 01 – 2012 03 31

Task 6.4: Assessment of waste performance

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

Deliverable: D-6.4.4

SEVENTH FRAMEWORK PROGRAMME

Authors:
Asta Narkuniene
Povilas Poskas
Raimondas Kilda

CARBOWASTE

Kaunas, 2012

Distribution list

Person and organisation name and/or group	Comments
WP6 members	
Executive Board	
Co-ordinator	
European Commission	

CARBOWASTE		
Work package: 6 **Task: :** 6.4	**CARBOWASTE document no:** CARBOWASTE-1203-D-6.4.4-a (e.g. May 2008 as date of issue: 0805)	**Document type:** T=Technical Report
Issued by: LEI, LIT **Internal no.:** CW1203-Deliverable-6-4-4-a		**Document status:** Final

Document title

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. 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. It depends on the leaching rate which is demonstrated (expected) for the particular option.
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. 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 significantly.

Revisions

Rev.	Date	Short description	Author	Internal Review	Task Leader	WP Leader
00	dd/mm/yyyy	Issue	Name, Organisation	Name, Organisation *Signature*	Name, Organisation *Signature*	Name, Organisation *Signature*
01	09/03/2012	Final issue	A. Narkuniene LEI	P. Poskas LEI		Grambow Subatech

Table of Contents

1 Introduction 5
2 Methodology 5
3 Development of radionuclide transport models from the RBMK-1500 graphite disposed of in the crystalline rocks 6
3.1 Repository concept 6
3.1.1 RBMK-1500 graphite 6
3.1.2 Waste package 7
3.1.3 Disposal tunnel 8
3.2 Geosphere 10
3.3 Radionuclide release from repository containing irradiated graphite 11
3.4 Modelling of radionuclide release through the EBS 16
3.4.1 Conceptual model 16
3.5 Modelling of radionuclide release in the geosphere 19
4 Results and discussion 21
4.1 Release through the engineered barriers 21
4.1.1 Reference near field model 21
4.1.2 Alternative near field model (encapsulated waste) 27
4.1.3 Release of ^{36}Cl 32
4.1.4 Summary 34
4.2 Release through the natural barriers 34
5 Summary and conclusions 38
6 References 39

1 Introduction

LEI team is involved in the **Task 6.4: Assessment of waste performance** in 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 being organized was done in several stages as summarized in Fig. 1.

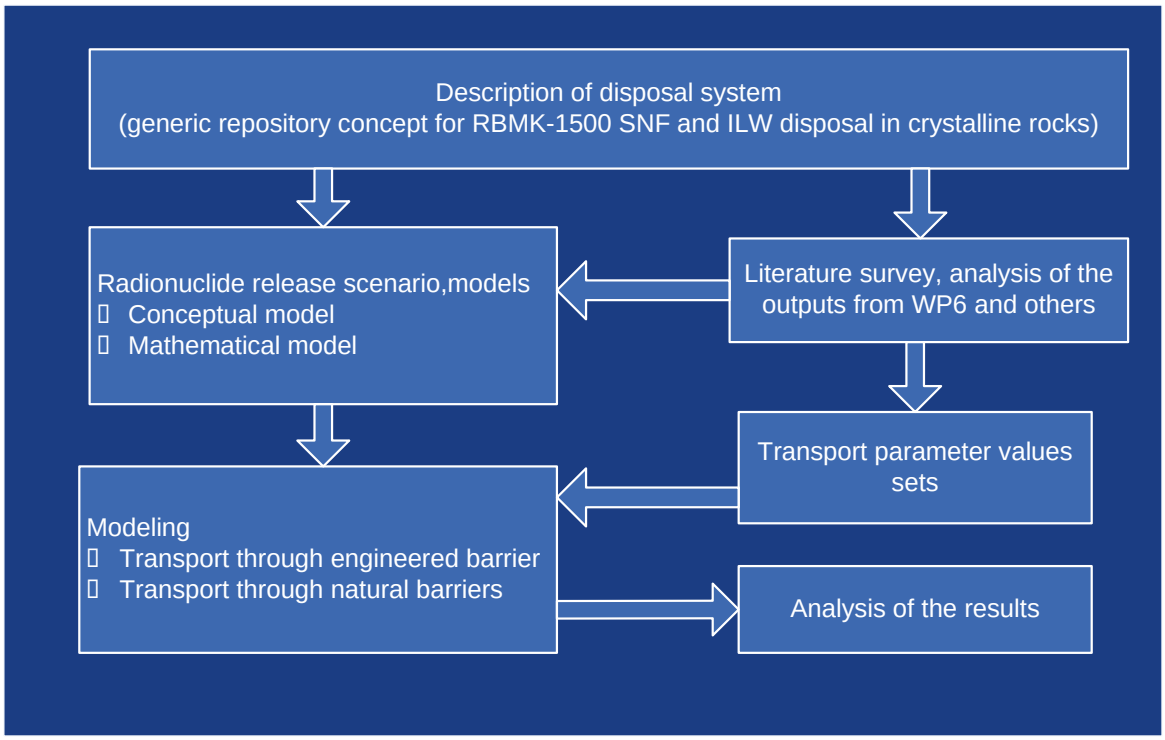

Fig. 1. Stages of the LEI work within task 6.4

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 the various data on the various physical/chemical processes determined the release of the radioactive material 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. The ILW (and spent graphite) are planned to be stored in the concrete 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. After the emplacement of the ILW the void regions within the tunnels would be backfilled with cementitious grout (backfill) as proposed in NIREX concept (United Kingdom) or with gravel as proposed for Swedish repository.

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 consisting of a graphite as a moderator and reflector. This means that after the dismantling of the reactor cores app. 3600 tones of graphite blocks and sleeves will be accumulated. 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 maximal initial nitrogen impurities in graphite matrix and nitrogen is present in all (open and closed) graphite pores the maximum activity of GR-280 type graphite was used (higher than for 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]	~3600
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].

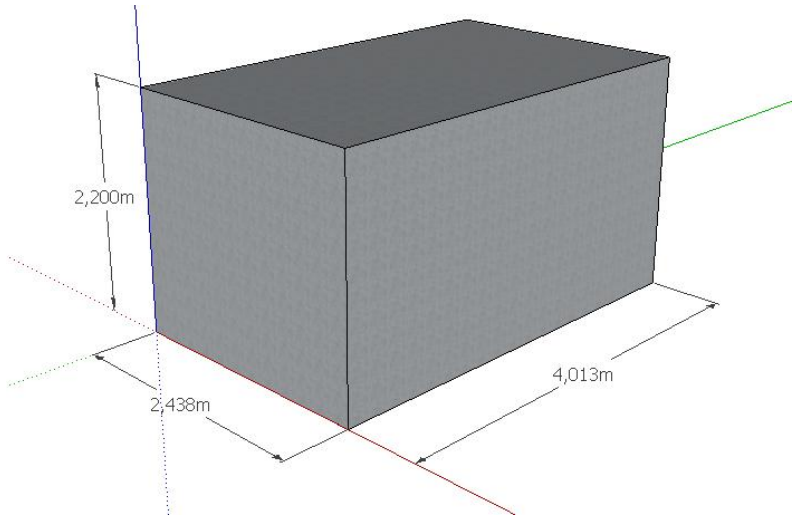

Fig. 2. Steel container considered for the RBMK-1500 graphite waste in this analysis

Total internal volume of the container of these dimensions is 18.9 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. 163 with total internal volume to be occupied by waste 12.7 m^3.

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).

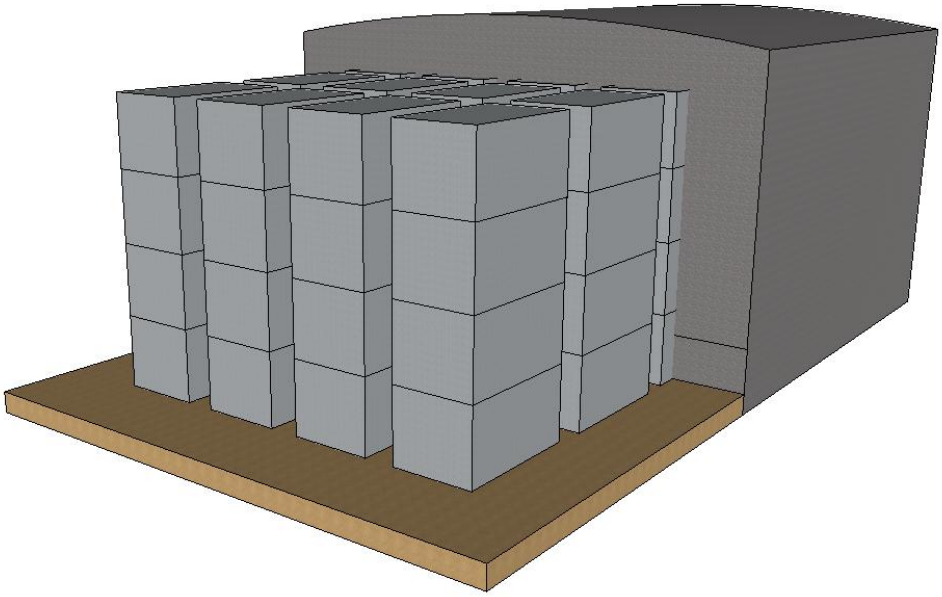

Fig. 3. Graphite waste disposal tunnel considered for the modelling

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]	Hydraulic conductivity (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.

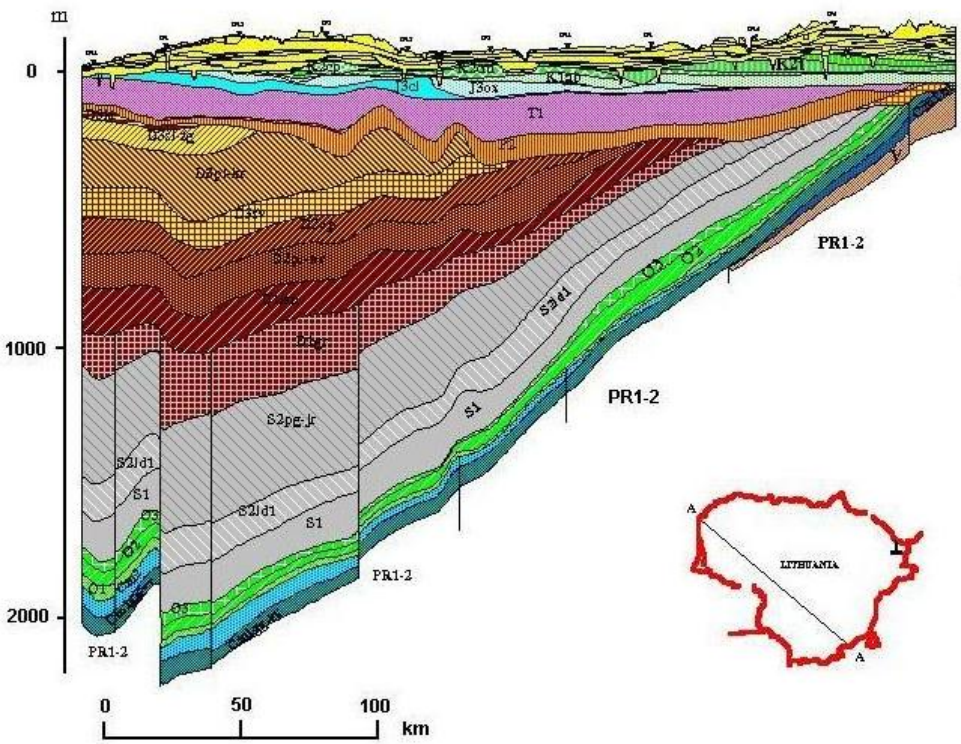

Fig. 4. Hydrogeological cross-section of Lithuania; crystalline basement is marked as "PR1-2" (Authors: L. Kilda, S. Šliaupa, J. Lazauskienė)

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. The prospective area of the crystalline basement was confirmed as occurring in the southern Lithuania with 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 [11, 12]. 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. 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 groundwater 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 [13], 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 [14]. Reported ^{14}C dissolution rate was derived from G2 experiments is 3.10^{-7} $g \cdot m^{-2} \cdot j^{-1}$ [14]. 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).

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

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

^{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 [15]. 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 [15].

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 is probably less than 0.01.

Chlorine release

As indicated in [16] 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 [14] 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 transport 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 [18] 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 [18] 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 [19] 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 ^{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. The sorption of ^{14}C in NIREX cementitious 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 [19]

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 is 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 [20] 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 [21] 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 preliminary models had been developed and numerical modelling of ^{14}C radionuclide transport in the near field was performed. The computer code AMBER [23] 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 released from the waste matrix interact with the groundwater percolating the waste matrix (solubility limit has been taken into consideration);
- Solubility limit is based on the chemical conditions and internal volume inside the container;
- Subsequently 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;

- 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 different 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] while evaluating the advective flow through the disposal tunnel.

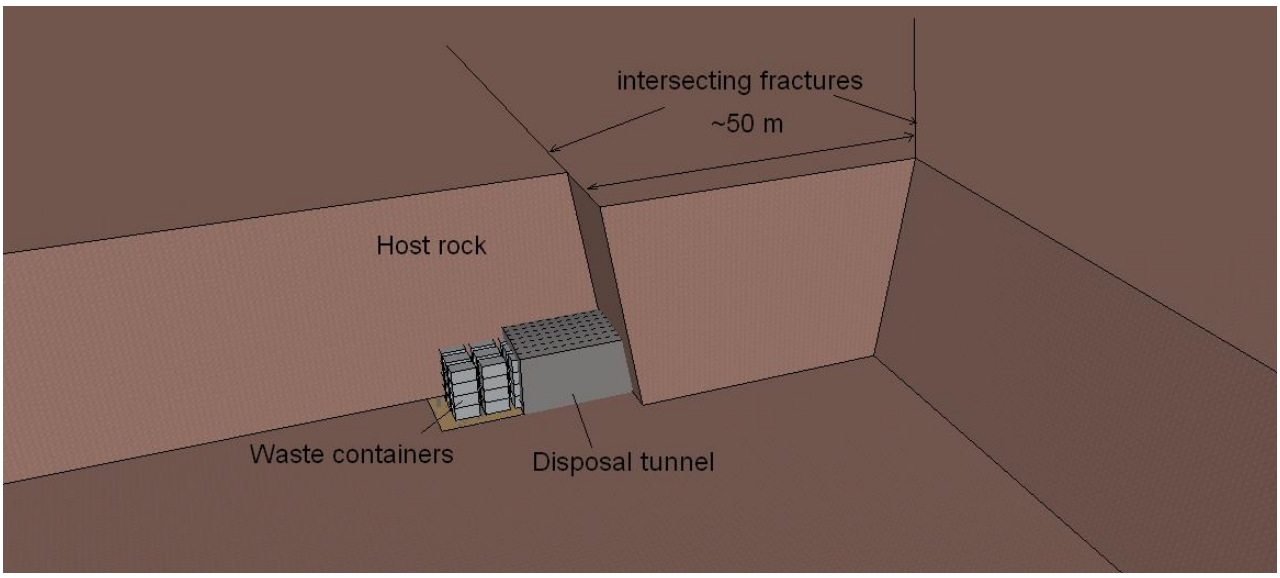

Fig. 5. Scheme of disposal tunnel and intersecting fractures

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 groundwater, 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. This 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 preliminary. 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:

- **Case A**;
- **Cases B1 and B2** (leaching rates used corresponds to the possible release from the non treated graphite waste);
- **Cases C1, C2, C3** (leaching rates used corresponds to the possible release from the treated graphite waste);
- **Case D** (conservative assumption of 100 % available ^{14}C inventory to be released from the container).

The leaching rates used in the simulations are of more indicative nature rather than the precise ones (Table 4).

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

	Fractional leaching rate, 1/yr						
Time after repository closure, yr	**Case A**	**Case B1 (not treated waste, rate 1)**	**Case B2 (not treated waste, rate 2)**	**Case C1 (treated waste, rate 1)**	**Case C2 (treated waste, rate 2)**	**Case C3 (treated waste, rate 3)**	**Case D**
1	$1.83 \cdot 10^{-5}$	0.1	0.1	0.1	0.01	0.001	Instant release from graphite
10	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	
10^2	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	
10^3	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	
10^4	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	
10^5	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	
10^6	$1.83 \cdot 10^{-5}$	0.01	0.001	0.1	0.01	0.001	

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.

Data on solubility limit (0.01 mol/m^3) and 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 [22], while in [19] for porous concrete range of $1\cdot10^{-10}$-$6\cdot10^{-10}$ m^2/s is reported. 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 software:

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

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 release 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 hypothetical repository is based on the conservative assumption that the hypothetical 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 [24] (Fig. 6).

Table 5. Hydrogeological data of natural barriers [24]

Layer no.	Depth (m)	Hydraulic conductivity (m/s)	Porosity (m^3/m^3)	Description
1	0–100	$1.2 \cdot 10^{-5}$	0.25	glacial loam
2	100–200	$2.3 \cdot 10^{-5}$	0.35	sand
3	200–250	$5.8 \cdot 10^{-5}$	0.5	anhydrite
4	250–270	$1 \cdot 10^{-6}$	0.5	limestone
5	270–290	$5.8 \cdot 10^{-5}$	0.05	sandstone
6	290–310	$1.16 \cdot 10^{-10}$	0.13	clay
7	310–410	$6 \cdot 10^{-6}$	0.05	sandstone
8	410–420	$1.16 \cdot 10^{-7}$	0.01	weathered crystalline rocks
9	420–520	$1.16 \cdot 10^{-13}$	0.0038	monolithic crystalline rocks

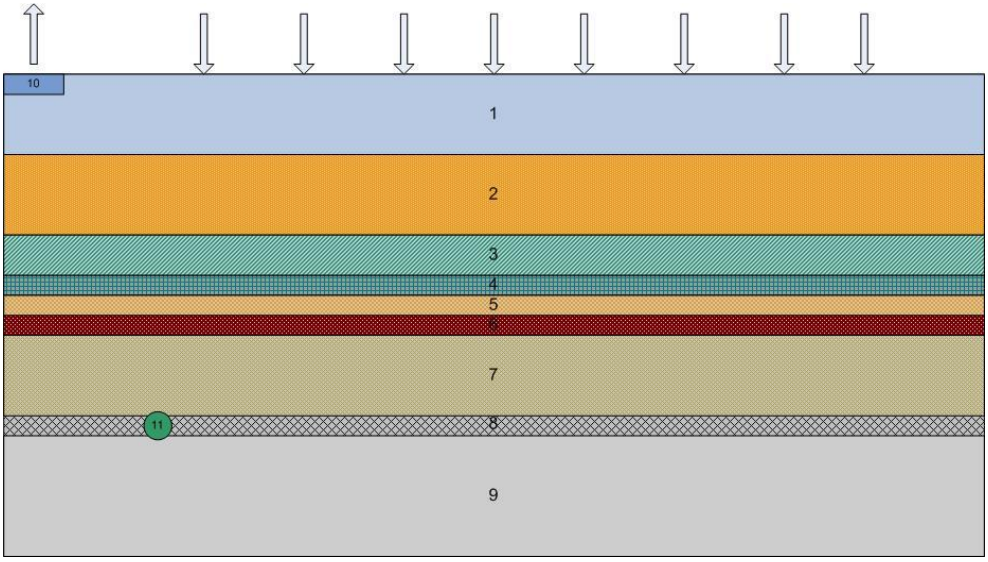

Fig. 6. Far field model (1 – glacial loam, 2 – sand, 3 – anhydrite, 4 – limestone, 5 – sandstone, 6 – clay, 7 – sandstone, 8 – weathered crystalline rocks, 9 – monolithic crystalline rocks, 10 – groundwater discharge area, 11 – radionuclide flux from the crystalline rock)

The results of ^{14}C release through the EBS in case of instant release from the graphite (Case D) and in 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:

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

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 Release 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.

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. The potential interaction with the backfill material is accounted by the linear sorption coefficient of K_d=0.2 m^3/kg.

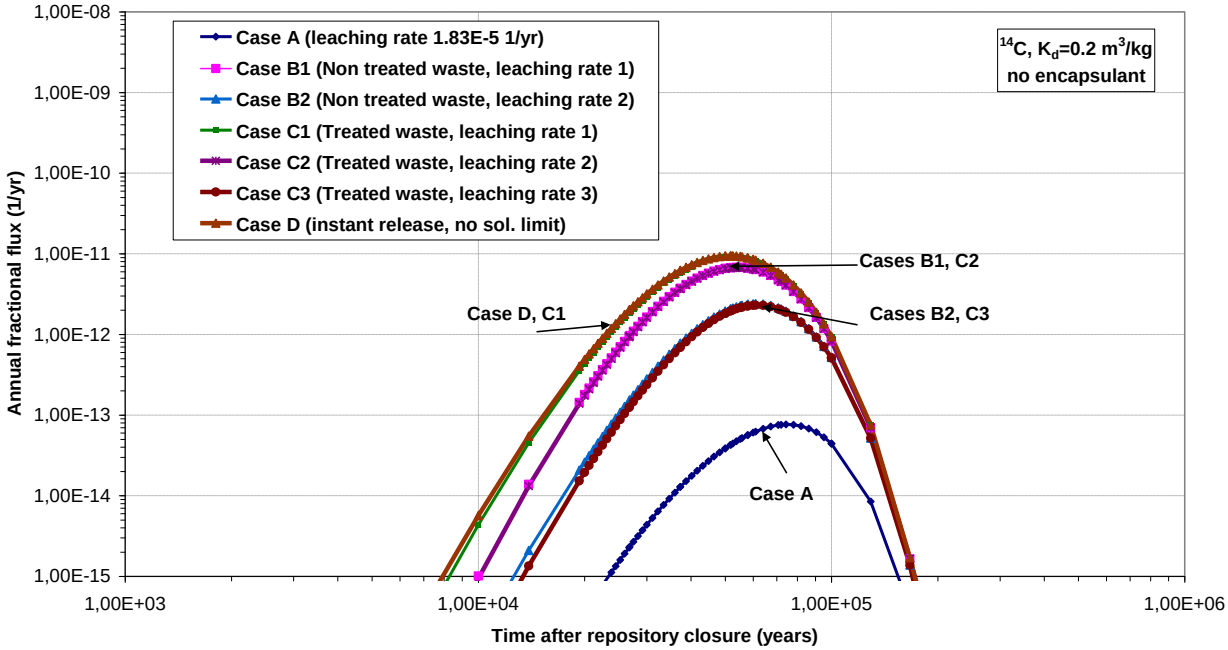

Fig. 7. Release of ^{14}C to the geosphere from one disposal container with non encapsulated waste

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 (Case A and Case D (instant release)) (Table 3). As it could be expected the lowest peak flux is observed in Case A, due to the lowest leaching rate. 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 indicated that the fractional flux profiles for cases B2 (non treated waste) and C3 (treated waste) differs only slightly. The profile of radionuclide flux to the geosphere in cases B1 and C3 is almost identical. Comparing the results of case C1 and D, where the instant release from the waste matrix is assumed, it was observed that the maximal flux to geosphere is not sensitive to the leaching rate of 0.1 1/yr vs. the assumption of instant release. In case of treated waste the maximal (peak) fractional flux differs by a factor of 4 while the leaching rate varies from 0.001 to 0.1 1/yr and the impact of this uncertainty on the maximal flux to geosphere from the EBS is limited.

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 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 (Case D) (~$5.2 \cdot 10^4$ yr) . For the evaluation of the prolongation 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 longer time period past closure. The comparison of maximal fluxes and its observation times is summarized in Fig. 8.

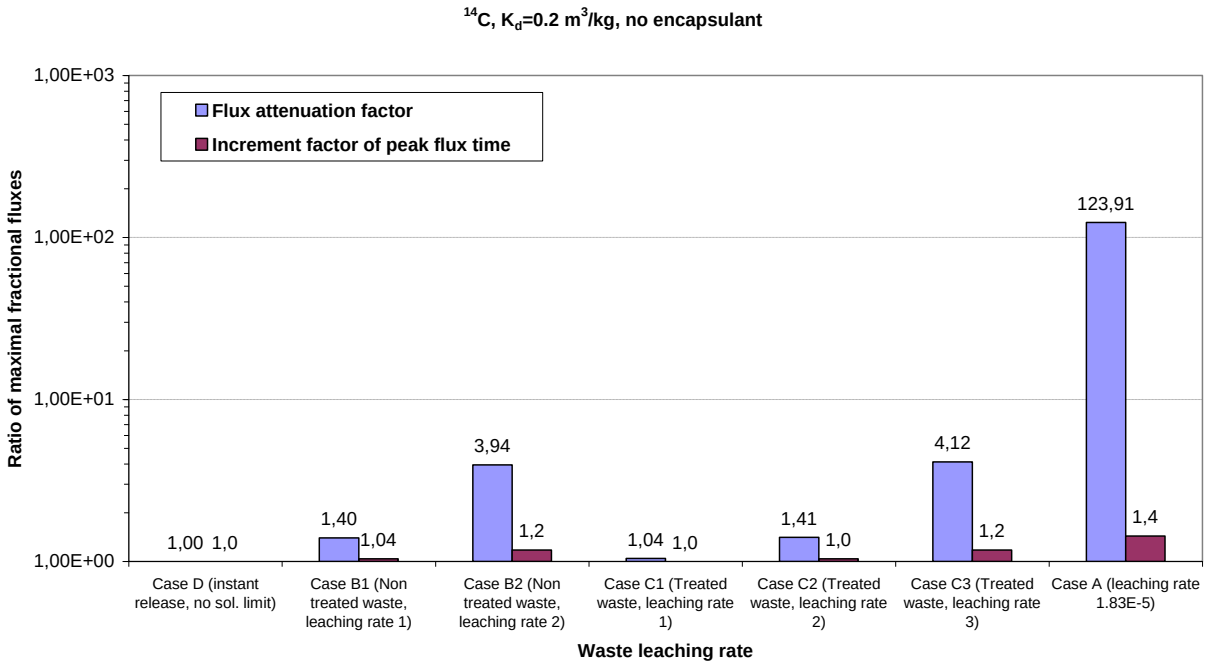

Fig. 8. Variations of peak fractional flux and time of maximal flux within the cases being analyzed

As it could be seen in Fig. 8 the most effective decrease of maximal flux is demonstrated if leaching rate is the order of 10^{-5} 1/yr. In this case the maximal flux to the geosphere would be

decreased by a factor of ~124. If the leaching rate is of the order of 10^{-3} 1/yr, the fractional flux would be lower by a factor of ~4. If the leaching rate is of the order of 10^{-2} 1/yr, the fractional flux would be lower by a factor of ~1.4. The reasoning of leaching rate of the order 10^{-1} 1/yr does not provide any significant benefit, as the fractional flux would be lower by a factor of ~1.04.

Time of peak flux is prolonged mostly in case of low leaching rate (Case A). In other cases it slightly increase (B2, C3), while in the rest cases is the same as in case of instant release.

Fig. 9 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 flux coming from the waste take places over the orders of magnitude.

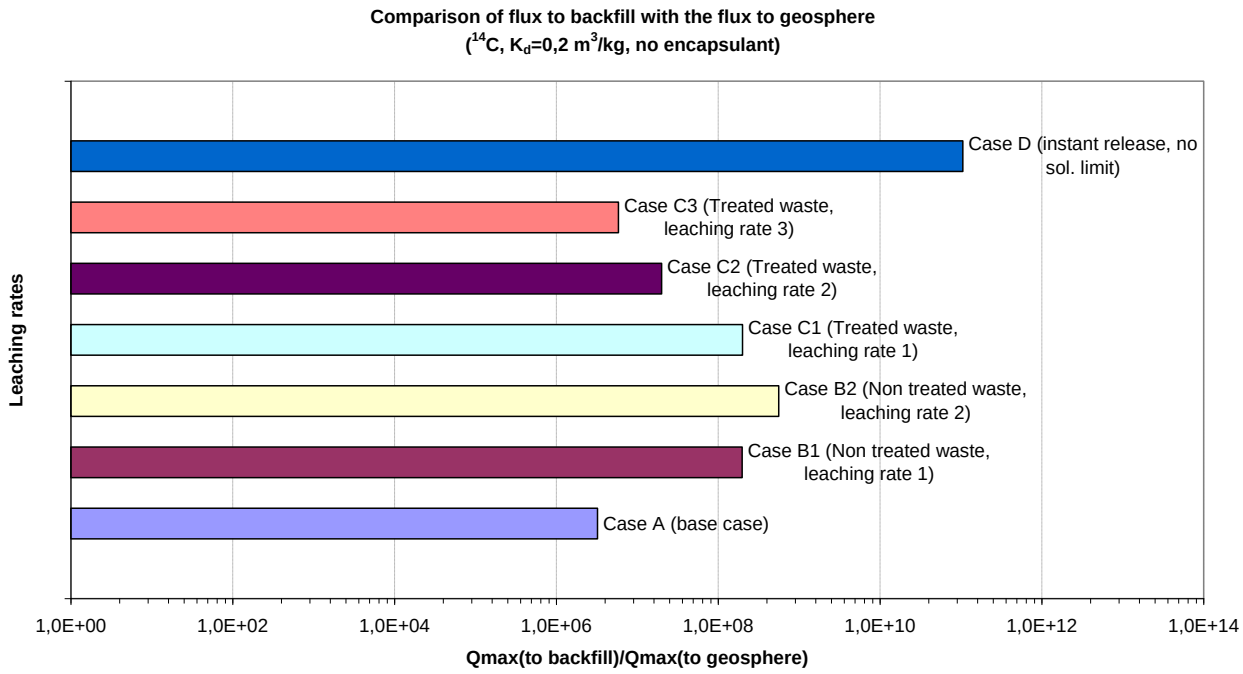

Fig. 9. Comparison of ^{14}C flux to the backfill and to the geosphere under different waste leaching rates

The highest ^{14}C fractional flux to the backfill was observed for the case of instant release from graphite (case D), when all inventory is released instantly to the backfill. However due to transport and interaction with a tunnel backfill material the radionuclide flux to the geosphere is decreased and the maximal fractional flux for different leaching conditions is in the range of $8 \cdot 10^{-14}$ - $9 \cdot 10^{-12}$1/yr. 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.

No sorption

The results of ^{14}C flux to the geosphere out of EBS disregarding the sorption in the backfill material are presented in Fig. 10.

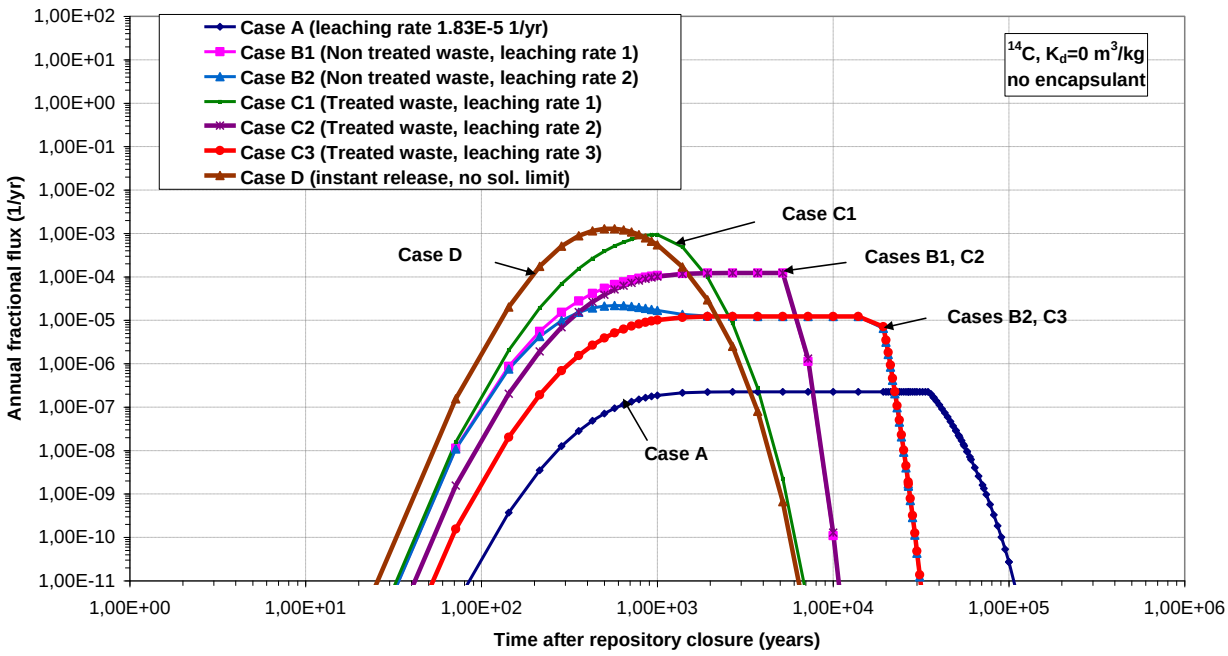

Fig. 10. Release of ^{14}C to the geosphere from one disposal container with non encapsulated waste (sorption disregarded)

If there is no interaction with backfill material the leaching rate importance is obvious. For the cases being analyzed the peak fractional flux varies within the rage of app. 4 order of magnitude ($1 \cdot 10^{-7}$ – $1 \cdot 10^{-3}$ 1/yr). For the cases of non treated waste (B1 and B2) the release profiles are identical up to app. several hundred years after the repository closure and starts differ later. The difference in the release rate in the plateau of annual fractional flux profile is of one order of magnitude and is directly related to the differences in the leaching rate profile (0.01 and 0.001 1/yr respectively). The time of peak flux is $5 \cdot 10^{3}$ yr past closure in Case B1, while in case B2 it is observed earlier ~570 years after the repository closure.

Differences of the potential leaching rate for the treated waste (Cases C1, C2, C3) are reflected in the differences of radionuclide flux profiles: the highest flux to the geosphere is governed by the highest leaching rate (0.1 1/yr). The time of peak flux is the shortest for case C1 as well.

Comparison of the time of maximal fractional flux under different leaching conditions showed that in cases D (instant release) and B2 (non treated waste, initial leaching rate of 0.1 1/yr, and subsequent constant release of 0.001 1/yr) the maximal release rate is observed before $1\cdot10^3$ years past closure. In the rest cases the maximal flux is estimated after $5\cdot10^3$ – $7\cdot10^3$ years past closure. As it could be expected the fastest transport of radionuclide occurs for case D, where the entire radioactive inventory is assumed to be available for instant release from irradiated graphite. Estimated maximal fractional flux to the geosphere depends on the leaching rate and varies within the range of $2\cdot10^{-7}$-$1\cdot10^{-3}$ 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. 11.

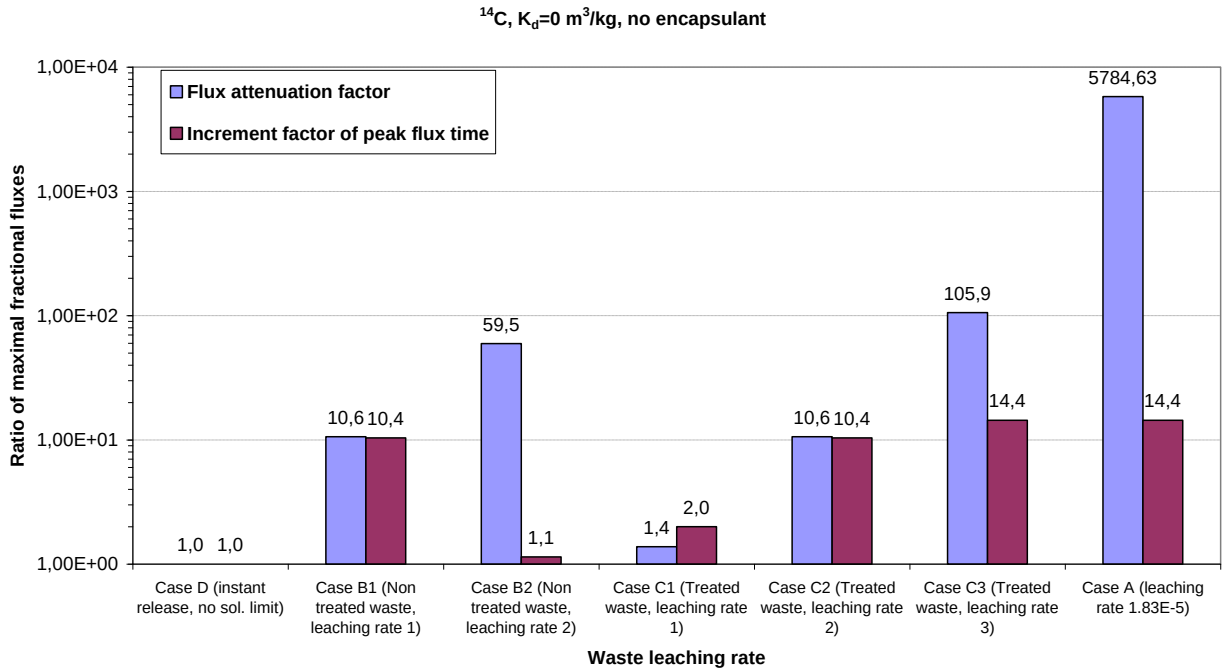

Fig. 11. Variations of peak fractional flux and its observation time within the cases being analyzed (sorption disregarded)

As it could be seen in Fig. 11 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 ~5800. If the leaching rate is of the order of 10^{-3} 1/yr, the fractional flux would be lower by a factor of ~100. However if the higher release occurs within short period (as in

Case B2) the maximal flux will be lower app. 60 times. If the leaching rate is of the order of 10^{-2} 1/yr, the fractional flux would be lower by a factor of ~10. The reasoning of leaching rate of the order 10^{-1} 1/yr does not provide a significant benefit, as the fractional flux would be lower by a factor of ~1.4. Time of peak flux is prolonged mostly in case of low leaching rate (Case A, Case C3) (by a factor of 14.4). In other cases it increases about 10 times (B1, C2), while in case B2 is almost the same as in case of instant release.

The modelling results presented and their comparison indicated the decreased importance of differences of leaching rate if the retention in the repository backfill occurs. 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. For example, in case B1 and C2 the flux from non treated granular form and treated granular form (see Table 3) of graphite show a similar behaviour and diminished impact of the higher release during the first 10 years.

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 capsulated waste. The maximal fractional flux in case of instant release (case D) is observed ~$6 \cdot 10^4$ years after the repository closure. The variations of peak fractional flux's time are slightly smaller in comparison to non encapsulated waste (Fig. 12).

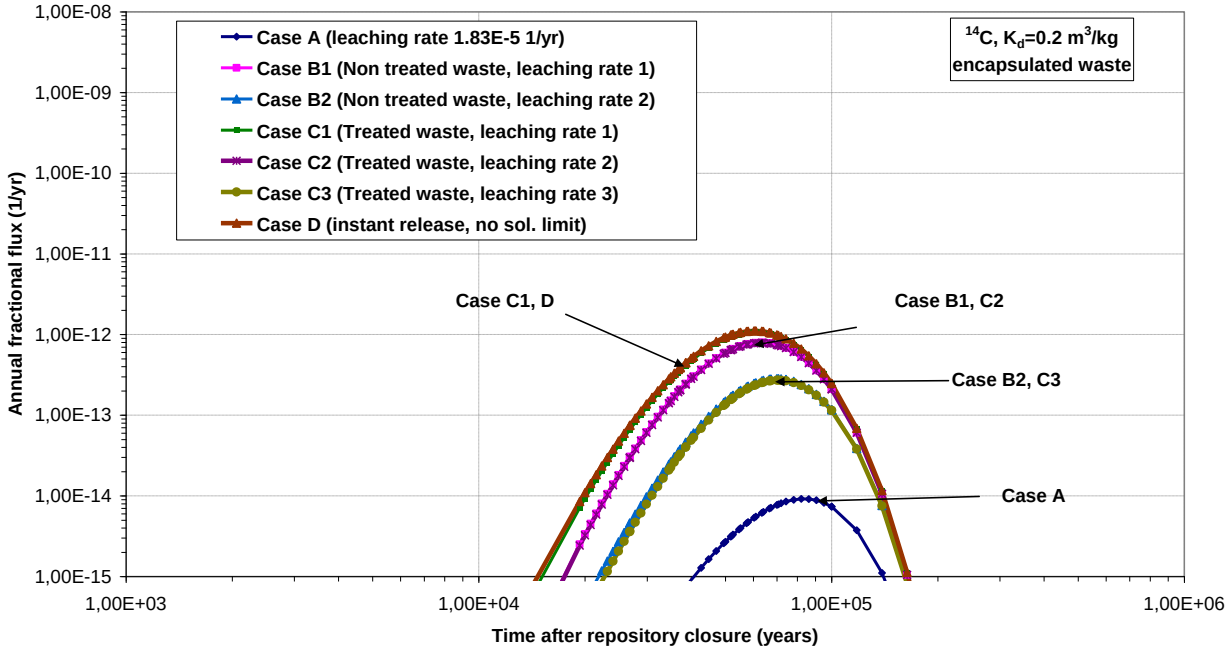

Fig. 12. Release of ^{14}C to the geosphere from one disposal container with encapsulated waste

Estimated maximal fractional flux to the geosphere depends on the leaching rate and varies within the range of $9 \cdot 10^{-15}$-$1 \cdot 10^{-12}$ 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. 13.

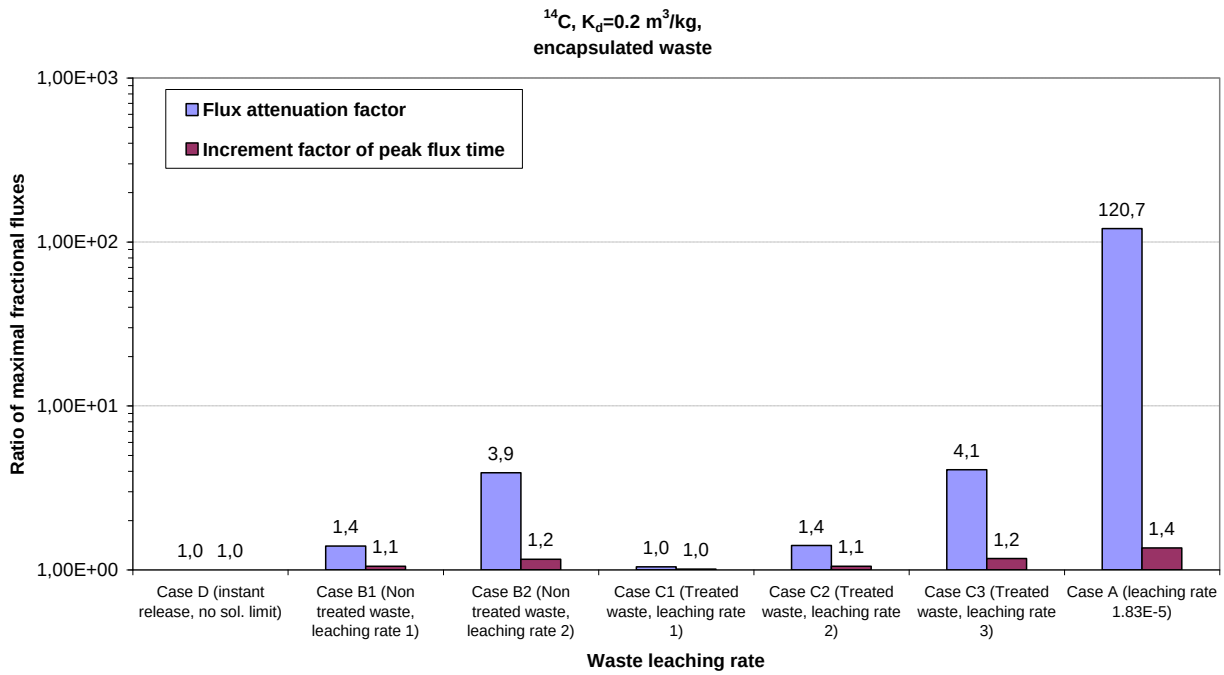

Fig. 13. Variations of peak fractional flux and its observation time within the cases being analyzed

As it could be seen in Fig. 13, 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 ~120. If the leaching rate is of the order of 10^{-3} 1/yr, the fractional flux would be lower by a factor of ~4. If the leaching rate is of the order of 10^{-2} 1/yr, the fractional flux would be lower by a factor of ~1.4. The reasoning of leaching rate of the order 10^{-1} 1/yr does not provide any significant benefit, as the maximal fractional flux is almost the same.

Time of peak flux is prolonged mostly in case of low leaching rate (Case A) (by a factor of 1.4). In other cases it increases about 1.2-1.4 times, while in case B2 is almost the same as in case of instant release.

No sorption

If no sorption 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. 10, Fig. 14).

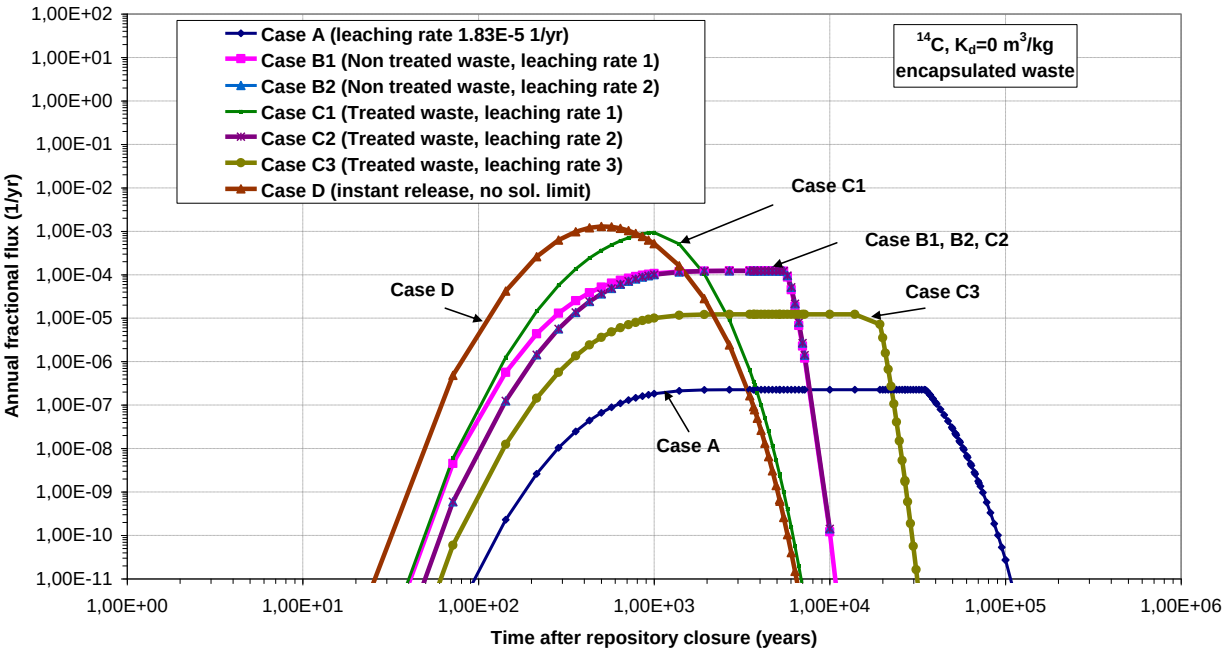

Fig. 14. Release of ^{14}C to the geosphere from one disposal container with encapsulated waste (sorption disregarded)

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.

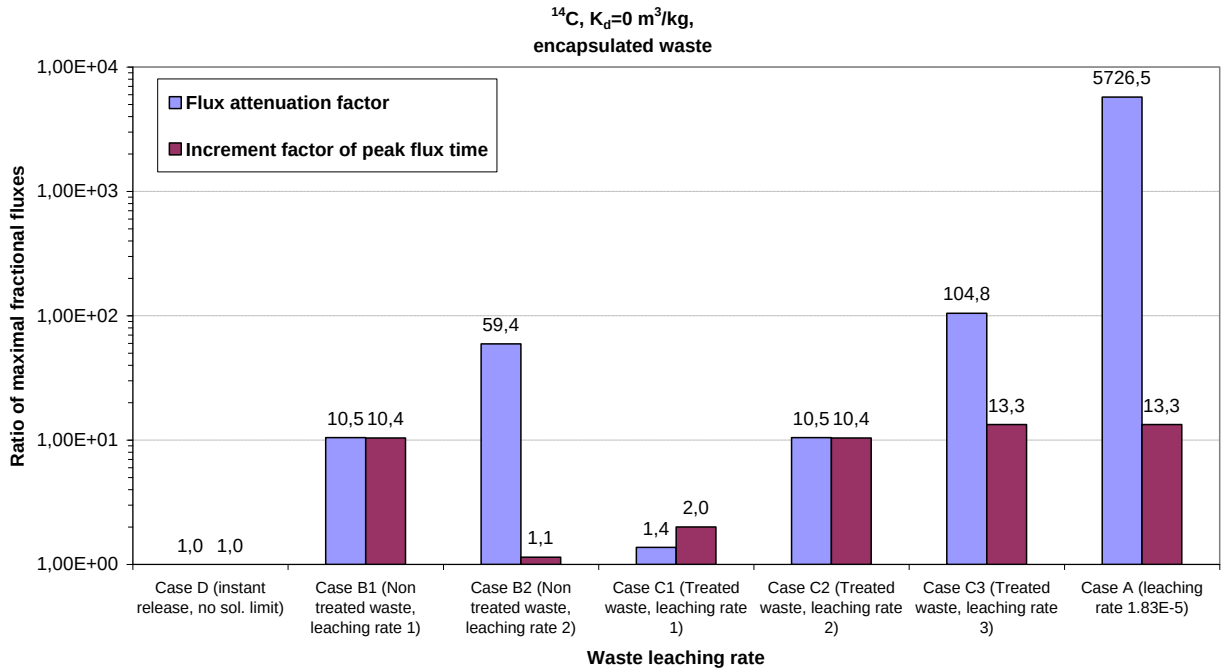

Fig. 15. Variations of peak release rate and time of peak release within the cases being analyzed (sorption disregarded)

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

<u>Comparison of the modelling results for not encapsulated and encapsulated waste</u>

In comparison to the release from the EBS to geosphere with the presence of encapsulant in the container, it was observed that the maximal release rate from the backfill to the surrounding geosphere is about 6-9 times lower in comparison of the modelling results do not considering the encapsulation (Fig. 16).

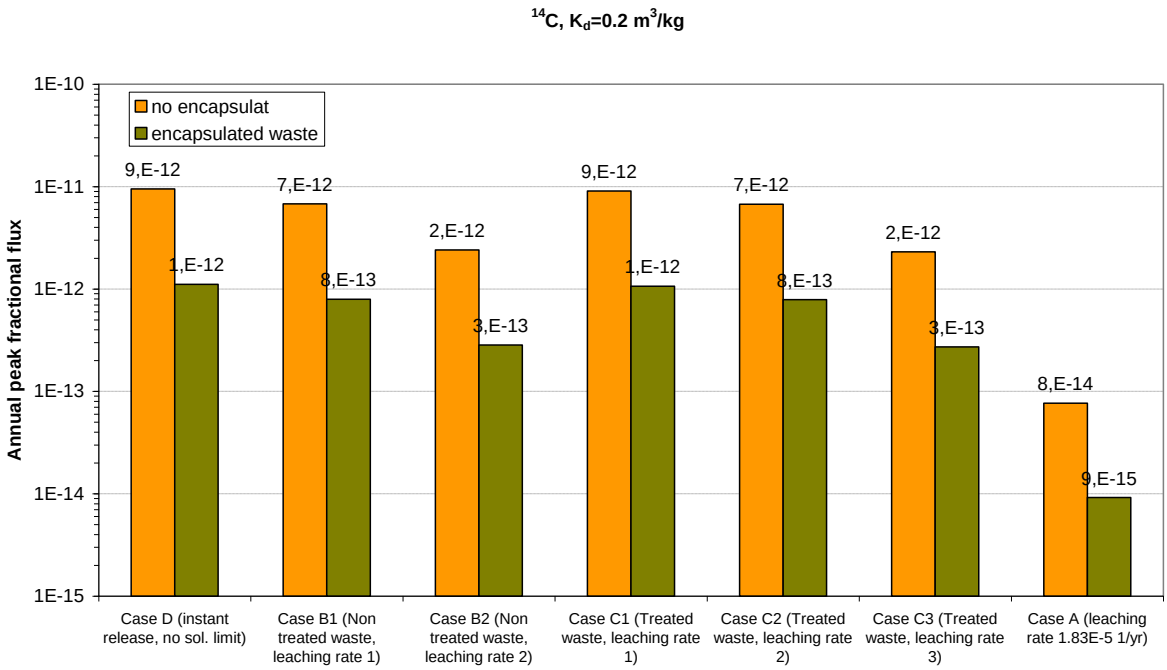

Fig. 16. Comparison of peak release for non encapsulated and encapsulated waste

As it has been mentioned above there is no credit taken for the sorption in the encapsulant and in the backfill material as well, the estimated maximum of radionuclide flux are very close to that of determined in the reference near field model (without waste encapsulation) (Fig. 17) The diffusive and advective transport through the encapsulant material does not result in a significant decrease of radionuclide flux to geosphere and its delay.

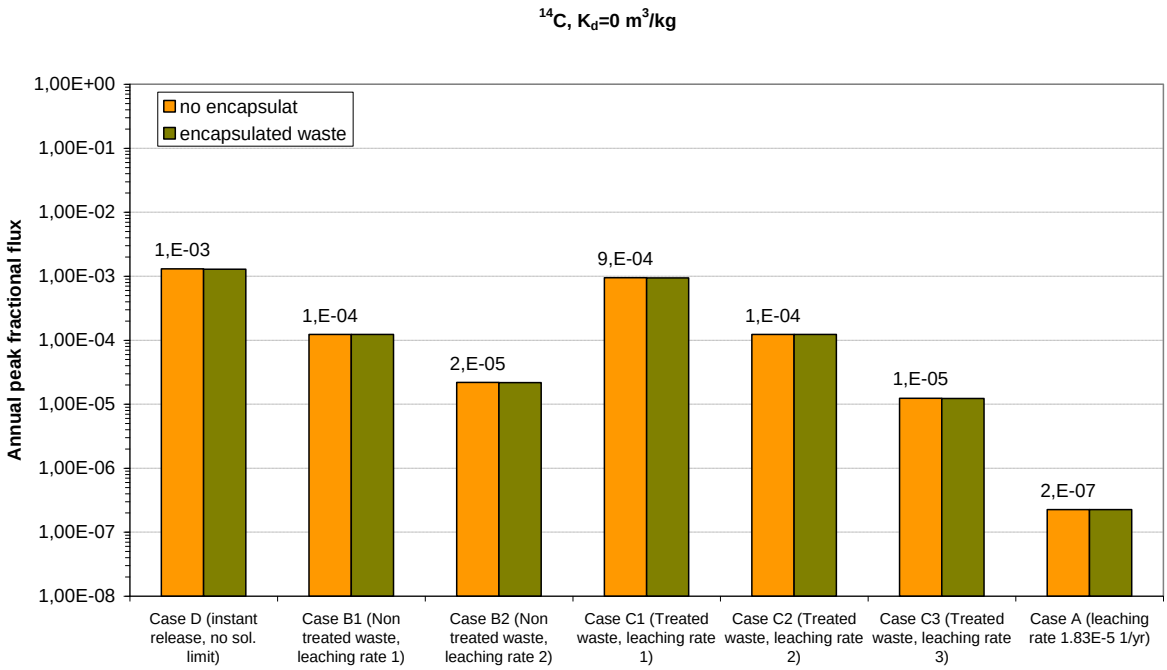

Fig. 17. Comparison of peak release for non encapsulated and encapsulated waste (sorption disregarded)

4.1.3 Release of ^{36}Cl

Chlorine release through the EBS from the waste matrix and considering no interaction with the environment as well no solubility limitation was modelled too. The leaching rate of $6.3 \cdot 10^{-2}$ 1/yr and instant release were assumed. The modelling results are presented in Fig. 18.

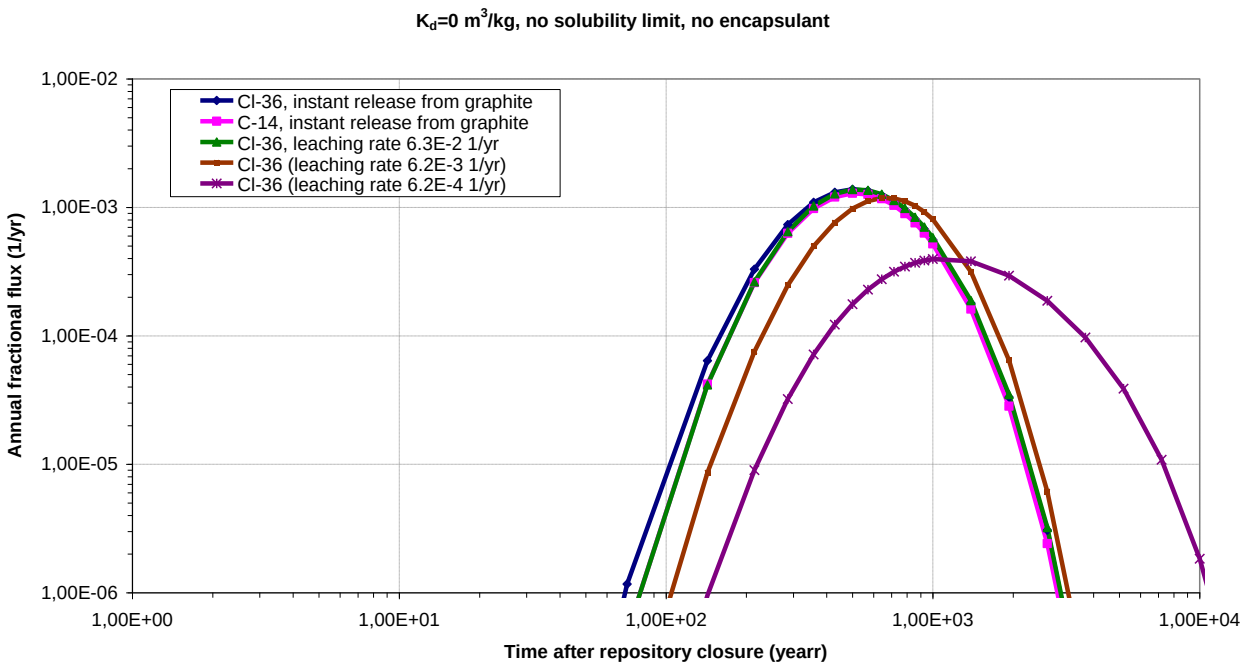

Fig. 18. Release of ^{36}Cl to the geosphere from one disposal container with non encapsulated waste (sorption disregarded)

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 published leaching rate of $6.2 \cdot 10^{-2}$ 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 $6.2 \cdot 10^{-2}$ decreased by 1-2 orders of magnitude and are presented in the Fig. 18. The results have showed that only in case of leaching of the order of $1 \cdot 10^{-4}$ 1/yr the flux to the geosphere becomes lower (by a factor ~3.4). This differs from ^{14}C where in case of leaching rate of the order of 10^{-2} 1/yr, the fractional flux would be lower by a factor of ~10 or in case of leaching rate of the order of 10^{-3} 1/yr, the fractional flux of ^{14}C lower by a factor of ~100. This is mainly attributed to the differences in the half-life of these nuclides. During ^{14}C transport in the EBS the process of radioactive decay contribute the decreasing the flux to the geosphere. In case of ^{36}Cl due to its much larger half-life, 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 played by the geological barriers around the EBS.

4.1.4 Summary

The results presented in section 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). In case of sorption of the 0.2 m^3/kg the peak fractional flux of ^{14}C dissolved in groundwater would vary 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. Comparison of the radionuclide flux (^{14}C) released to the backfill and from it indicates the backfill being of key importance as well. If the retention do not occur the differences in the leaching rate and its profile corresponds to the differences in the annual fractional flux profiles and its maximum rate. 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.

Thus the decreased importance of variations in leaching rates is indicated if the retention in the repository backfill could be demonstrated. Otherwise the needs of precise definition of leaching rates should be considered further according to the results of subsequent transport analysis in the geosphere.

The radionuclide transport analysis has been carried out considering the possible graphite waste encapsulation in the cement 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.

4.2 Release through the natural barriers

Release of ^{14}C in the far field (geosphere) was modelled using the computer code Petrasim [25] 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 and is transported to the geosphere. No interaction with the geological media assumed also. The simulations were performed a) assuming that ^{14}C from the repository is transported in liquid phase and b) all the inventory released is allowed to dissolve in the groundwater, become volatile and to be transported in two phases.

^{14}C transport through natural barriers in liquid form

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. 19 represents the annual fractional flux radionuclide ^{14}C from the geosphere (in term 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.

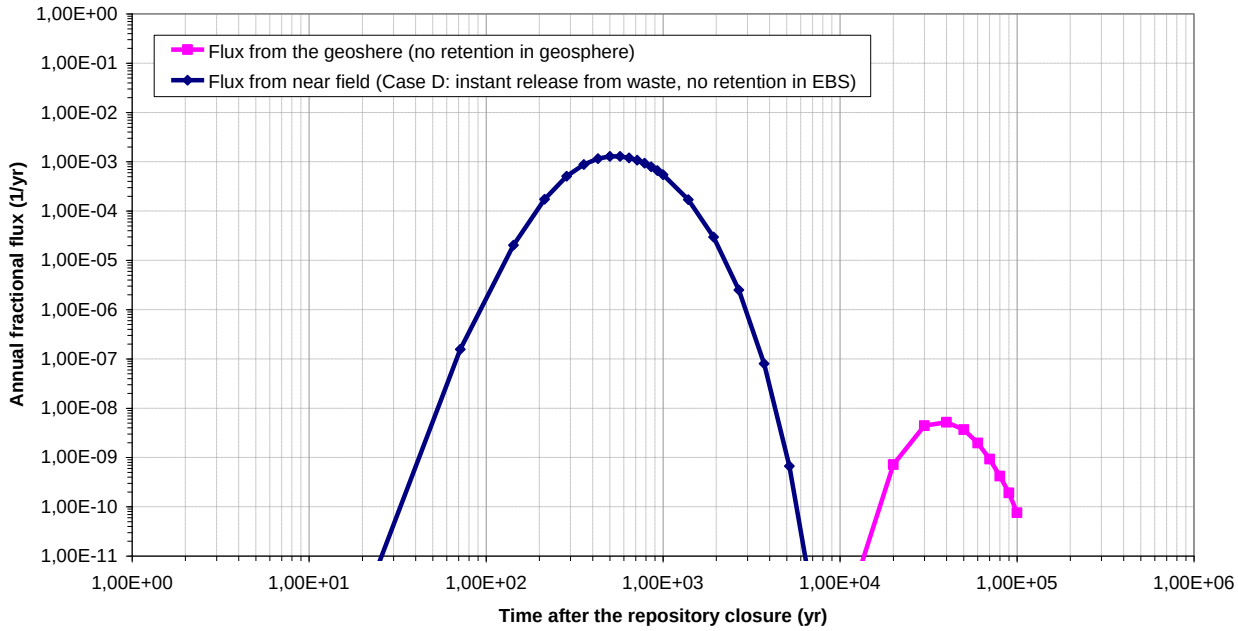

Fig. 19. Release of ^{14}C from the geosphere (instant release from graphite, no retention in the EBS)

As it could be seen in the figure, the system of natural barrier even disregarding the sorption in geologic environment do contribute 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 near field is decreased by app. 5 order of magnitude. Sorption on the geological barriers will lead to the lower flux than indicated in the Fig. 19 under the same flow conditions. Thus the importance of waste leaching rate depends 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). Thus in case of no sorption 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 from graphite waste becomes an important issue. Based

on the comparison of the releases from the near field in case of instant release (Case D) and leaching rate of $1.83 \cdot 10^{-5}$ 1/yr (Case A) the flux from the near field would be lower app. 5 orders of magnitude thus the flux from the geosphere is expected to be decreased at least by the same factor.

The distribution of dissolved radionuclide in the environment surrounding the repository is presented in Fig. 19 after 40 thousand years of repository closure.

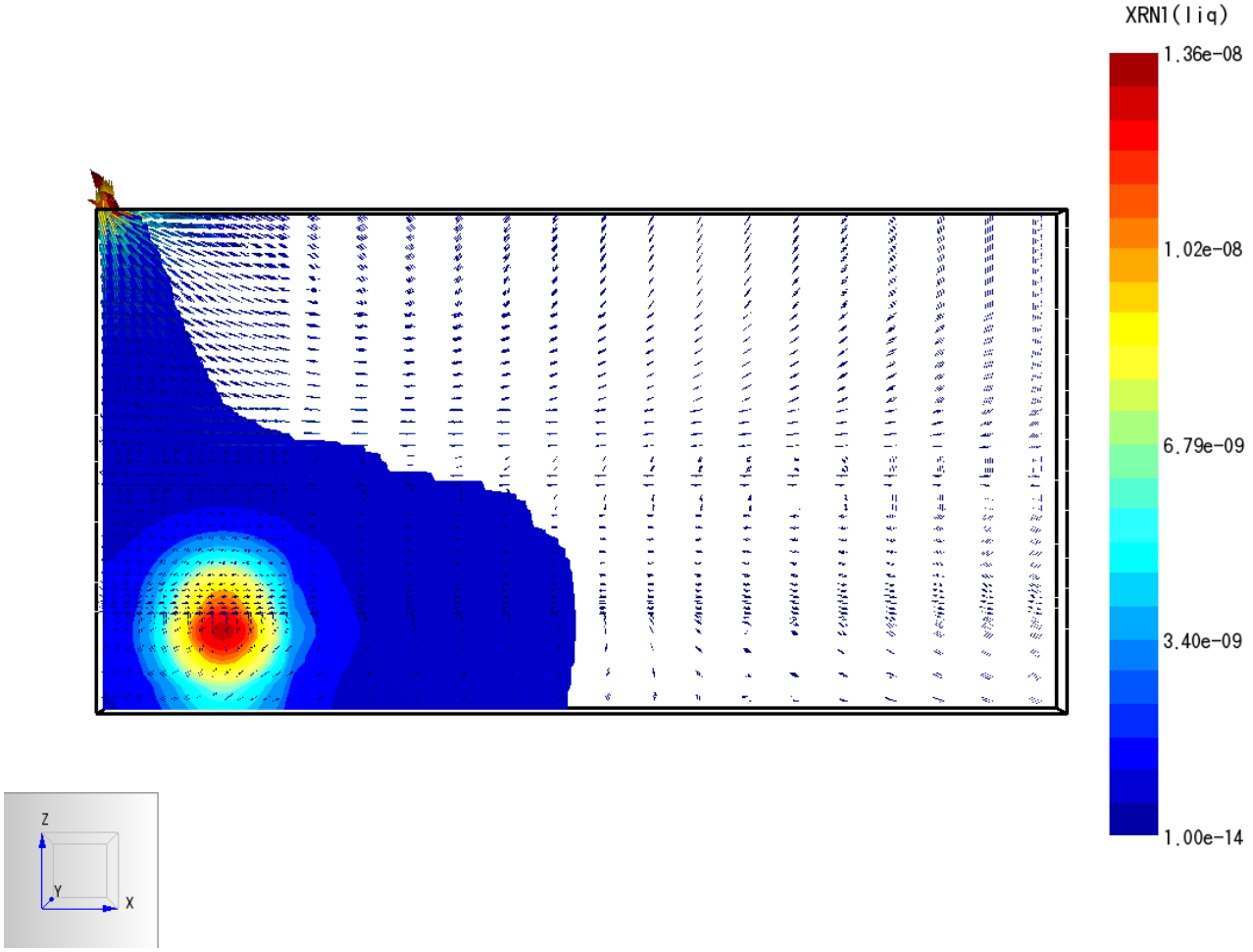

Fig. 20. Distribution of dissolved ^{14}C in the geological environment surrounding the repository (after 40 thousand years of repository closure)

As it could be seen there is radionuclide flux dispersion in the geosphere and it is distributed more in the direction of groundwater flow towards the surface.

<u>^{14}C transport through natural barriers in gaseous form</u>

The EOS7R module of software Petrasim (TOUGH2) allows performing the radionuclide transport analysis not only for dissolved fraction. In case of ^{14}C its transport as volatile radionuclide is also

possible. All inventory released from the near field was allowed to interact with the groundwater, dissolve there and later volatilize from the solution. For the evaluation of partial gas pressure according to Henry's law the inverse of Henrys' coefficient ($1 \cdot 10^{-8}$ Pa) was used. Based on scoping calculation free gas phase of ^{14}C is formed around the repository. The results in the Fig. 21 represent the region around the hypothetical repository where the rocks are not fully saturated by the groundwater due to free gas phase in it.

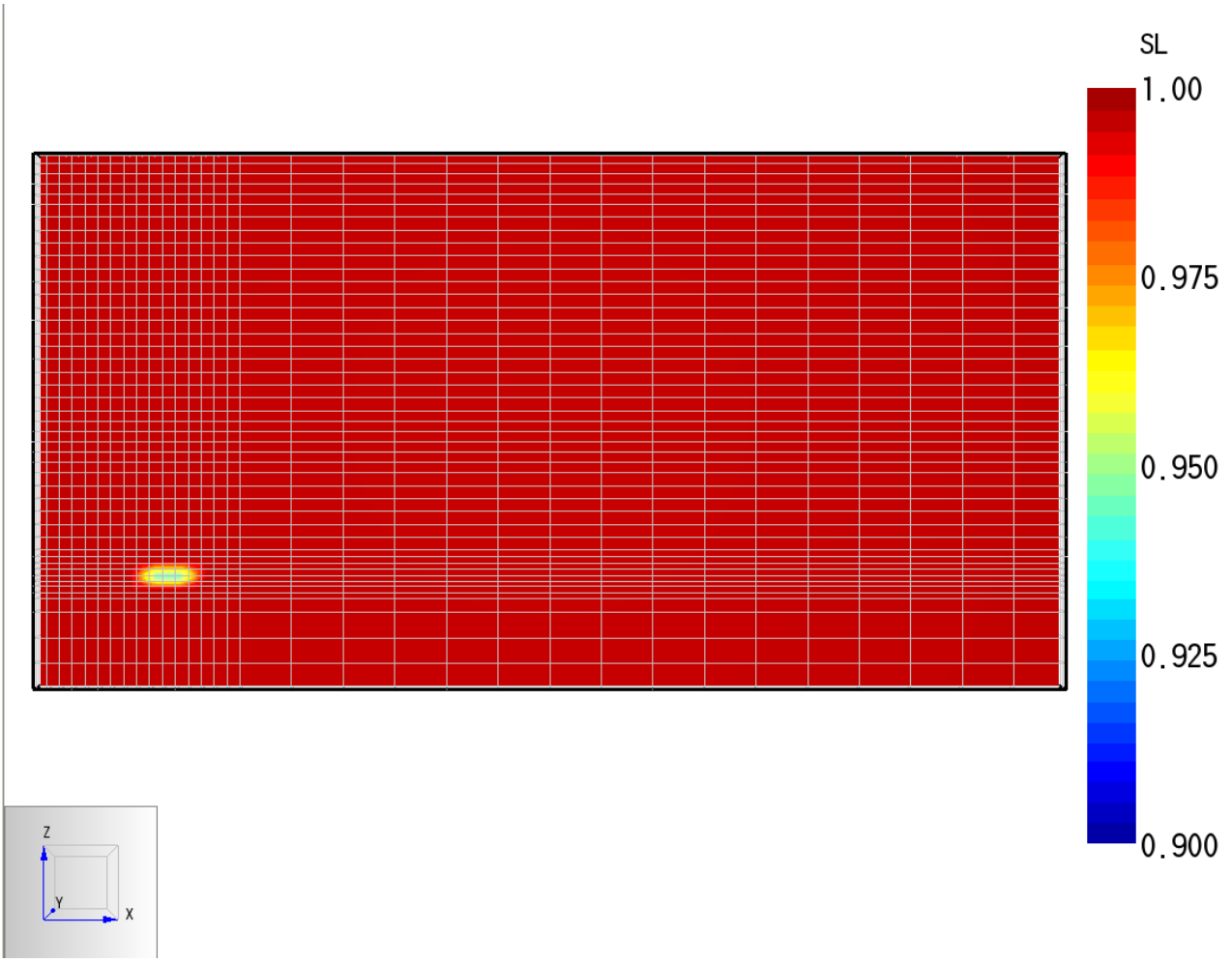

Fig. 21. Liquid saturation in the geological environment (SL<1 corresponds to the region with free gas phase) (after 40 thousand years of repository closure, when the maximal flux to the surface is observed)

Obviously the distribution of ^{14}C by continuous flow requires that considerable amount of it would be released to the environment and at sufficient rate to be enough to build up the pressure and form a continuous flow in gas phase.

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. Thus in case of gaseous release these constitutes to the preferential pathway for gas transport. 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. It depends on the leaching rate which is demonstrated (expected) for the particular option.

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. 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 significantly.

6 References

1. Final Decommissioning Plan for Ignalina NPP Units 1&2 A1.1/FDP/0004. Ignalina: Ignalina NPP DPMU, 2004.
2. Investigations of possibilities to disposal of spent nuclear fuel in Lithuania: a model case. Vol. 2. Concept of repository in crystalline rock, Vilnius, 2005, vol. 1.
3. Assessment of isotope-accumulation data from RBMK-1500 reactor. Technical Report T-3.4.2, CARBOWASTE project.
4. *Hacker, P. J.; Neighbour, G. B.; Levinskas, R.; Milcius, D.* Characterization of Ignalina NPP RBMK Reactors Graphite. Materials Science, 2001, no. 1, p. 62–66.
5. *Bondarkov, M. D.; et. al.* Activity Study of Graphite from the Chernobyl NPP Reactor. Bulletin of the Russian Academy of Science: Physics, 2009, no. 2, p. 261–265.
6. Report on UK waste packaging conception for irradiated graphite // NDA Deliverable T-6.3.4, 2011.
7. Generic Repository Studies. Generic Waste Package Specification. Volume 1. 2007 // NIREX report N/104 (issue 2), 2007.
8. Towler G., Penfold J., Limer L., Metcalfe R., King F. PSPA: Consideration of non-encapsulated ILW in the Phased Geological Repository Concept // Quintessa report, QRS-1378ZD-R1, December 2010.
9. Limer L., Smith G., Thorne M. Disposal of graphite: A modelling exercise to determine acceptable release rates to the biosphere // Quintessa report, QRS-1454A, 2010.
10. Why a cementitious backfill? // NIREX report No. N/034, 2001.
11. Suitability of geological environment in Lithuania for Disposal of spent nuclear fuel: Investigations of possibilities to disposal of spent nuclear fuel in Lithuania: a model case, Vilnius, 2005, vol. 1, 66 p.

12. Investigation of Lithuanian crystalline basement rock and alternative geological formation suitability for the construction of deep radioactive waste repository: Information report on the second stage. Vilnius, 2004, 16 p.
13. Water impregnation kinetics in nuclear graphite samples from UNGG reactors // CEA report Technical Report (WP 6 Task 1), 2010.
14. Behaviour of radionuclides in UNGG graphite in aqueous media // ANDRA&CEA Technical Report (WP 6 Task 1), 2011.
15. Carbon-14 in Low-Level Waste // EPRI report TR-107957, September 1999.
16. Behaviour of ^{36}Cl in irradiated graphite samples from UNGG G2 reactor – first results // CEA Technical Report (WP 6 Task 1), 2010.
17. Compilation of waste package concepts as envisioned for the ANTARES Project// AREVA Technical Report T-6.3.1, 2010.
18. Retention properties of cement for ^{36}Cl report // CEA report T6.3.2/D6.3.3, 2010.
19. SFR 1. Vault Database // SKI report 02:53, 2002
20. Report on model ^{14}C migration in gas phase in porous media // INR Technical Report T-6.4.2, 2011.
21. *Thorne M. C.* Development of Increased Understanding of Potential Radiological Impacts of Radioactive Gases from a Deep Geological Repository: Form of Release of C-14 // Mike Thorne and Associates Limited MTA/P0011b/2005-4: Issue 2.
22. Investigation of Alternative Waste Forms for GTCC ^{14}C Filters // EPRI report.
23. AMBER 4.4. Reference guide // Enviros report, 2003.
24. *Jakimavičiūtė-Maselienė, V.; Mažeika, J.; Petrošius, R.* Modeling of coupled groundwater flow and radionuclide transport in crystalline basement using FEFLOW 5.0. Journal of environmental engineering and landscape management, 2006, vol. XIV, no. 2, p. 101-112.
25. Petrasim 5. User manual // Thunderhead engineering, 2011.