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- OPERATIONAL MODELLING DESCRIBING RN MIGRATION OUT OF THE WASTE PACKAGES -

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Operational modelling describing radionuclides migration out of the waste packages

Executive summary

This report is produced under the framework of the Carbowaste European project for which the CEA/DEN/DTN/SMTM/LMTE proposed to carry out an operational model (MOP) for the release of radionuclides by waste packages designed to immobilize nuclear graphite.

After a reminder on the origin of French carbonaceous waste and on the nature of the waste packages, a synthetic point is made on the evolution of the operational modelling.

Then, the configurations and parameters currently used for calculations are pointed out.

The calculation results concerning the performances of containment for chlorine 36 are shown for each configuration. It is noted that the performances of the waste packages based on CEM V are higher than those of the waste packages based on CEM I, when one considers a stable healthy material or a material evolving in time. In addition, it is noted that the performances of the waste packages are degraded when one takes into account the decalcification of cementitious materials and also their early fractures.

Lastly, taking into account of parameters (porosity, diffusion coefficient, source term) obtained from measurements on irradiated graphite coming from GGNU G2 reactor has very little impact on the results.

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1 Introduction

In the framework of the Carbowaste European Project, the CEA/DEN/DTN/SMTM/LMTE proposed to carry out an operational modelling (MOP) for the release of radionuclides from the waste packages designed to immobilize nuclear graphite.

Nuclear graphite was used in France in the GGNU (Graphite Gas Natural Uranium) technology developed jointly by the CEA and EDF. Graphite was used as neutron moderator or reflector, like lining fuels. GGNU reactors being currently in the process of dismantling, the expected total volume of graphite elements to be evacuated is assessed to be 23 000 tons. The inventory of this waste shows that ^{36}Cl is present in small quantities in nuclear graphite, but this radionuclide constitutes a penalizing element because of its long radioactive half-life (301 000 years) and its high mobility in the natural environment.

After a reminder on the origin of French graphite waste and on the nature of the waste packages, a synthetic point is made on the evolution of the operational modelling. Initially, this modelling considered only healthy materials. Then the decalcification of constitutive cementitious materials of the waste packages was taken into account in order to better describe the package in disposal conditions. Early fractures of cement were then taken into account.

In order to have a comprehensive description of the data used in the operational modelling, the configurations and parameters currently retained for calculations are pointed out.

Lastly, we will presente the results for the modelling carried out with healthy cementitious materials, then with degraded materials and then with a material including early fractures.

Parameters (porosity, coefficient of diffusion, source term) resulting from measurements on irradiated materials coming from G2 reactors are then considered in the simulations.

2 The graphite waste

2.1. Origin of the nuclear graphite waste produced in France

As indicated above, graphite waste originate from the GGUN reactors which are now abandoned. These reactors belonged either to the CEA or EDF.

There were nine reactors, 6 EDF (Chinon A1, A2, A3, Saint-Laurent 1 and 2, Bugey 1) and 3 CEA (on Marcoule site, G1, G2 and G3). In addition, experimental reactors (EL2 and EL3) on Saclay site also have graphite elements.

2.1.1. The graphite waste from EDF

The graphite elements used as neutron moderator in the GGNU reactors are separated in two groups [1]:

The first group, or deconstruction waste, is made up by the elements constituting the framework of reactors, namely stacks, the reflectors and graphite from the surface support. The majority of these elements are massive blocks, with hexagonal form and variable length. They represent a total of 18,349 tons.

The second group, or exploitation waste, is made up by the elements associated with fuel claddings, such as sleeves and stacks, and it represents 3678 tons. The sleeves are hollow rolls with constant length and are stored in silos.

Figure 1 presents different graphite elements

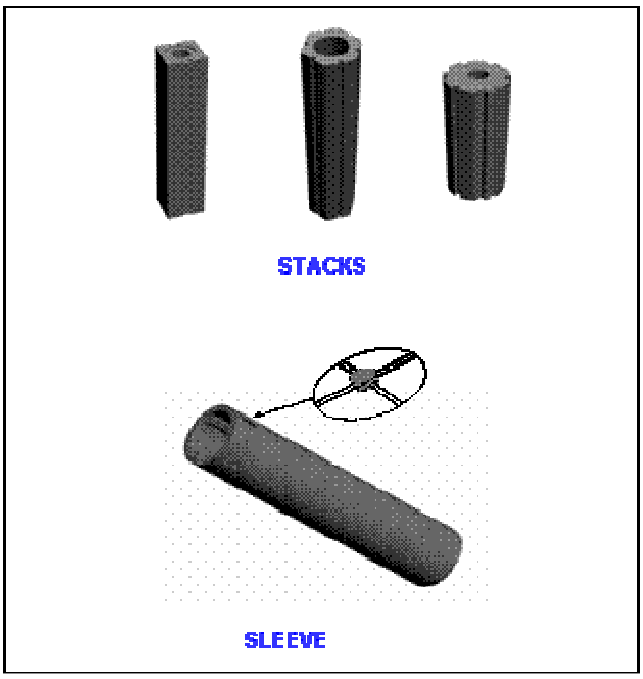

Figure 1: geometry of different graphite elements

2.1.1.1. Deconstruction Waste

Deconstruction Waste is consisted by stacks, reflectors and the elements provided radiobiological shielding located at the base of the reactor.

Stacks (figure 2), constituting the heart of the reactor, form a set of graphite columns formed by building block superposition. They are either prisms with hexagonal section (Saint-Laurent 1 and 2, Bugey 1 and Chinon A2 and A3 reactors), or prisms with square section (Chinon A1 reactor). For each reactor, stacks have different lengths, generally understood in the range of 400 - 1,500 mm. Only, stacks from Chinon A1 are prisms with square section of 200 mm by 200 mm.

The reflectors and the surface support graphite elements are in general massive parts with variable lengths and of size comparable with stacks.

Figure 2: example of stacks with hexagonal section

2.1.1.2. Exploitation Waste

These waste surround the uranium fuel element (sleeves). These elements are withdrawn during the unloading of fuel at the end of a three years period within the reactor. The sleeves are separated from the fuel element. Only the hearts can remain attached to the sleeves. The sleeves are currently stored on the two sites of La Hague and Marcoule and on the EDF site Saint-Laurent.

The sleeves stored on the sites of La Hague and Marcoule come primarily from the Chinon reactors. They are crushed until 1976 and separated from their heart. After this date, the sleeves were not any more treated this way and remained entirely intact.

From the conditioning and handling points of view, this differences concerning management thus brings to separate them in 2 groups because of very different level of cobalt activity:

- group 1 gathers the whole of the sleeves crushed and separated from their heart. The level of activity is of the same order of magnitude as that of deconstruction waste,
- group 2 gathers the intact sleeves stored on the site of Saint-Laurent. The level of activity is higher at least of one order of magnitude because of the presence of the cobalt 60 radionuclide.

Figure 3 presents a picture of a sleeve coming from the power station of Saint-Laurent.

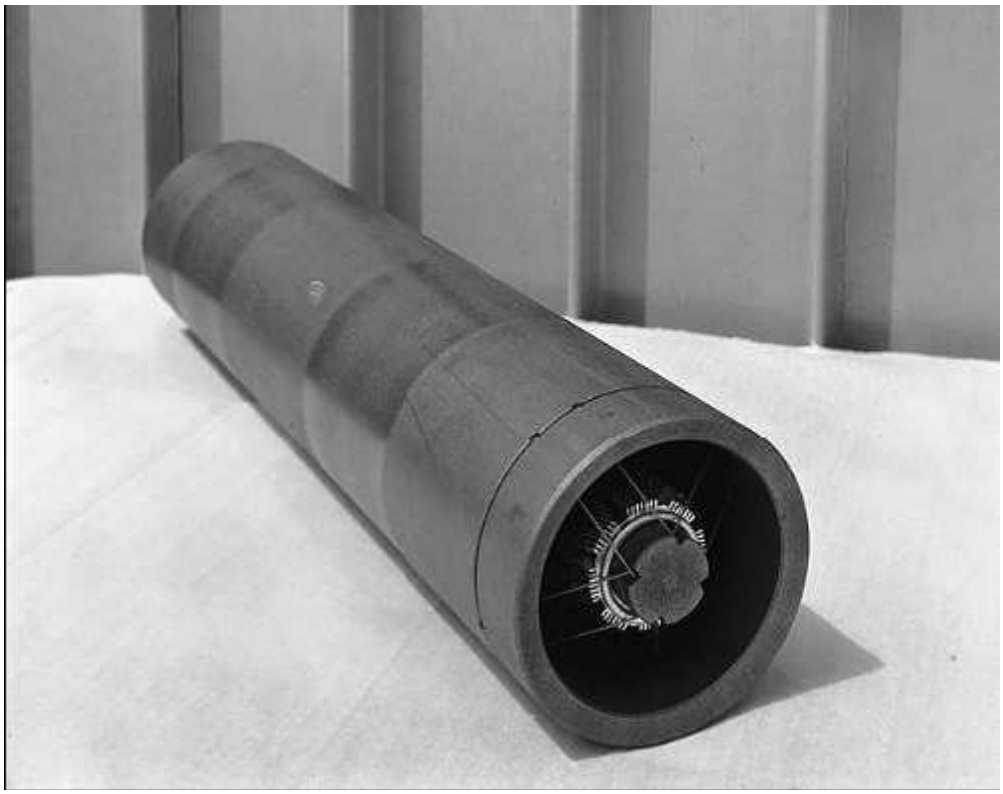

Figure 3: sleeve from Saint-Laurent reactor

2.1.2. The graphite waste from CEA

The shape of the graphite elements produced by CEA is comparable with those of the EDF elements, however dimensions can vary [2].

The CEA carried out in August 2007 an inventory as complete as possible of their graphite waste (and radiferous waste) intended for their specific storage [3]. These waste, from reactors G1, G2 and G3 (3804 tons), EL2 and EL3 (109 tons), Phoenix (59 tons), Rhapsody (12.2 tons), associated with those stored on the site of Marcoule (729.5 tons) or on the other sites of the Direction de l'Energie Nucléaire of the CEA (0.5 ton), represents a mass balance in graphite waste of 4 714.2 tons.

2.1.1. Quantitative inventory of the volume of waste

Total tonnage of graphite waste which will be sent on storage is thus estimated at approximately 23 000 tons.

2.2. Radiological inventory

The radiological inventory of graphite waste is essential to the good dimensioning, the storage components which will have the function to reduce the dose rate of the workers during the exploitation phase, and the elements providing a function of containment of the radionuclides during the phase of monitoring and post closing. Thus, many studies focussing on the radiological inventory of graphite elements coming from the sleeves and their heart and stacks, were carried out, either during series of direct measurements on real samples or estimates. These studies are listed in the document [4].

A first selection of the relevant radionuclides was carried out [5]. The selection criteria are the following:

- radionuclides whose radioactive half-life is higher than 30 years,
- radionuclides whose total activity is higher than 1 GBq,

- radionuclides whose total transfer time is considered higher than 100 000 years.

The application of these 3 criteria results in selecting only 6 radionuclides of interest: chlorine 36, carbon 14, calcium 41, silver 108m, molybdenum 93 and beryllium 10. Among those, chlorine 36 and beryllium 10 are those which have the higher contribution to the impact.

3 Waste package description

The storage package is composed of waste graphite coated with a mortar matrix and its concrete container. It constitutes the first confinement barrier.

The package considered in this study is a parallelepipedic object of 10 m^3 made of concrete in which a cart containing graphite waste is inserted. A schematic presentation is given in figure 4 where:

1) cart with primary graphite waste,
2) concrete structure containing the cart,
3) mortar matrix that fills the space between the cart and the concrete structure,
4) concrete cover.

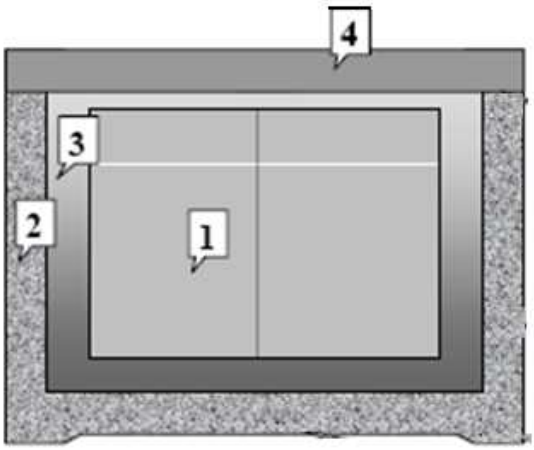

Figure 4: schematic view of the waste package

Currently, two concepts of package are considered. They primarily differ by the performance criterion relating to the long term radiological impact. Indeed, one can consider either that the package takes little part or not in the safety demonstration, taking into account the margins on

the other components of storage. Otherwise, one can also consider that it is an element taking part to the safety demonstration. The performances in time are thus different according to these two concepts, which leads to dimensional differences, but especially to differences in the choice of cementitious materials constitutive of the package. In the first case the concrete constituting the body of the package is rather based on CEM I while in the second case it will be preferentially based on CEM V.

Moreover, for each one of these two concepts, dimensions of the containers and the quantity of graphite waste conditioned in the cart vary, according to the radiological inventory of the graphite elements and thus of their origin.

3.1. Waste package based on CEM I concrete description

Various alternatives of packages based on CEM I concrete under consideration according to the origin of the graphite elements are described in table 1 [6]. One finds there informations concerning dimensions of the package, the mass of graphite waste and the number of packages to be realized.

Origin	Nature	Length (m)	Width (m)	Height (m)	Structure thickness (m)	Graphite per package (kg)	Waste package number
Bugey 1	Stacks	2.7	1.9	1.8	0.3	2 140	960
Bugey 1	Biological shieldings	2.8	1.9	1.8	0.2	3 300	160
Saint-Laurent A1	Stacks	2.7	2	1.8	0.2	3 990	650
Saint-Laurent A1	Biological shieldings	2.9	1.9	1.8	0.2	5 040	140
Saint-Laurent A2	Stacks	2.9	1.9	1.8	0.2	3 990	620
Saint-Laurent A2	Biological shieldings	2.7	2	1.8	0.2	4 670	170
Chinon A1	Stacks	2.7	2	1.8	0.2	5 020	230

Chinon A2	Stacks	2.7	2	1.8	0.2	4 480	500
Chinon A3	Stacks	2.7	2	1.8	0.2	3 140	810
Saint-Laurent disposals	Sleeves	2.7	2	1.8	0.3	2 890	700
Areva disposals	Sleeves	2.7	2	1.8	0.2	4 320	240

Table 1: list of various alternatives for waste packages based on CEM I concrete

The reinforced concrete constitutive of the container (body and cover) is a high performances concrete, F44 type, containing CEM I with silica fume and black steel reinforcements. The mortar used as stamps filling is containing CEM I and corresponds to that used for the packages intended for the Aube's site. Their compositions are given in table 2 [7].

	F44 concrete (kg)	Mortar (kg)
Cement (CEM I)	400	600
Water	131,5	260
Sand	746	1 450
Fine gravel	1 145	-
Silica fume	40	-
Additives	8.4 to 11.2	0 to 1.8
Black steel reinforcements	30	-

Table 2: concrete and mortar composition (to 1 m^3) for packages based on CEM I

One can note the existence of a metal grid (structure identical to that of the cart) positioned under the cover and intended to retain the graphite waste during the injection of the mortar and thus to limit the presence of supernatants [6]. This grid, located 10 cm under the cover, ensures

a 10 cm thickness of mortar between the top of the cart and the cover. It is represented on the sight of the package given of figure 5.

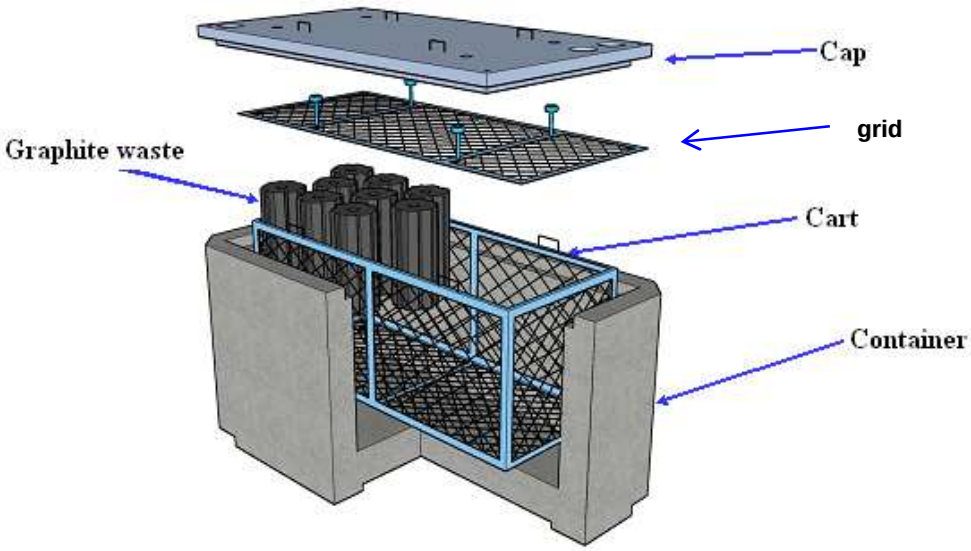

Figure 5: waste package based on CEM I concrete concept

For this F44 concrete formulation, measurements of diffusion coefficients with tritium water realized on laboratory scale samples led to a very low value, equal to $5.8.10^{-14}$ m ² /s. In order to take into account the various phenomena met during the manufacture of an industrial object (for example the early fractures), this value was voluntarily degraded by an order of magnitude to be brought back to 5.10^{-13} m ² /s.

For the mortar, the value of the specified diffusion coefficient obtained by tritium diffusion (5.10^{-12} m^2/s) is an estimate (i.e not measured value) based on data from the literature.

3.1. Waste package based on CEM V concrete description

The various alternatives of these packages based on CEM V concrete, according to the origin of the graphite elements, are described in table 3 [6]. One finds in this table informations concerning dimensions of the packages, the mass of graphite waste and the number of packages to be realized.

Origin	Nature	Length (m)	Width (m)	Height (m)	Structure thickness (m)	Graphite per package (kg)	Waste package number
Bugey 1	Stacks	2.7	2	1.8	0.3	2 130	960
Bugey 1	Biological shieldings	2.9	1.9	1.8	0.2	3 140	170
Saint-Laurent A1	Stacks	2.8	1.8	1.8	0.2	3 280	790
Saint-Laurent A1	Biological shieldings	2.8	1.9	1.8	0.2	4 430	160
Saint-Laurent A2	Stacks	2.7	2	1.8	0.2	3 690	670
Saint-Laurent A2	Biological shieldings	2.7	2	1.8	0.2	4 410	180
Chinon A1	Stacks	2.7	1.8	1.8	0.2	2 750	410
Chinon A2	Stacks	2.7	2	1.8	0.2	3 170	700
Chinon A3	Stacks	2.8	1.8	1.8	0.2	2 600	980
Saint-Laurent disposals	Sleeves	2.7	2	1.8	0.3	2 600	770
Areva disposals	Sleeves	2.7	2	1.8	0.2	3 940	260

Table 3: list of various alternatives for waste packages based on CEM V concrete

The concrete constitutive of the packages body is made of CEM V cement coming from the factory Calcia Airvault and with stainless steel fibres. The mortar is also containing CEM V cement. It was formulated by the “Materiaux and Durabilité des Constructions” Laboratory (LMD) of the INSA Toulouse in order to optimize the processing, the retention of chlorine 36 and the effective diffusion coefficient [8]. The compositions of these two hydraulic binders are given in table 4.

	Concrete (kg)	Mortar (kg)
Cement (CEM V)	459	600

Water	197	190
Aggregates (Boulonnais)	1 637	-
Sand	-	1 290
Silica fume	25	60
Additives	14,5	12
Stainless steel fibres	86	-

Table 4: concrete and mortar composition (to 1 m^3) for package based on CEM V

4 Operational modelling (MOP) « graphite waste package »

Initially the operational modelling takes into account only healthy constitutive materials.

For the development of the MOP, the generalized transport code PORFLOW [9] developed by the ACRi company was selected because this numerical tool makes it possible to deal with multidimensional problems of migration in saturated or unsaturated porous environments in systems with complex geometries.

4.1. MOP with healthy materials

Initially, the operational modelling considers only materials with constant properties in time, for the concrete and mortar as well for graphite waste itself [10], and this, throughout all the storage life.

In this model, the transport of the radionuclides in materials constituting the package is described using the following traditional equation (Equation 1):

$$\frac{\partial(\omega \cdot R_{RN} \cdot C_{RN})}{\partial t} = div\left(D^{eff} \cdot \overrightarrow{grad} C_{RN}\right) - \lambda_{RN} C_{RN} \qquad (1)$$

where:

D^{eff} : effective diffusion coefficient for aqueous species (m^2/s),

R_{RN} : retardation factor for radionuclide ($R_{RN} = 1 + \frac{1-\omega}{\omega} K_{dRN} \cdot \rho_s$ where K_{dRN} is partition coefficient (m^3/kg solid phase) and where ρ_s is the volumic mass of solid phase (kg/m^3 solid phase)),

C_{RN} : solution radionuclide concentration (mol/m^3),

λ_{RN} : radioactive half-life (s^{-1}),

ω : porosity.

It is considered that the radionuclides are present in the porosity of the graphite matrix and entirely dissolved when this one is put in contact with water. The radionuclides are then released by diffusion through graphite. This model requires to know the radiological inventory and the transport properties of the matrix: diffusion coefficient, porosity, and partition coefficient (Kd) if the sorption of the element on graphite must be taken into account.

4.2. MOP with decalcificated cementitious materials

The Diffu-Ca model is an operational model making possible to describe, in a simplified way, the chemical degradation of cementitious materials containing CEM I or CEM V cement [11]. In this model, it is considered that decalcification is the central phenomenon of the degradation and that decalcification itself is controlled by the evolution of the calcium concentration in the interstitial aqueous solution of materials. Consequently, the chemistry of the system is simplified so as to express the major parameters (porosity, solid calcium concentration, diffusion coefficient, etc) according to the aqueous calcium concentration. Besides decalcification, the model also describes the diffusion of the radionuclides through cementitious materials, this migration itself being influenced by the evolution of materials.

The version of Diffu-Ca used for this study only considers diffusion phenomena in saturated media.

4.2.1. Cementitious materials description

In a general way, the cementitious materials are regarded as composed of:

- portlandite, noted CH, with volumic fraction V_{CH},
- CSH or calcium silicate hydrates, with volumic fraction $V_{C\text{-}S\text{-}H}$,
- ettringite, noted Ett, with volumic fraction V_{Ett},
- aggregates, with volumic fraction V_{Gra},
- saturated porosity (ω) , with a volumic fraction $\omega = 1 - V_{CH} - V_{C\text{-}S\text{-}H} - V_{Ett} - V_{Gra}$.

4.2.2. Decalcification modelling

The description of decalcification is based on a simplified relation between the solid phase and the aqueous calcium concentrations, respectively noted S_{Ca} and solution C_{Ca}. From there results a description of the evolution of the volumic fractions of various minerals, and thus of porosity. Decalcification is described while being based on the following assumptions:

- the portlandite (CH) dissolves with a linear trend. This dissolution takes place as soon as aqueous calcium concentration C_{Ca} is lower than 22 mol/m^3 and is considered total when C_{Ca} hasreached 21 mol/m^3;
- when the portlandite is entirely dissolved (i.e. when $C_{Ca} < 21$ mol/m^3), the CSH start to evolve. It is considered that this decalcification is done without significant opening of porosity;
- the progressive dissolution of the ettringite occurs concurrently to the decalcification of the CSH. It is regarded as total when C_{Ca} reaches 2 mol/m^3.

The evolution of porosity according to C_{Ca}, concentration of calcium in solution, results from what precedes. Its characteristics are represented in figure 6.

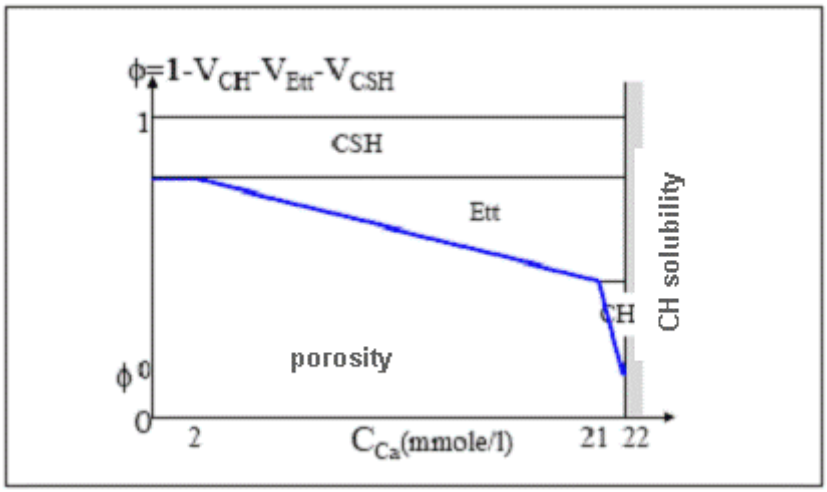

Figure 6: evolution of the porosity of a cement paste according to calcium concentration in solution [11]

In the same way, the evolution of the calcium concentration in solid phase results from the previous observations. This concentration is expressed as a function of the ratio (C/S) (or S_{Ca}/S_{Si}, i.e. the ratio of the calcium concentration in solid phase on the silicon concentration in solid phase) and its typical evolution is represented in figure 7.

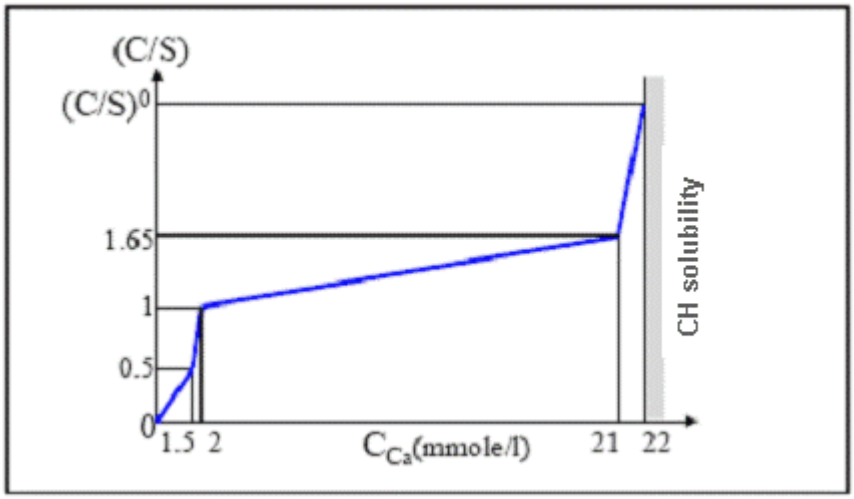

Figure 7: evolution of ratio (C/S) for a cement paste according to calcium concentration in solution [11]

4.2.3. Transport equations

A second equation (Equation 2) is added to the traditional transport equation for radionuclides used in the "healthy materials" modelling, in order to represent the calcium diffusion:

$$\frac{\partial(\omega \cdot C_{Ca} + S_{Ca})}{\partial t} = div\left(D^{eff} \cdot \overrightarrow{grad} C_{Ca}\right) \quad (2)$$

Where C_{Ca} and S_{Ca} are respectively calcium concentrations in the liquid and solid phase (mol/m^3).

It is noted that in these equations, the terms ω and S_{Ca} are linear functions per part of the C_{Ca} variable, specified according to the properties of cementitious material.

In addition, the effective diffusion coefficient in cementitious materials evolves with porosity. Various relations were proposed in the literature to connect these two parameters (law of Archie, exponential law, etc) [11, 12]. For materials based on CEM I, the following law (Equation 3) is retained in the Diffu-Ca model:

$$D^{eff} = D_0 * \exp(\omega / \omega_r) \quad (3)$$

where D_0 and ω_r are constants. This law was obtained by tritium water diffusion experiments with tritium water on healthy materials whose various values of porosity were obtained while ratio water / cement was varying.

One will note that for materials based on CEM V cement, this relation is not valid anymore and is replaced by successive constant values of D^{eff}, according to C_{Ca} [13].

4.3. Taking into account the early fractures

Early fractures are prejudicial for the durability of the concrete structures because they can cause a deterioration of the mechanical properties and because they can support the penetration of aggressive external agents.

4.3.1. Origin of early fractures

Four causes are at the origin of early fractures [14]: segregation, plastic shrinkage, thermal shrinkage after water intake and auto-desiccation shrinkage.

Segregation

Segregation is the surface exudation of a part of the mixing water to the higher face of the freshly-mixed concrete. It is a phenomenon which appears before the water intake, in relation to a progressive compressing of the skeleton under the gravity effect. This vertical deformation can be important (a few percent) and accompanied, in the extreme cases, by creation of open cracks.

Plastic shrinkage

It is an exogenous shrinkage due to the desiccation of the concrete which appears before and during the water intake. This deformation is limited in time and appears only when exposed surface is free from water of segregation.

Thermal shrinkage

The water intake of cement is accompanied by a heat emission and a heating of the concrete. After the water intake, the heat emission slows down and the concrete cools down. The thermal shrinkage that occurs after water intake is the contraction of the concrete due to this cooling. This phenomenon occurs of a few tens of hours at a few weeks after the water intake and varies with the dimension of the parts.

Shrinkage due to auto-dessication

This shrinkage originates in the difference in volume between water and initial cement on the one hand and the formed hydrates on the other hand. The least volume of the formed hydrates

causes a contraction phenomenon. This contraction increases when the ratio water /cement decreases. So this phenomenon is more important for the concretes with high efficiencies.

4.3.2. Evaluation of early fractures

There are no standard rules imposing a maximum depth or a maximum opening of early fractures for the civil engineering facilities. Also, a literature study is realized to search the level of early fractures which it is possible to note for works of the size of the waste package. Collins and Sanjayan [15] give a report on presence of microscopic cracks going up to 0.3 mm of depth for samples of small sizes (10 cm length cylinders for 6 cm in diameter) of mortar. In addition, Jensen and Chatterji [16] give a report on relations between the depth (p) and the opening (or diameter d) of the cracks on the one hand, and the width (L) of the structures considered on the other hand:

- $d / L = 5.10^{-5}$
- $p / d = 2\ 000$
- what gives : $p / L = 0,1$

These relations, in dissension with the observations of Collins and Sanjayan [15], would result in considering depths of fractures of about 2 or 3 cm, according to the configuration, with openings of cracks of about 10 or 15 microns respectively.

The analysis of these documents can lead us to define two types of early fractures: the first one (type 1) being realistic and the second one (type 2) being overestimated:

- cracks separated by 4 cm, 2 cm depth and 350 µm diameter, noted type 1 (40, 20, 035),
- cracks separated by 2 cm, 4 cm depth and 700 µm diameter, noted type 1 (20, 40, 070).

5 Various configurations and various parameters retained for simulations

In this chapter we will summarize the principal data relative to the standard waste package, materials and configurations of calculations used in this document to model the release of the radionuclides from the graphite waste package.

5.1. Selected waste packages

As indicated previously, the concept of graphite waste package is always under investigation and the technical selected options differ according to the nature of the graphite elements. Consequently, a choice of the most representative packages was carried out, while being based on the following criteria:

- an important number of packages to be produced,
- choice of a packages based on CEM I concrete and a packages based on CEM V concrete.

The following packages were considered for this study:

- package based on CEM I concrete for stacks from Chinon A3,
- package based on CEM V concrete for stacks from Chinon A3.

The general characteristics of the selected packages, resulting from document [6], are given in the following table.

	Length (m)	Width (m)	Height (m)	Structure thickness (m)	Cover thickness (m)	Graphite per package (kg)	Waste package number
CEM I							
Stacks from Chinon A3	2.7	2	1.8	0.2	0.17	3140	810
CEM V							
Stacks from Chinon A3	2.8	1.8	1.8	0.2	0.2	2600	980

Table 4: selected packages characteristics

The representation within the space of package is simplified. Thus, the following assumptions are made:

- the package is compared to a right-angled parallelepiped,
- one considers a constant thickness of mortar between the cart and the body out of concrete. The thickness is fixed at 5 cm for the sides and the low part of the package. For the high part, the radiobiological shielding constraints impose a 10 cm thickness between the top of the cart and the cover,
- the package is regarded as symmetrical, calculations are thus carried out by considering a vertical half-section in the width direction.

The geometrical configurations of the half-packages are represented figure 8.

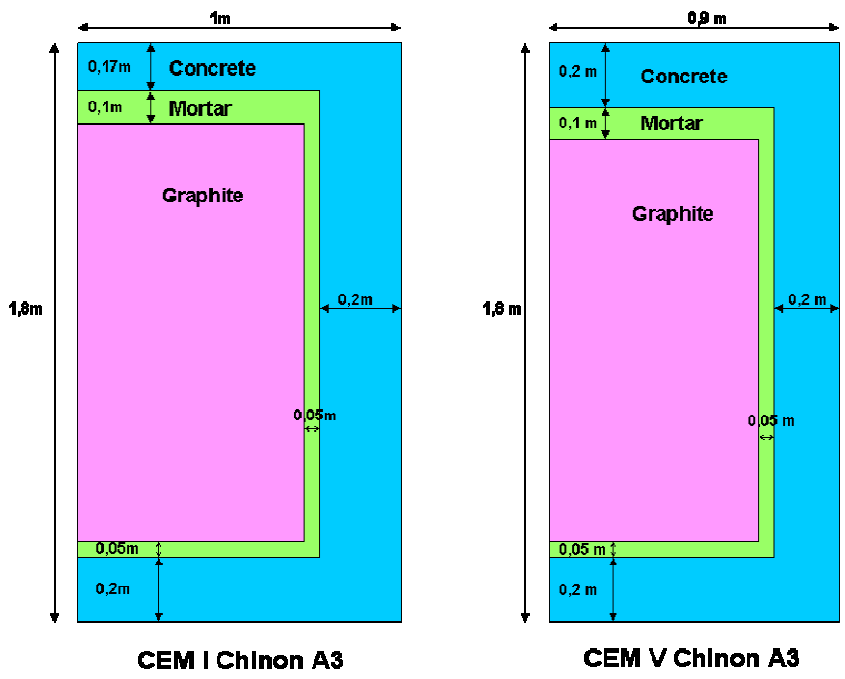

Figure 8: simplified geometrical configuration of waste package

5.2. Nature and composition of cementitious materials

Nature of cementitious materials CEM I and CEM V are in table 5.

	Concrete	Mortar
CEM I waste package	F44 concrete with black steel reinforcements	Mortar with no reinforcements
CEM V waste package	Concrete with stainless steel fibres	Mortar C2-1 with no fibres

Table 5: general characteristics of concrete and mortar

5.3. Graphite characteristics

The physical parameters (porosity, density, diffusion coefficient) of graphite material result from measurements on samples of stacks from Bugey [17]. These parameters are gathered in table 6.

Density ρ (kg/m^3)	1 700
Porosity ω (-)	0.2
Effective diffusion coefficient $\mathbf{D^{eff}}$ (m^2/s)	10^{-11}
^{36}Cl partition coefficient $\mathbf{Kd_{Cl}}$ (m^3/kg)	0
^{36}Cl half-life λ_{Cl} (s)	$9.52.10^{12}$

Table 6: transport parameters for graphite material

5.4. CEM I concrete characteristics

It will be noted that:

- for the concrete, the ratios calcium /silica (C/S) were corrected so as to take into account the presence of silica fume. A hydration rate of 13% of silica fume is considered to obtain a

porosity of healthy material of 9% as indicated in the document [7]. For this material, measurements of diffusion coefficients realized with tritium water on laboratory scale samples led to a very low value, equal to $5.8.10^{-14}$ m² /s. In order to take into account the various phenomena met during the manufacture of an industrial object (for example fractures due to shrinkage), this value was voluntarily lowered by an order of magnitude to be brought back to 5.10^{-13} m² /s,

- for the mortar, in the absence of additional detail on the aggregates, a volumic fraction of the (V_{Gra}) aggregates of 40% was taken into account so as to obtain a porosity of healthy material equal to 15%. It is noted that for this material, the specified value of the effective diffusion coefficient D^{eff} (5.10-12 m2/s) [7] is an estimate (i.e not measured value) based on data of the literature obtained with tritium diffusion experiments. The computed value (1.10^{-12} m^2/s) with the exponential law for a porosity of 15% seems closer to the actual values for this kind of mortar [11],
- density of materials was calculated by considering that all the water of the mixture is consumed by the hydration of the components,
- the presence of the additives as those of the reinforcements is not taken into account in the properties of materials.

The parameters used for calculations are gathered in table 7, together with the parameters describing the decalcification of materials. The other parameters of interest are given in table 8.

	F44 concrete		Mortar	
C_{Ca} (mol/m^3)	(C/S)	porosity	(C/S)	porosity
22	2.632	0.090	3	0.150
21	1.448	0.138	1.65	0.281
2	0.877	0.174	1	0.353
1.5	0.439	0.174	0.5	0.353
0	0	0.174	0	0.353

Table 7: parameters used to describe the CEM I cementitious materials decalcification

		Concrete	Mortar
ρ_s	Density (kg/m^3)	2 471	2 310
ω (calculated)	Calculated porosity (-)	0.09	0.15
ω (spécified)	Specified porosity (-)	0.09	0.15
D^{eff} (calculated)	Calculated effective diffusion coefficient (m^2/s)	$5.6.10^{-13}$	1.10^{-12}
D^{eff} (spécified)	Specified effective diffusion coefficient (m^2/s^1)	5.10^{-13}	5.10^{-12}
Kd_{Cl}	Partition coefficient for chlore 36 (m^3/kg^1)	0.001	0.001
(C/S)°	Initial ratio C/S for cement paste (-)	2.632	3
$(C/S)°_{C\text{-}S\text{-}H}$	Initial ratio C/S for C-S-H (-)	1.448	1.65
S_{Si}	Si concentration in cement paste (mol/m^3)	4 987	4 900
$V^m_{C\text{-}S\text{-}H}$	Molar volume for C-S-H (m^3/mol)	84.10^{-6}	84.10^{-6}
V^m_{CH}	Molar volume for portlandite (m^3/mol)	$33.1.10^{-6}$	$33.1.10^{-6}$
V^m_{Ett}	Molar volume for ettringite (m^3/mol)	$7.15.10^{-4}$	$7.15.10^{-4}$
$V°_{Ett}$	Initial volumic fraction for ettringite in cement paste (-)	0.12	0.12
$V°_{CH}$	Initial volumic fraction for portlandite in cement paste (-)	0.162	0.219
$V°_{CHS}$	Initial volumic fraction for C-S-H in cement paste (-)	0.419	0.412
$V°_{Gra}$	Initial volumic fraction for aggregates (-)	0.7	0.4
D_0	1st coefficient for porosity law (m^2/s)	$2.3.10^{-13}$	$2.3.10^{-13}$
ω_r	2nd coefficient for porosity law (-)	9.95	9.95

Table 8: cementitious materials parameters used in the calculations

5.5. CEM V concrete characteristics

The properties of cementitious materials based on CEM V depend on the composition of the cements used. This later can vary as a function of the proportions and the nature of the components of the cement (clinker, slag, ashes, gypsum). It is thus necessary to have the detailed composition of the paste in order to determine the distribution of the mineral phases once the material is hydrated. In this study, the methodology applied is identical to the one used in the document [12]. This methodology is based on the following assumptions:

- calculations are based on the composition of cement CEM V/A 32.5 N CE from the Airvault factory of Calcia group. Anhydrous cement is considered as composed of 55% with clinker, 22% with slag, 23% with ashes, to which are added 5% of gypsum,
- one considers the following hydration rates:100% for the clinker, 75% for the slag, 65% for ashes and100% for the gypsum,
- for the concrete, the presence of silica fume is neglected (small initial quantity). For the mortar (the formulation selected is C2.1 from the document [8]), a hydration rate of 10% is considered for silica fume, so as to optimize calculated porosity to the value of to 13.7%, compared to that specified in the document [7] which is equal to15.9%,
- the hydrated cement paste is considered as constituted of portlandite (CH), ettringite (Ett), jennite (CSH with initial ratio $(C/S)^{\circ}_{C\text{-}S\text{-}H} = 1.5$) and hydrotalcite (magnesium aluminate, considered as inert during decalcification). Contrary to CEM I materials, the calcium of the ettringite is taken into account in the calculation of the (C/S) ratios for the cement pastes,
- the volumic fraction for each mineral species is deduced directly from the chemical composition of the hydrated cement paste,
- for the concrete, the volumic fraction of aggregates is fixed at 0.7 (optimized, in absence of data on the aggregates); for the mortar this fraction (equal to 0.487) is calculated from the specifications concerning the sands used [8],
- the densities were calculated similarly to that obtained for the packages based on CEM I,
- the presence of additives and:or fibres is not taken into account in the material properties .

One notes that the modelling of cementitious materials based on CEM V cement is appreciably different from those based on CEM I cement [11, 18], because of a different mineralogical assembly (the mineralogical composition obtained from the above assumptions is deferred in table 12) for these two types of hydraulic binders. In a general way it is considered that the materials subjected to decalcification are composed of three different zones:

- a healthy zone for which C_{Ca} ranges from 22 to 21 mol/m^3, corresponding to the dissolution zone of portlandite,
- a degraded zone for which C_{Ca} ranges from 21 to 2 mol/m^3, corresponding to the evolution zone of ettringite, and in which the (C/S) ratio of the CSH lies between initial value $(C/S)^{\circ}_{C\text{-}S\text{-}H}$ and 0.8,
- a surfacic zone for which C_{Ca} is lower than 2 mol/m^3, in which some CSH remain with a (C/S) ratio lower than 0.8.

The parameters used to describe the decalcification of materials are gathered in table 9.

	Concrete		Mortar	
C_{Ca} (mol/m^3)	(C/S)	porosity	(C/S)	porosity
22	1.97	0.101	1.90	0.137
21	1.66	0.117	1.66	0.161
2	0.8	0.147	0.80	0.217
0	0	0.147	0	0.217

Table 9: parameters used to describe the CEM V cementitious materials decalcification

As indicated previously, for materials based on CEM V, the diffusion coefficient D^{eff} is specified in the form of constant values for each zone described above. The diffusion coefficients were selected in agreement with the values used in the document [18]. With regard to the mortar, the value of the diffusion coefficient for the healthy zone corresponds to that

measured on material of formulation C2.1 [8]. It should be noted that this value is obtained by diffusion of chloride ions under electric field. The specified values are given in table 10.

	Concrete	Mortar
D^{eff} ealthy zone (m^2/s)	$1.5.10^{-13}$	$1.2.10^{-13}$
D^{eff} degraded zone (m^2/s)	$3.0.10^{-13}$	$3.0.10^{-13}$
D^{eff} surfacic zone (m^2/s)	$3.0.10^{-12}$	$3.0.10^{-12}$

Table 10: specified values for diffusion coefficients for each decalcification zones

However, this kind of specification (constant values by zones) is difficult to implement during calculations (it generates numerical oscillations when the different zones are changing). Consequently, the definition of the diffusion coefficients is replaced by an exponential law. This law (Equation 4) is expressed in the form:

$$D^{eff} = D_1 * \exp[-(C_{Ca}/b_1)^{a1}] + D_2 \quad (4)$$

The values of the coefficients of this law are deferred in table 11.

	a1 (-)	b1 (mol/m^3)	D_1 (m^2/s)	D_2 (m^2/s)
Concrete	0.7	2	$2.85.10^{-12}$	$1.50.10^{-13}$
Mortar	0.7	2	$2.88.10^{-12}$	$1.20.10^{-13}$

Table 11: values of coefficients of the exponential law

The other parameters of interest for the packages based on CEM V concrete are given in table 12.

		Concrete	Mortar
ρ_s	Density (kg/m^3)	2 420	2 152
ω (calculated)	Calculated porosity (-)	0.101	0.137
ω (spécified)	Specified porosity (-)	0.08	0.159
D^{eff} (calculated)	Calculated effective diffusion coefficient (m^2/s)	$1.6.10^{-13}$	$1.3.10^{-13}$
D^{eff} (spécified)	Specified effective diffusion coefficient (m^2/s^1)	$1.5.10^{-13}$	$1.2.10^{-13}$
Kd_{Cl}	Partition coefficient for chlore 36 (m^3/kg^1)	0.001	0.001
(C/S)°	Initial ratio C/S for cement paste (-)	1.97	1.9
$(C/S)°_{C\text{-}S\text{-}H}$	Initial ratio C/S for C-S-H (-)	1.66	1.66
S_{Si}	Si concentration in cement paste (mol/m^3)	5 132	5 845
$V^m_{C\text{-}S\text{-}H}$	Molar volume for C-S-H (m^3/mol)	$4.58.10^{-4}$	$4.58.10^{-4}$
V^m_{CH}	Molar volume for portlandite (m^3/mol)	$33.1.10^{-6}$	$33.1.10^{-6}$
V^m_{Ett}	Molar volume for ettringite (m^3/mol)	$7.15.10^{-4}$	$7.15.10^{-4}$
$V°_{Ett}$	Initial volumic fraction for ettringite in cement paste (-)	0.052	0.047
$V°_{CH}$	Initial volumic fraction for portlandite in cement paste (-)	0.10	0.11
$V°_{CHS}$	Initial volumic fraction for C-S-H in cement paste (-)	0.39	0.45
$V°_{Gra}$	Initial volumic fraction for aggregates (-)	0.7	0.487

Table 12: cementitious materials parameters used in the calculations

5.6. Modelling approaches

The basic assumptions of the model used are the following ones:

- transport of radionuclides is done only by diffusive way, by considering that the waste

package is water saturated (the water intake by the packages is regarded as very fast as soon as water is available; for long term operational modelling, it will thus be regarded as instantaneous), and the flow conditions within the environment of storage are not taken into account,

- it was agreed focus only on the case of the ^{36}Cl. This radionuclide is considered as uniformly distributed within the matrix graphite,
- the cart is considered as filled only with graphite waste. Spaces between the various graphite elements as well as the geometry of these elements are not taken into account,
- the waste package is considered as composed of three materials: the cart with graphite waste, the mortar and the body of the concrete container.

5.7. Initial and boundary conditions

The following initial and boundary conditions were applied:

- the initial calcium concentration in solution is considered equal to 22 mol/m^3 in all the system,
- the initial ^{36}Cl concentration in the interstitial solutions of concrete and mortar is considered to be zero,
- the initial ^{36}Cl concentration (in Bq/m^3) in graphite waste is calculated by considering an inventory of 3300 Bq/kg for stacks from Chinon A3 [19]. This inventory is supposed entirely soluble and present in the porosity of graphite at the initial moment of calculation,
- by symmetry, one considers a null flux on the medium vertical axis of the package,
- so as to ensure a conservative approach, a zero and constant ^{36}Cl concentration is considered on the surface of the package during the whole computations (i.e one considers that the environment medium has the capacity to instantaneously evacuate the ^{36}Cl),
- in the same way, a zero and constant calcium concentration is considered at the surface of the package.

5.8. Implementation of calculations

With the simplified geometrical representations adopted for the package, calculations were carried out by considering structured grids on right-angled fields. In a general way, meshes of 5 mm by 5 mm dimension were used.

Calculations are carried out with a value of diffusion coefficient of water in water equal to $2.2.10^{-9}$ m²/s.

5.9. Output parameters

To analyse the results, different indicators were selected in order to highlight the variations obtained as a function of the various calculation cases. In a general way, the results were exploited using the following parameters:

- containment time (expressed in years), defined as the period of time when the cumulated activity released by the package does no longer change over time (variation lower than 0,001% in 100 years),
- maximum relative activity flux (expressed in $year^{-1}$), expressing the maximum value reached by the activity flux (in Bq/year) released by the package, relative to the initial total activity contained in the package (in Bq),
- characteristic time at which the maximum reported activity flx is reached.

6 Results

We will present first the results of calculations for packages based on CEM I and on CEM V concretes, whose materials are either healthy, or decalcified or decalcified and early fractured.

In the second time, sensitivity studies are carried out on the value of the porosity of irradiated graphite, on the value of the diffusion coefficient of chlorine 36 in graphite and on the ^{36}Cl source term. The data used in these studies result from the work undertaken within the framework of the Work Package number 3 of the Carbowaste Project.

6.1. Results for packages based on CEM I

The comparative curves of cumulated activity released by the package and of relative activity flux for the various configurations of package based on CEM I for stacks from Chinon A3 are presented in figures 9 and 10.

The values of the indicators are gathered in table 13.

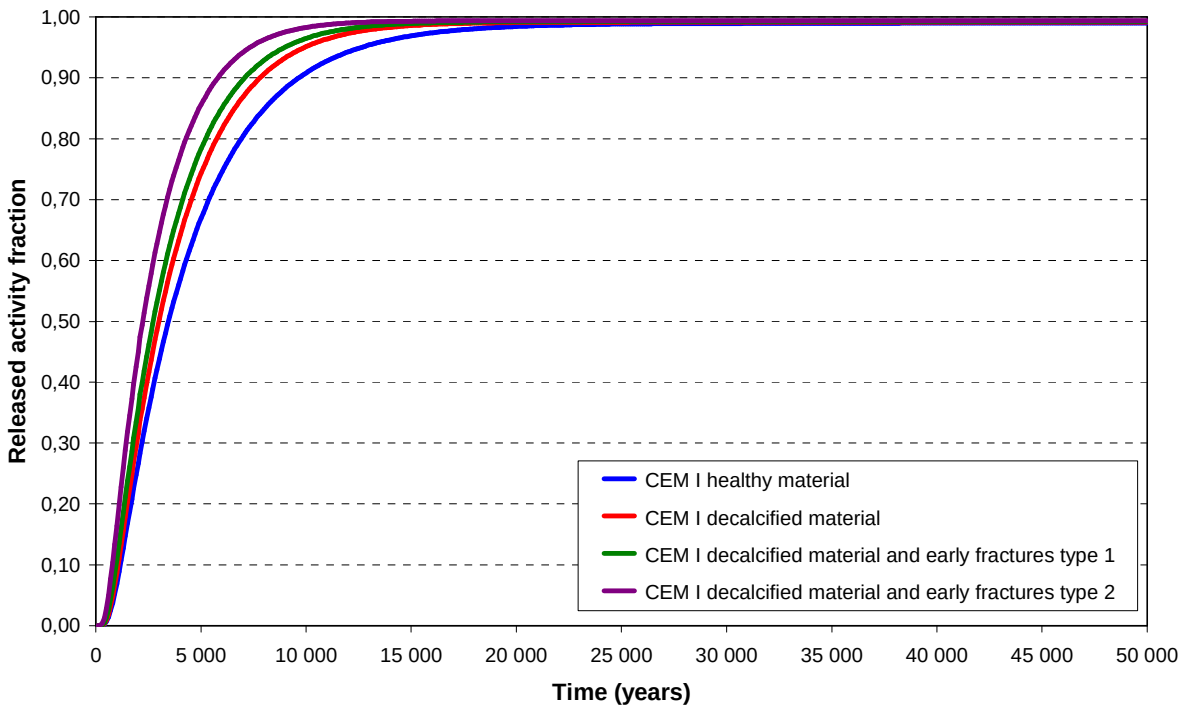

Figure 9: evolution released activity versus time for CEM I

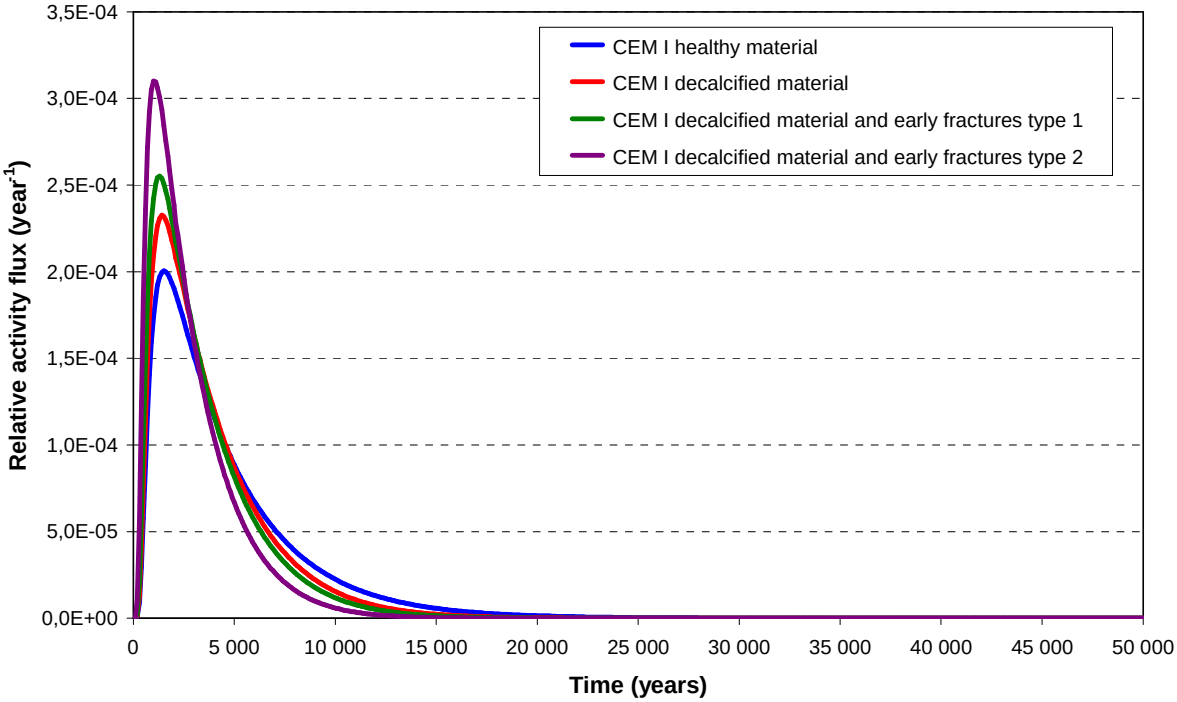

Figure 10: relative activity flux versus time for CEM I

	Containment time (year)	Maximum relative activity flux ($year^{-1}$)	Time of maximum relative activity flux (year)
Healthy concrete	32 000	$1.93.10^{-4}$	1 600
Concrete with degradation	26 600	$2.24.10^{-4}$	1 500
Concrete with degradation and type 1 early fractures	22 600	$2.42.10^{-4}$	1 300
Concrete with degradation and type 2 early fractures	18 900	$3.10.10^{-4}$	1 100

Table 13: containment performances for CEM I concrete

6.2. Results for packages based on CEM V

The comparative curves of cumulated activity released by the package and of relative activity flux for the various configurations of package based on CEM I for stacks from Chinon A3 are presented in figures 11 and 12.

The values of the indicators are gathered in table 14.

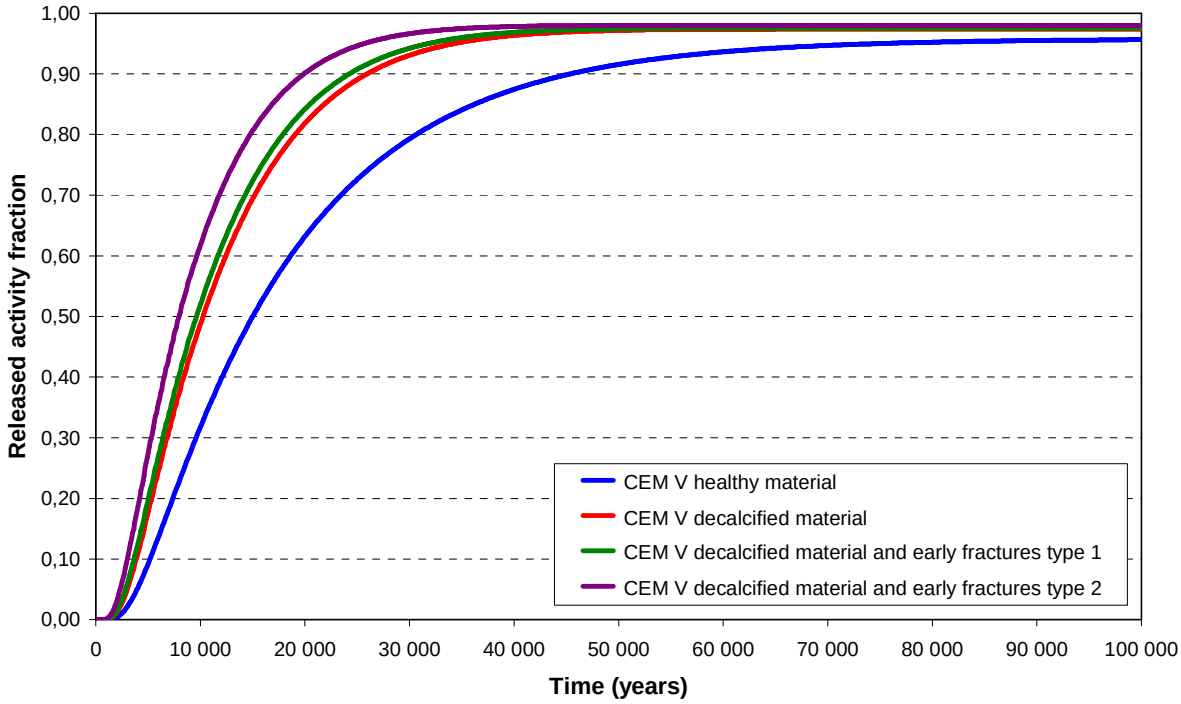

Figure 11: evolution released activity versus time for CEM V

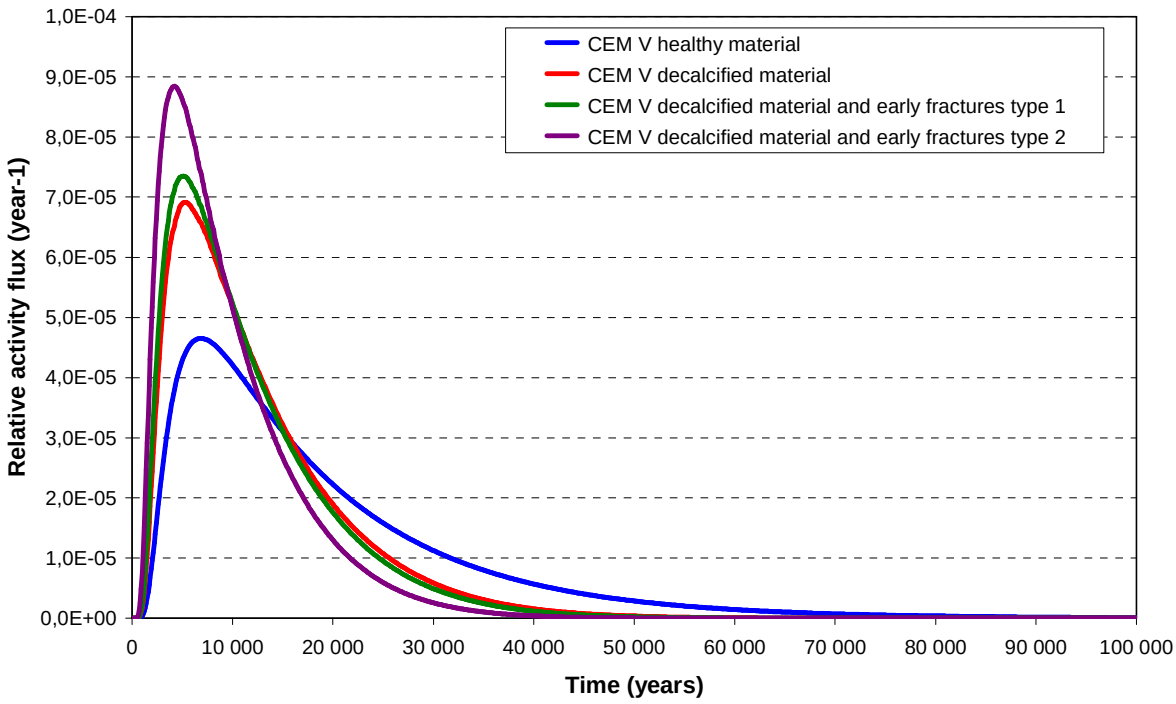

Figure 12: relative activity flux versus time for CEM V

	Containment time (year)	Maximum relative activity flux ($year^{-1}$)	Time of maximum relative activity flux (year)
Healthy concrete	102 700	$4.65.10^{-5}$	6 900
Concrete with degradation	60 100	$6.91.10^{-5}$	5 300
Concrete with degradation and type 1 early fractures	57 000	$7.35.10^{-5}$	5 100
Concrete with degradation and type 2 early fractures	49 200	$8.84.10^{-5}$	4 200

Table 14: containment performances for CEM V concrete

6.3. Sensitivity studies

The value of graphite porosity, equal to 20%, is resulting from measurements on non irradiated graphite. In order to refine the operational modelling of the waste packages, it is better to use a value of porosity determined on irradiated graphite. A study carried out by the CEA [20] highlights values of porosity, for samples of graphite coming from the G2 reactor, varying from 23% to 26%, so that calculations are carried out with these two values. For such variations of porosity, which remain weak, the results are almost unchanged compared to the calculation carried out previously.

In the same way, the value used for the diffusion coefficient of chlorine 36 in graphite, equal to 5.10^{-11} m²/s, is updating according to measurements carried out on samples coming from G2 reactor [20] which give a range of values between $0.9.10^{-11}$ m²/s and 4.10^{-11} m²/s. Such variations do not lead to a significant effect on the release of chlorine 36 from the waste packages.

The very little impact due to the adjustment of the values of porosity and diffusion coefficient of chlorine 36 in graphite on the release of this radionuclide from the package is certainly due to the fact that the retention is mainly due to cementitious materials (concrete and mortar) and not to graphite itself.

Lastly, a sensitivity study was also undertaken on the source term (chlorine 36). Measurements on samples from G2 [20] show strong heterogeneities. Values going from 15 to 1300 Bq/g (the value of reference being of 3.3 Bq/g) were taken into account. The relative activity fluxes are not modified with the source term. On the other hand the durations of containment slightly decrease when the source term increases. This is certainly due to an increase in the chlorine 36 concentration gradients involving an acceleration of the diffusion phenomenon.

6.4. Conclusion

In a first time, one notes that the performances of the packages based on CEM V are appreciably better than those of the packages based on CEM I, with containment times

respectively equal to 102 700 years (CEM V) versus 32 000 years (CEM I) when healthy materials are considered. This conclusion remains true for the configurations with decalcified materials, with or without early fractures.

For the packages based on CEM I, the containment time, equal to 32 000 years when considering healthy materials, decreases by 23% if one considers a decalcification leading to an opening of porosity, and thus to an increase in the diffusion coefficient of chlorine 36. The presence of realistic early fractures (type 1) led to a light additional degradation (29% compared to healthy materials), which is still accentuated (40%) when a penalizing early fractures (type 2) is considered.

For the packages based on CEM V, the same tendency is observed, with however a stronger loss of containment time. Indeed, the containment time, equal to 102,700 years when considering healthy cementitious materials, decreases by 41% if one takes into account the decalcification phenomenon. The presence of realistic early fractures (type 1) led to a light additional degradation (44% compared to healthy materials), which is also accentuated (52%) when penalizing early fractures (type 2) is considered.

Lastly, the sensitivity studies carried out with the parameters of irradiated graphite (porosity, diffusion coefficient of chlorine 36 and source term) do not modify the results.

7 General conclusion and perspectives

In this note, the principal available data on the origin of waste graphite in France are pointed out (chapter 2). The waste packages based on CEM I and CEM V concrete are described (chapter 3).

The following chapter presents the evolution of the operational modelling (MOP) describing the release of the radionuclides by the different packages (chapter 4). With the initial modelling which did take into account only healthy materials with an only diffusive transfer for the radionuclides, we then added functions making it possible to describe the decalcification in time of cementitious materials and early fractured materials.

In the following part (chapter 5), all the parameters selected and the assumptions made for each calculation configuration are listed.

In chapter 6 are presented the results of performance calculations for each configuration. It is noted that performances of the packages based on CEM V are appreciably higher than those of the packages based on CEM I, for materials healthy in time as for materials evolving in time, with or without early fractures. In addition, it is noted that the performances of the packages are decreased when one takes into account the decalcification and the early fractures.
Taking into account the parameters (porosity, diffusion coefficient, source term) resulting from measurements on irradiated materials coming from reactors do not modify the results.

Let us note however that these performances are calculated without the presence of the engineering barriers (cells or galleries), which should then be appreciably improved.

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