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Journal of Nuclear Materials 358 (2006) 82-95

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Some principles of service life calculation of reinforcements and in situ corrosion monitoring by sensors in the radioactive waste containers of El Cabril disposal (Spain)

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Received 21 February 2006; accepted 19 June 2006

Abstract

Reinforced concrete is the most usual material used in engineered barriers in low-level nuclear waste disposal facilities. The record of modern concrete is no longer than about 100 years. During this time, it has been noticed that the material gives a good performance in many environments, however several chemical aggressive species in water, soil or the atmosphere may react with the cement mineralogical phases and perturb its integrity. El Cabril repository has a design life objective of longer than 300 years and therefore, these structures should maintain their main characteristics during this target service life. The potential aggressive conditions that the cement-based materials can suffer have been identified to be: carbonation, water permeation (leaching) and reinforcement corrosion. More unlikely may be the biological attack. Chlorides are not in the environment but they are inside the drums as part of analytical wastes. Vaults and containers are made of a very similar concrete composition while the mortar is specifically designed to be pumpable, with low hydration heat, low shrinkage and of low permeability. In this paper results of concrete characteristics are given as well as the monitoring of the behaviour of reinforcement corrosion parameters from 1995 on the same environmental conditions of the actual waste. This monitoring has been made in a buried structure with embedded sensors. The effect of temperature is commented.

1. Introduction

Cement-based materials (CBM) may be very durable. Examples, of 2000–3000 years old, have survived until present days showing still good performance and therefore, they are suitable candidates to play an important role in the long term immobilization of radioactive wastes. However, the record of modern concrete is no longer than about 100 years. During this time, it has been noticed that the material presents a good performance in many environments, however several chemical aggressive species in the water, soil or the atmosphere may react with the cement mineralogical phases and disturb its integrity. The challenge of the concrete as engineering barrier is its long term stability.

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In Spain, Enresa has selected the place of El Cabril (Córdoba) for the placement of a repository of low and medium radioactive wastes and cementitious materials have been identified for being an important part of the engineering barrier.

In other countries, similar experiences have been developed for the low and medium radioactive waste disposals. Concrete is normally the most suitable material used for the vault construction.

Fig. 1 shows a general view of El Cabril installations devoted to waste disposal. The different main structures in which CBM are used, as part of the engineered barrier, are:

- 1. The so named 'vaults' (large structures).
- 2. The containers (cubic form and smaller).
- 3. The mortar filling the containers around the drums.
- 4. The drum matrices that contain the waste.

The main cement-based materials used in engineered barriers at El Cabril repository are the vaults, the containers and the mortar filling the gaps between the drums introduced in the containers. Vaults and containers are made of very similar concrete compositions, while the mortar was specifically designed to be pumpable and with high impermeability.

The vaults and the containers were made of the same concrete, whose composition is given in Table 1. The composition of the mortar is also given

Table 1Composition of concrete and mortar

	Concrete	Mortar
Cement I-42.5 R/SR	400	_
Cement IV-B-32.5 SR/BC	_	708.3
Sand (0–2.5 mm)	297	_
Sand (0–5 mm)	614	_
Sand 'ASIROSA'	_	1239.4
Aggregate (6–16 mm)	949	_
Superplastifier Melcret 222	4.8 (1)	_
Admixture Rheobuild 1000	_	7 (1)
w/c Ratio	0.45	0.37
w/(c+blending agents) Ratio	0.45	_

there. The IETcc was responsible of their definition. In both cases the cement selected was a low C3A and alkali content in order to prevent sulphate or alkali-aggregate attack. Its chemical analysis is given in Table 2.

The mortar after being designed is prepared as pre-packed material by two manufacturers and transported in dry conditions. Its mixing with water is made in El Cabril at the location where the drums are introduced into the containers. There, the containers are filled with the mortar and covered with a concrete cap to be then transported and placed into the vaults.

The containers are prefabricated in a special devoted plant. The reinforcement bars are placed in the moulds and then, concrete is casted, to be finally cured in a number of steps, including steam



Fig. 1. General view of structures with CBM in El Cabril.

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Table 2		
Chemical	analysis	of cement

Compound %	SiO ₂	CaO	Al_2O_3	Fe ₂ O ₃	SO ₃	MgO	Na ₂ O	K ₂ O
Cement	20.20	65.84	2.17	4.1	3.8	1.85	0.11	0.65
Fly ash	48.71	12.09	25.18	5.18	0.43	1.61	0.61	3.28

cured temperatures lower than $60 \,^{\circ}$ C and wet curing. Real characteristic strengths of around 60 MPa are typically achieved at 28 days.

The vaults were all built at the same time and made with the same type of concrete that is used for the containers. When filling of the containers is completed, a closing reinforced concrete slab is constructed to seal the particular vault. After completion of the operation of the facility, the vaults will be covered with an engineered earthen cap approximately 3 m thick.

In addition to the day to day quality control established in the Quality Assurance Programme, microstructural periodic checking is made by IETcc. This control was very frequent (once per month) during construction of vaults and developing of the prefabrication plant procedures. After verification of the constancy in the results of such checking, the frequencies were extended and nowadays a yearly based control is being maintained.

The main parameters measured and their typical values are given in Table 3.

The aim of present paper is summarize some of the requirements defined for the cement-based materials in order to achieve a target service life of 300 years and to illustrate the basis of models currently being developed to improve the simplified assessment of the disposal system durability. The monitoring of corrosion parameters in a container which has been buried to reproduce real conditions foreseen in the long term is described as well.

Table 3

Parameters	related	to	concrete	durability	and	typical	values
obtained on	El Cab	ril c	concrete				

Parameters	Typical mean values
Compression strength (MPa)	61.13
Porosity (% in volume)	7.25
Density (g/cm ³)	2.31
Coefficient of water absorption $(kg/m^2 s^{0.5})$	1.83E-3
Coefficient of resistance to water penetration (s/m^2)	6.32E+8
Oxygen permeability (m ²)	11.18E-18

2. Some principles of service life prediction of CBM in El Cabril

As mentioned above, the target service life was defined as 300 years. Taking into account that the record of modern reinforced concrete is less than 100 years, reaching the target life is a challenge. Much older concretes have survived to present, but they are not reinforced with steel and are based on mixes of lime and pozzolan which result in analogue not readily usable to assess modern cements. In addition, the experience about structures made in the 20th century is difficult to extrapolate to present cements beyond around 30 years, because cement production technology has changed dramatically during the century. Present cements have a much higher C_3S content, with high CS_3/CS_2 ratio and much higher finesse.

All these uncertainties make the prediction of performance for 300–500 years to be impossible to be fully validated. Therefore, as a general characteristic, robust and conservative models for degradation have been developed.

Same conservatisms were applied to the election of material: a pre-requisite for the CBM selection was that only materials with experimental record of about 25 years were preferred. No new materials with records of performance less than 25 years were considered as candidates for making part of the CBM. So, materials such as silica fume or fibres of glassy metals were rejected in the initial mixes studied. Due to the long term performance required no new material is going to be recommended for El Cabril. Therefore, the main principle was to fabricate a good performance concrete, but with no blending agents.

Another general statement concerns organic compounds. Their acceptance was limited to those with enough experimental performance history and their presence was minimized due to the potential bio-attack when the structures are buried.

After stating these general pre-requisites, two main aspects need to be considered for the life model: the type and level of attacks (environmental conditions or actions) and the models to calculate the resistance to them of the CBM. As preliminary framework, the life model to be considered will be of the type:

$$t_i = t_0 + t_1 + t_2 + t_3 + t_4 + \dots + t_n$$

With t_i representing time taken for the aggressive substances (mainly water from the rain or from the water table) to penetrate the different engineering barriers (ground cover, concrete, mortar, etc.). This model is conceptual and indicates the need to describe the two main scenarios of the rain water penetrating from the top of the vaults or the water raising by capillarity from the water table and reaching also the bottom of the vaults. Depending on the composition of these waters when in contact with the concrete of the vaults the possible risks of deterioration will depend on the types of attack.

3. Environmental conditions at El Cabril

Following these conceptual scenarios, the possible aggressions that CBM can suffer, identified as significant, were: (a) sulphates, (b) alkali-aggregate, and (c) corrosion of reinforcements due to carbonation or chlorides (which are not in the environment but they are inside the drums as part of analytical wastes). Leaching was not initially considered of importance in a first step due to the buried conditions above water table, although it was studied afterwards. More unlikely might be the bio-attack.

Two main different environmental situations can be considered at El Cabril facility: The operational period where there are atmospheric and the buried parts in the engineered barriers, and the postoperational phase configuration, where the whole concrete vaults will be protected by an earthen engineered cap, built to minimize water percolation and isolating the concrete barriers from temperature changes.

That is, the total life time t_t is composed of several steps, t_i , which will depend on the particular environmental action considered and the number of barrier layers. In the case of only considering corrosion due to carbonation, the t_t will be the time for the carbonation front to reach the reinforcements by passing through the external coating and the cover (in the first 30 years). Only two t_i would be under consideration in this preliminary assumption, of considering steel depassivation the limit state.

For the case of chlorides incorporated in the waste drum matrices, the t_t would be the time taken by the chloride to diffuse through an assumed limited drum wall failure, then through the surround-

ing mortar and through he cover of the container to reach the reinforcement.

3.1. Buried structures

The most important characteristic of the ground, regarding CBM durability, is the water table geochemistry and depth. This was identified as being a few meters below the foundations of the vaults and the water composition contained very low sulphate and chloride contents. Therefore, from the perspective of the ground, it was not considered as possible attack mechanism to concrete, except possibility for bio-attack. In this first step, was foreseen so low that was neglected.

Anyway, technical specifications were set up to prevent sulphate and alkali-aggregate attack (i.e. by the use of a cement of low C_3A and alkali contents and carefully controlled selection of aggregates). Therefore, only corrosion of reinforcement has been studied in detail.

Regarding corrosion of reinforcements in buried parts, a difficulty is the scarce information found in the literature about observations of reinforcement corrosion in foundations. Corrosion rates have been measured very seldom. No corrosion due to carbonation is expected because carbonation does not develop in buried structures and, in general, the observation is that corrosion has developed in the foundations only when chlorides or other aggressive ions exist in the groundwater. The concrete then cracks and looses its ability to protect the steel. Existing chlorides, sulphates or other deleterious substances towards CBM were not found in the groundwater of El Cabril, so significant risk of corrosion in the buried parts was not identified.

Nevertheless, as part of the research program developed, a particular test was made through the corrosion monitoring of a pilot buried container. This will be described later.

3.2. Structures exposed to the atmosphere

The atmosphere in El Cabril is controlled by the climate. The climate is of continental and rural type with the mean values shown in Table 4 of the parameters relevant for CBM durability.

With the absence of chlorides, the only aggressive substances identified in this atmosphere are the water and the carbon dioxide. Both are very ordinary and of no particular risk.

Table 4 Mean values of HR, temperature and rain in El Cabril

Climate parameters	Relative humidity (%)	Temperature	Rain (mm/year)
Mean values	61.03	17.12	546
Standard deviation	15.81	6.72	

In summary, regarding reinforcement corrosion the only expected aggression considered was corrosion induced by carbonation in the atmospheric zones. In the interior of the vaults some diffusion of carbon dioxide may occur from the atmosphere through the drain collectors.

Regarding carbonation in the atmospheric parts, the time action assumed was not a target of 300–500 years, but of some 30 years, because this will be the length of the period where unburied conditions are expected. That is, the vaults will be exposed to the atmosphere during a maximum of 30 years before covered with ground.

3.3. Presence of chlorides and other aggressive ions in the matrices with waste

The amount of chlorides inside the drums is very variable. The chloride attack would start by perforating the metal of the drums and diffusing through the backfilling mortar, reaching the reinforcement of the containers. For this attack to develop, the presence is needed of enough water in the concrete pores and the perforation of the drum walls, at least, locally.

4. Tests to characterize parameters for the life model

For the preliminary life model assumed, the main tests that have been carried out to study the risk of reinforcement corrosion can be grouped into four main areas:

- 1. Measurement of carbonation velocity in indoor and outdoor conditions.
- 2. Monitoring of concrete air permeability in natural conditions.
- 3. Monitoring of corrosion parameters in buried and atmospheric conditions.
- 4. Characterization of resistance to chloride transport of concrete and mortar and modelling chloride diffusion from the drums.

Other complementary tests are foreseen in support of the general life time model improvements. These were for the sake of characterization of CBM, during construction of the vaults.

4.1. Measurement of concrete carbonation velocity

Carbonation occurs in concrete because the material has an alkaline nature and the calcium bearing phases present are attacked by carbon dioxide of the air and converted to calcium carbonate [1]. If CO_2 from air or from water penetrates, the concrete will carbonate according to

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O_3$$

The carbonation process requires the presence of a certain amount of water. If the concrete is too dry (RH < 40%) CO₂ cannot dissolve and no carbonation occurs. If on the other hand it is too wet (RH > 90%) CO₂ cannot enter the concrete and the concrete will not carbonate. Optimal conditions for carbonation occur at a RH of 50% (range 40–90%).

The advance of carbonated front through the concrete cover can be determined by using a pH indicator (Phenolphtalein). In this way, it is possible to determine the carbonation velocity (V_{CO_2}) of a concrete in different environments by making different measurements with time. The expression used in



Fig. 2. Specimen treated with the phenophtalein indicator.



Fig. 3. Evolution with time of the carbonation rate calculated on the control specimens in El Cabril.

 Table 5

 Quality concrete cover classification [3]

Quanty concrete cover enconnection [5]			
Quality of concrete cover	Class	$KT (10^{-16} \text{ m}^2)$	
Very poor	5	>10	
Poor	4	1–10	
Normal	3	0.1 - 1	
Good	2	0.01 - 0.1	
Very good	1	< 0.01	

the present study for the determination of the (V_{CO_2}) is the well known

$$X = V_{\rm CO_2} \cdot t^{0.5},\tag{1}$$

where X is the depth of the carbonated front at each time (t).

Because of the destructive character of the technique used for carbonation measurements, the study of carbonation penetration of the container concrete has been made by the exposure of cylindrical control specimens corresponding to two different vaults and stored in three different environments: El Cabril atmosphere (outdoors), the interior of a building (indoors), and buried samples.

Each year, two specimens on each exposure are broken and carbonation is observed by spraying phenolphtalein on both sides after splitting the specimen along a transversal axis (Fig. 2). These sides are two rectangular surfaces whose dimensions are 15×30 cm, that are impregnated with the colour indicator. In the example of the figure it is observed how the carbonation front is around 6 mm depth after 13 years of exposure to the environment.

The carbonation velocity is calculated using expression (1). As can be observed in Fig. 3, the rate of carbonation presents some scatter but it is quite stable, not showing an influence of the exposure outdoors or indoors of a building. The low values found, with an average of (V_{CO_2}) around 1.5 mm/ year^{0.5}, confirm the good carbonation resistance of the concrete designed.

Finally, concerning the buried specimens they show no carbonation sign until present.

4.2. Air permeability measurements in concrete

Air permeability is considered a rough estimation of concrete compactness. In the present study it is



Fig. 4. Results of permeability coefficient (KT) measured during several years over the internal walls of the real concrete container.

measured in the surface covering layers by a non destructive method [2]. The system is based on creating a vacuum inside a device placed on the concrete surface, and measuring the air flow after a certain time.

The ranking of cover quality is empirical, depending on the permeability coefficient value. Experimental tests using this method permit a classification of the concrete cover into five different classes (Table 5). These measurements have been made two times per year directly over the concrete surface of the vaults. In the same points corrosion rate measurements are also made. The trend shows that the coefficient of the values *KT* changes between 10^{-2} and $10^{-1} \times 10^{-6}$ m² (Fig. 4). These values provide a concrete class between 1 and 3, following the Torrent's classification, which is shown in Fig. 5. That means a normal, good or very good quality of the concrete cover.



Fig. 5. Relationship between resistivity and *KT*. Nomogram for the connection by resistivity of the quality cover classification. No connection was necessary due the high values of resistivity.



Fig. 6. Results of resistivity measured during several years over the internal walls of the real container.



Fig. 7. Results of corrosion potential measured during several years over the internal walls of the real container.



Fig. 8. Results of corrosion potential measured during several years over the internal walls of the real container.



Fig. 9. Reinforcement of the pilot container with corrosion sensors and its aspect once buried. There is a main hole to have access to the chamber where the corrosion-data logger is placed besides the container.

Since the permeability measurements depend on the concrete moisture, resistivity values are measured simultaneously. There is no criteria for the relation between resistivity and permeability, but as mentioned in [2], that when the concrete is not completely dry, it is necessary to calibrate the classification with the resistivity values (measured in the same points than permeability), as is presented in Fig. 5.

4.3. Monitoring of corrosion parameters in atmospheric and buried conditions

4.3.1. Atmospheric conditions

In the same places where permeability, resistivity, corrosion potential and corrosion rate of reinforcement are determined. The electrochemical parameters are measured in the concrete vaults in El Cabril – Cordoba – Spain. These three parameters are measured twice a year with the corrosion rate meter Gecor 6. Fig. 6 shows the results of resistivity measured during several years. As said, resistivity has shown values between 100 and 300 K Ω cm, these values indicate that concrete is dry. More than 90% of the corrosion potential values are more positive than -200 mV (using Cu/CuSO₄ as reference electrode), (Fig. 7) indicating a non-active corrosion.

The corrosion rate values (Fig. 8) are below the depassivation limit $(0.1 \,\mu\text{A/cm}^2)$, so they indicate the passivity of reinforcement, as expected.

4.3.2. Buried conditions

The monitoring of corrosion parameters has been made by the instrumentation of a pilot container (Fig. 9) filled with drums that also have been instrumented (Fig. 10) [4]. This pilot container has been monitored from 1995 by embedding 27 sets of electrodes. The parameters measured are: temperature, concrete deformation, corrosion potential, resistivity, oxygen availability and corrosion rate. The impact of yearly cycle of temperature on several of the parameters is remarkable, and therefore, care has to be taken when interpreting on-site results.

There is a main hole to have access to the chamber where the corrosion-data logger is placed besides the container.

Electrochemical and non electrochemical measurements were made using a Geologger measurement system installed on a gallery beside the



Fig. 10. Instrumented drum.



Fig. 11. Data logger.

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container (Fig. 9 right part). The system has 50 available channels for recording the several groups of sensors (Fig. 11).

For measuring the corrosion potential and the corrosion rate, either the main rebar of the container or the bare metal surface of the drum, were used as working electrodes. In order to obtain the real value of the corrosion rate, the measurement method used was the polarization resistance determined from a galvanostatic pulse. However, as the surface of the working electrode is bigger than the counter (small disk) a technique based on the measurement of the slope of the transient pulse after application of a step of current, has been used. This technique is not as accurate as the one based on 'sensorized guard ring'. However, as the technique is continuously recorded, any scatter or wrong measurement can be easily identified. On the other hand, it has the advantage of being very quick, and it allows a very small disturbance of the system.

Regarding the resistivity, it is measured by means of the current interruption method from a galvanostatic pulse. The oxygen flow at the rebar level is measured by applying a cathodic constant potential of about -750 mV (SCE) and measuring the current of reduction of oxygen after a steady-state is achieved.

From the 27 groups of sensors installed, only less than 10% of them have failed. The rest show a good response even 10 years after their installation. As an example, Figs. 12 and 13 show the results obtained from one group of sensors placed in one wall of the concrete container (group 13), and from another that is placed over one drum (group 26). From these figures is possible to deduce that the temperature evolution due to the seasonal changes is the most influencing factor for the trends recorded. The reinforcement remains passive as expected $(I_{corr} < 0.1 \,\mu A/cm^2)$.

The 10 years recording has enabled the deduction that a progressive decrease of the amount of oxygen is detected, without this being noticed by the values of the corrosion potential. The progression of hydration is well reflected by the increase in electrical resistivity and the strains are very good detectors of the presence of water in liquid state.



Fig. 12. Evolution with time of corrosion parameters in the sensors embedded in the pilot container (reinforcement structure).



Fig. 13. Evolution with time of corrosion parameters in the sensors embedded in the pilot container (drum).

4.4. Characterization of resistance to chloride transport of concrete and mortar and preliminary modelling of chloride diffusion from the drums

4.4.1. Chloride testing

Concerning the chloride transport, an extensive research program has been developed to characterize the diffusion coefficients, D, and the chloride threshold for steel depassivation. Tests for measuring the D in steady and non-steady conditions have been undertaken by using migration (accelerated) experiments [5]. Fig. 14 shows the cell type and the arrangement used. The tests were performed with different types of concrete (mixes 1– 4) and mortar mixes (mixes 5 and 6) in different conditions, in order to collect data for future modelling.

The steady-state diffusion coefficients [6], calculated according to the procedure and equations given in [7], are presented in Fig. 15, where it can be seen that for concrete mixes (1-4) there is not a noticeable decrease in the values obtained as the age of the matrixes increases. As far as the mortars



Fig. 14. Arrangements for the accelerated testing of chloride diffusion by means of migration experiments.

are concerned, a more pronounced decrease has been produced.

The non-steady-state diffusion coefficients are given in Fig. 16. From Fig. 16, it can be deduced that for the data corresponding to concrete dosage1, cast with ordinary Portland Cement (OPC) low in aluminates, there is not a decrease in the diffusion coefficient as long as the test or age of the specimens increases. In addition, they can be correlated in function of the duration of the test all together, independently of having been exposed to the



Fig. 15. Steady-state chloride diffusion coefficients obtained for the six mixes after curing in humid chamber for 28 days and after 1 and 3 years sealed.



Fig. 16. Non-steady-state chloride diffusion coefficients for different time of exposure to chlorides and at different delay in exposing the six mixes studied.

chloride solution after curing for 28 days or after one year sealed. Therefore, for that concretes, it seems that there is not a significant influence in the duration of the time previous to the exposure to chlorides, and of the age of the concrete in the coefficient obtained. The case of the rest of mixes, especially the case of mortars (dosages 5 and 6) is different, as there is a difference depending on the duration to the exposure to chlorides which, in addition, does not follow the same trend for the different mixes studied.

Concerning the chloride threshold values for depassivation of the rebar, numerous studies have

been already carried out to study the chloride threshold value for depassivation of the steel embedded in the different types of concrete using an accelerated method, whose set up is shown in Fig. 17 [8]. For making this test, a container is sealed to one of the surfaces parallel to the rebar and it is filled up with a 1 M NaCl solution. In the solution a stainless iron mesh is immersed and it is connected to the negative pole of a power supply. As positive electrode, a steel plate is electrically connected to the bottom surface of the sample, maintaining the electrical contact by a wet sponge. A voltage drop of 12 V is applied between the



Fig. 17. Experimental arrangement used for the chloride threshold value tests.

electrodes. During the tests, it is necessary to neutralize the solution with a continuous dropping of diluted acid, because of the increase in the concentration of hydroxyl ions, due to the hydrolysis of water. During the test, the electrical potential of the rebar have to be monitored, in order to detect the instant of the depassivation of the steel. Periodically, the power supply is switched off to also have the data of potential without the electrical field applied. The evolution of the potential of the steel indicates the depassivation of the rebar embedded in the specimen. Once this variation has been detected, the power supply has to be switched off and wait until depassivation in natural conditions. The averaged results obtained for the concrete and the filling mortar, in percentage of total chlorides on binder, are of 1.17% and 1.04% respectively, much higher than the typical value commonly accepted of 0.4%.

4.4.1.1. Modelling. In the case of the values of *D* and chloride threshold obtained have been introduced in a 3D model that helps to study the different possible scenarios and chloride concentrations in the drums. Fig. 18 shows one of the examples which consider constant and decreasing chloride concentration in the interior of the drums.

Being the attack by chlorides the cause of the most premature failures, during last years numerous publications can be found in the literature on the modelling of the chloride diffusivity [9–14]. However, these models have not been validated due to they are relatively modern and enough time has not passed to apply them in real structures with reliability. Thus, the exercise shown has to be considered theoretical. It tries to limit the chloride content in the drums but cannot be rigorously taken.

5. Final comments

Corrosion of reinforcement can be approximately modelled and accurately measured on-site. The periodical corrosion rate measurements from monitoring seems very necessary to assess present conditions of concrete structures and is a very useful tool in the case of cooling towers of power plants. Techniques based in the measurement of polarization resistance have been implemented in portable corrosion rate meters to obtain corrosion rate values, and corrosion-data loggers are now operative in different structures to monitor corrosion related parameters.



Fig. 18. Results of the 3D numerical model developed to study scenarios of chloride release and diffusion from the drums.

The establishment of a suitable model of service life of CBM used in repositories of radioactive wastes has the main difficulty of the lack of proper calibration of existing models and their use for much shorter service lives. In addition, there are still important items to be measured and collected as they provide information for characterization of the environment and for the correct identification of key parameters involved in long term durability.

Acknowledgements

Recognition is made to the funding provided by Enresa to support the investigation and Geocisa for the collaboration in the corrosion data collection.

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