Cesium diffusion in mortars from different cements used in radioactive waste repositories

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ABSTRACT

Cement-based materials are widely used for stabilizing and conditioning radioactive waste in low- and intermediate-level repositories.

In this study, the adequacy of four different types of mortars, obtained from four different commercial cements, CEM I, II and IV (A and B), to act as barrier to ^{137}Cs migration was analysed. Diffusion experiments using the in-diffusion (ID) method with constant tracer concentration in the reservoirs were carried out, at increasing experimental times (150, 380 and 1500 days approximately).

In the experiments of less duration, and especially in CEM II and CEM I, two different pathways for diffusion were identified (*fast* and *slow*), leading to a double porosity system. However, the "fast" contribution to Cs diffusion, in all cases, could be neglected at larger experimental time, indicating a rearrangement of the pore structure that becomes more tight and homogeneous over time. Thus, if the experiments last enough time, only one diffusion coefficient properly describe Cs transport in these systems.

Apparent diffusion coefficients, D_a, obtained for Cs, range between $6.0 \cdot 10^{-13}$ and $1.0 \cdot 10^{-14}$ m^2/s. Mortars produced with CEM IV (A and B) are the most efficient barrier for cesium transport, amongst those analysed in this study. Mortars with CEM II, which contain blast furnace slag, present the highest diffusion coefficient.

1. Introduction

Materials based on cements have many potential applications in the field of radioactive waste repositories. Concrete, mortar and cement have several uses in different repository concepts; for low and intermediate level waste the most important engineered barriers consist of cement-based materials. In high-level waste cement materials will be used for rock stabilization, limiting groundwater inflow, as well as for construction of engineered structures and for sealing of temporal or long-term service (Vieno et al., 2003). Also, cementitious materials may be used for conditioning/solidification and stabilization of the waste due to their favourable properties (Glasser et al., 1986; Atkinson and Nickerson, 1988; Atkins and Glasser, 1992; Batchelor, 2006; Ochs et al., 2016). Cements contribute both to physical and chemical containment of the radionuclides in the wastes because they can act as a barrier and because their large surface area provides high sorption capacity for radioactive species. Cementitious materials influence the chemical properties of the repository near-field, providing alkaline conditions, which favour radionuclide immobilization by sorption and low solubility. Under these conditions, radionuclide transport takes place

mainly by a diffusion process, slowed-down by sorption. Diffusion coefficients in cementitious materials are important parameters to be used in performance assessment calculations for low and intermediate level radioactive waste repositories.

Ion penetration in cement-based materials is largely controlled by their pore structure. The diffusion of ions is related to the volume of pores and their size and interconnectivity. Porosity, pore sizes and pore interconnectivity or distribution, are believed to be the main factors that affect diffusion of conservative ion through the mortars (Li et al., 2015).

Ion exchange resins used to purify coolant water in a nuclear power plant, frequently contain large amounts of ^{137}Cs, produced by the fission of uranium in nuclear reactors, and are typically part of low and intermediate level waste (LILW) (Ochs et al., 2016). There are few experimental data about Cs diffusion in mortars and the diffusion coefficients values show a very large variability, mainly due to the different experimental conditions. Sarott et al. (1992), performed through-diffusion experiments with hardened cement paste (HCP) and artificial pore water at pH 13.3 under oxic and anoxic conditions to study the cement carbonation; diffusion decreased with time by CaCO$_3$

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precipitation; in anoxic condition an average Cs effective diffusion coefficient, D_e, of $1.6 \cdot 10^{-10}$ m^2/s was found. Skagius et al. (1999) found for structural concrete a D_e (Cs) = $2 \cdot 10^{-12}$ m^2/s; Sato et al. (2015) fitted the experimental results on concrete with D_a values of $5 \cdot 10^{-13}$ and $4 \cdot 10^{-14}$ m^2/s.

Different authors (Idemitsu et al. (1997); Sato et al. (2015), Atkinson and Nickerson (1988)) observed that for Cs diffusion, mortars and cements should be considered as "double porosity" media; their assumption was that different diffusion paths exist in the shallow (surface) or deep part of the sample (matrix). In particular, Idemitsu et al. (1997) performed diffusion experiment with Cs in water-saturated mortar and found that the measured penetration profiles of the tracer were composed of two parts: a steep slope near the surface, related to diffusion path was through fissures with a width of a few microns, and a gradual slope in the mortar interior, corresponding to the intact mortar network of submicron pores. Sato et al. (2015) studied the diffusion of ^{137}Cs, ^{154}Eu and ^{241}Am in concrete and mortar and also in this case, they needed two different D_a to fit the experimental data for the "surface" and "deep" region. The D_a values obtained by Sato et al. (2015) for Cs are: $1 \cdot 10^{-13}$ m^2/s ("surface") and D_a $5 \cdot 10^{-13}$ m^2/s ("deep"). Atkinson and Nickerson (1988) also observed that for Cs diffusion, the cement showed a fast and slow diffusivity networks, and that the measured Cs profile appeared to be composed of two parts with different D_a (surface: $3.3 \cdot 10^{-14}$ m^2/s and deep: $4 \cdot 10^{-12}$ m^2/s). Idemitsu et al. (1997) fitted the experimental results obtained by Atkinson and Nickerson (1988) also using two different D_a ($1 \cdot 10^{-12}$ m^2/s and $1 \cdot 10^{-14}$ m^2/s).

Yuan (2009), studying chloride transport in concrete, also found that the experimental results are in agreement with the existence of different regions with their own retardation properties and with different diffusion coefficients, and concluded that the chloride diffusion coefficients are "depth dependent" because of the changes in pore structure.

The aim of this work is to obtain diffusion coefficients for Cs in mortars from different cements used for radioactive waste immobilization and to discuss the concept of "double porosity" on the bases of diffusion data obtained at different experimental times. Our hypothesis is that the porosity of the system is changing with time and tends to homogenise if enough experimental time is allowed, in contrast to the hypothesis of the existence of "shallow" and "deep" regions. Therefore, diffusion time must be considered a very important parameter for the D_a determination in these systems.

2. Materials and methods

Four commercial cements CEM I 42.5 R/SR, CEM II/A-S 42.5 N/SR, CEM IV A (V) 32.5 N and CEM IV B (V) 32.5 N, were used in this study. These cements have a low tricalcium aluminate (C3A in cement nomenclature) and alkali content, to prevent sulphate or alkali-aggregate attack (Andrade et al., 2006). General cement composition can be found in Table 1. CEM I, CEM IV A and B incorporate fly ash. Fly ash arises as a product of coal combustion: pulverised, micron-sized coal particles are combusted in air suspension where their mineral matter fuses. Fly ash is an extremely variable material and does not constitute a single material with well-defined properties (Atkins and Glasser, 1992). Blast furnace slags used for cements are usually chemically homogeneous

and constant in composition and normally glassy, to develop similar hydraulic properties than cement (Atkins and Glasser, 1992). CEM II is the only cement studied here that incorporates blast furnace slag.

A mortar is a homogeneous mixture of fixed proportions of cement, sand and water. In this work, silica sand with a size smaller than 2 mm was used to obtain mortars with a cement: sand: water relation of 1:1.75:0.45. "CEM I" samples were already prepared with 64% of CEM I and 36% of fly ash.

Mortars were prepared in PVC moulds and left to hydrate at room temperature and a relative humidity of 100%, for curing during 28 days. Mortars bulk densities determined form the ratio of the weight of the water saturated sample and its geometrical volume were very similar for the four used materials, between 2.1 and 2.2 g/cm^3.

The composition of the liquid phase influences the diffusion process. To prevent strong mortar degradation during the diffusion experiment, the aqueous phase must be in equilibrium with the solid phase. Furthermore, the sample must be fully saturated to carry out the diffusion tests. To perform the experiments, synthetic cement pore water was prepared under oxic conditions using boiled Milli-Q water, and adding crushed and sieved (< 1 mm) mortars in a solid/liquid ratio of 10 g/L.

The water in contact with each solid was maintained under stirring until constant pH and conductivity were reached, approximately 30 days. Chemical composition of these four waters, filtered by 0.45 μm, is presented in Table 2. CEM II water has the highest calcium content, alkalinity and electric conductivity and lowest aluminium content. These waters were stored in dark and closed bottles until their use.

Cylindrical mortars samples, of 5 cm length and 4.5 cm radius, were sealed completely with epoxy resin and then cut in a half, leaving one face of the mortar uncovered, Fig. 1 (Left). In this way, two samples of the same mortar were obtained from each test core.

Diffusion experiments were performed using the in-diffusion (ID) method with constant tracer concentration in the reservoir. Decrease of concentration due to Cs sorption on mortars was corrected periodically spiking a small quantity of tracer. Each sample was introduced in a reservoir with 100 mL of mortar pore-water (previously equilibrated with 10 g/L of the corresponding solid). As the mortar samples were stored in a humid chamber, it was considered that two months were enough to reach sample saturation. Afterwards the tracer, ^{137}Cs (supplied by Eckert & Ziegler Isotope Products as CsCl without carrier), was added to the reservoir and the diffusion experiment started.

The ^{137}Cs initial activity, C_0, was approximately 3100 cpm/mL. The tracer can diffuse through the uncovered mortar side and at the end of experiment (the diffusion time estimate for each set), the tracer concentration profile was obtained.

Three sets of experiments were performed for each mortar at increasing experimental times. The times selected for the different sets of experiments were: 150 days, 380 days and four years approximately.

Table 1
Cement composition.

Cement	CEM I 42.5 R/SR	CEM II/A-S 42.5 N/SR	CEM IV A (V) 32.5 N	CEM IV B (V) 32.5 N
Clinker	95–100	83	70	58
Fly ash	–	–	29	40
Slag	–	12	–	–
Minor compounds	1	5	1	2

Table 2
Chemical composition of the different mortars pore water (mmol/L).

| Element	CEM I	CEM II	CEM IV A	CEM IV B
Cl$^-$	$4.78 \cdot 10^{-3}$	$2.20 \cdot 10^{-2}$	$8.46 \cdot 10^{-2}$	$1.18 \cdot 10^{-2}$
SO$_4^{2-}$	$9.16 \cdot 10^{-2}$	$6.67 \cdot 10^{-2}$	$1.08 \cdot 10^{-1}$	$1.23 \cdot 10^{-1}$
NH$_4^+$	$< 2.77 \cdot 10^{-2}$	$< 2.77 \cdot 10^{-2}$	$< 2.77 \cdot 10^{-2}$	$< 2.77 \cdot 10^{-2}$
Ca^{2+}	4.77	9.96	4.96	4.09
Mg^{2+}	$< 1.23 \cdot 10^{-3}$	$< 1.23 \cdot 10^{-3}$	$< 1.23 \cdot 10^{-3}$	$< 1.23 \cdot 10^{-3}$
Na$^+$	$1.00 \cdot 10^{-1}$	$3.57 \cdot 10^{-1}$	$2.52 \cdot 10^{-1}$	$1.48 \cdot 10^{-1}$
K$^+$	$4.60 \cdot 10^{-1}$	$3.07 \cdot 10^{-1}$	$7.67 \cdot 10^{-1}$	$6.39 \cdot 10^{-1}$
Al	$1.37 \cdot 10^{-1}$	$1.74 \cdot 10^{-2}$	$1.56 \cdot 10^{-1}$	$2.00 \cdot 10^{-1}$
Fe	$< 5.37 \cdot 10^{-4}$	$< 5.37 \cdot 10^{-4}$	$< 5.37 \cdot 10^{-4}$	$< 5.37 \cdot 10^{-4}$
SiO$_2$	$4.66 \cdot 10^{-2}$	$2.33 \cdot 10^{-2}$	$3.16 \cdot 10^{-2}$	$4.99 \cdot 10^{-2}$
Alkalin. (meq/L)	11.82	21.49	8.86	10.51
pH	12.2	12.5	12.3	12.2
Cond. (μS/cm)	2300	3980	2750	2010
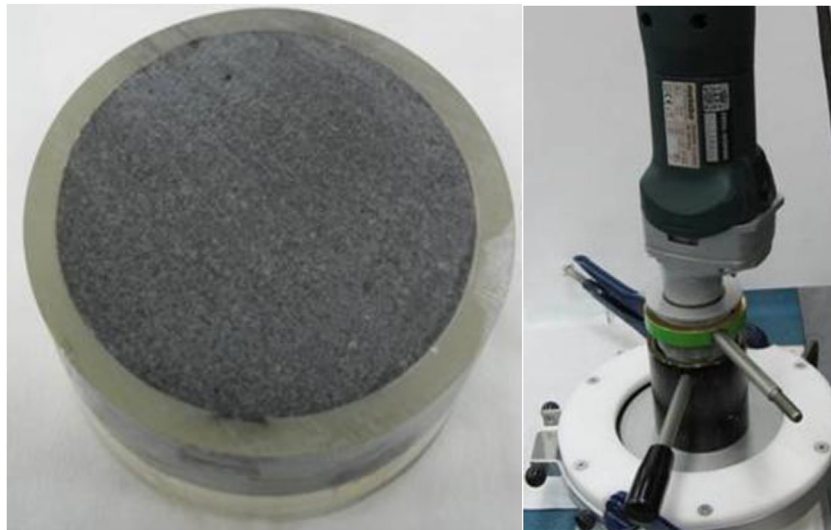
Fig. 1. (Left) Mortar sample sealed with a side free. (Right) Profile grinder used to obtain the concentration profile.

CEM II, was analysed only in the first two sets, because after 380 days, the penetration depth was larger than the sample thickness.

In-diffusion experiments are usually designed so that the tracer cannot reach the closed extreme of the sample and the experiment could be treated as a one-dimensional case of diffusion into a semi-infinite medium. The general diffusion equation for one-dimensional analysis under non-steady state condition is described by Fick's second law as:

$$\frac{\partial C}{\partial t} = D_a \frac{\partial^2 C}{\partial x^2}$$

(1)

where C is the solute concentration in the pore water that depend on time t and distance x; and D_a is the apparent diffusion coefficient, describing the diffusion of a reactive solute in a porous medium.

If solute concentration remains almost constant during the experimental time, and the following initial and boundary conditions are valid,

C (x > 0, t = 0) = 0

C(x = 0, t > 0) = C$_0$

C(x = ∞, t > 0) = 0

the concentration profile within the sample can be fit by the following analytical solution (Crank, 1975; Grathwohl, 1998):

$$\frac{C}{C_0} = erfc\left(\frac{x}{2\sqrt{D_a t}}\right)$$

(2)

where C is the cesium concentration at distance x from the solution-mortar interface at time t, C_0 is the concentration of cesium at the interface between mortar and solution, deduced from the experimental data by the fitting procedure of eq (2), and D_a the apparent diffusion coefficient, and $erfc$ denotes the complementary error function).

After the diffusion time, the ^{137}Cs concentration profile was obtained by slicing the mortar sample with a grinder machine (Grinder Profile from Germann Instruments) at depth increments, Fig. 1 (Right). The quantity of material extracted each time from a uniform sectional area was used for calculating the depth in the mortar sample. From the total mass extracted and total drilled volume a sample density (approximately 2 g/cm^3) is obtained. From these data a thickness for each sample can be calculated. These depths were in good correlation with the direct measurement of the lengths. In the graphs of concentration profiles, the distance corresponds to the middle of the sample.

The ^{137}Cs activity in the collected samples was measured through the gamma emission of ^{137m}Ba in equilibrium with Cs using a Cobra-II auto-gamma Packard counter (NaI detector). The ^{137}Cs concentration in the reservoir was controlled periodically. Taking in account the weighing procedure and counting efficiency the relative uncertainties calculated were < 5%.

3. Results and discussions

Fig. 2 shows the concentration profiles obtained for each mortar after a diffusion time of about 150 days. The experimental data clearly show a much deeper penetration of Cs in CEM II, whereas in CEM IV B the penetration is the smallest. In all the cases, the diffusion profiles shown in Fig. 2 could not be properly fitted using the simple analytical solution presented in equation (2), i.e. considering a single apparent diffusion coefficient, D_a. The fit of the curves with equation (2) is shown in Fig. 2 as a dotted line. As can be observed in Fig. 2, the simulation with a unique D_a, tends to under-estimate the diffusion depth, especially when it is deeper (CEM II and CEM I). This behaviour could be related to the heterogeneity of the materials pore structure, giving rise to different transport paths, as already observed by other authors.

Therefore, a way to fit the experimental data is considering a "double porosity system". In this approach, the sample volume can be divided into two different regions, each of which has own retardation properties described by individual apparent diffusion coefficients, i.e. two distinct diffusive pathways are considered. Part of Cs will penetrate faster through the large pores and slower in the small ones. In order to apply this approximation it is necessary to evaluate the fraction of Cs travelling in each path. By a trial and error procedure, it was observed that the best fit of the data for CEM I and CEM II mortars in the first set of experiments was obtained considering that a 25% of ^{137}Cs diffuses faster (fast apparent diffusion coefficient, D_{aF}), whereas the other 75% diffuses in the slower path (slow apparent diffusion coefficient D_{aS}). The percentage of mass in the two pathways was fixed at 25% and 75% for the rest of the experiments.

It can be considered that the first porosity corresponds to the connected pores in the mortar (micro-sized), and the second to the cement matrix, which possess larger surface area, increasing the retention by sorption.

Considering that the sample volume can be divided in two different regions each one with its retardation properties, the equation to fit the results is:

$$\frac{C}{C_0} = 0.25 \cdot erfc\left(\frac{x}{2\sqrt{D_{aF} t}}\right) + 0.75 \cdot erfc\left(\frac{x}{2\sqrt{D_{aS} t}}\right)$$
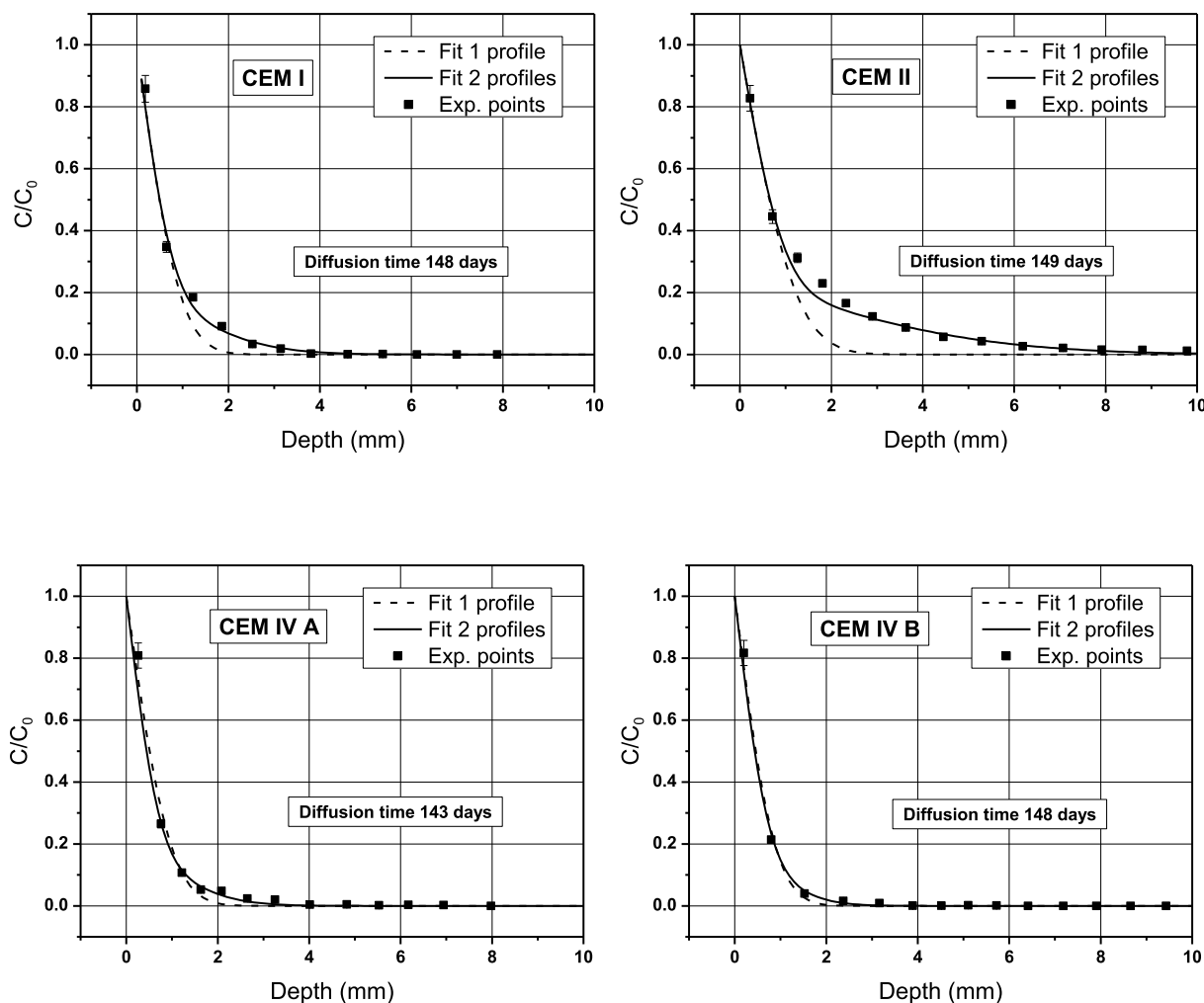
(3)

Fig. 2. Diffusion profiles obtained for CEM I, CEM II, CEM IVA and CEM IV B, for the first set of experimental time. The fit considering one and two contributions to diffusion are shown.

This approximation improved the data fit, as indicated in Fig. 2 (continuous line). The apparent diffusion coefficients obtained by the fits are summarised in Table 3.

Fig. 3 shows the diffusion profiles obtained for the four materials in the second set of the experiments (380 days approximately). Also in

these experiments and especially in the case of CEM II and CEM I, the simplest approach described by equation (1) with a single diffusion coefficient, was not satisfactory enough to fit the experimental data (dotted line).

Therefore, also in this case, two diffusion coefficients were used. In

Table 3
Apparent diffusion coefficients (m^2/s) obtained in the different tests.

| Mortar with Cement	Diffusion time (days)	1 profile D_a	2 profiles D_{aS} "slow"	2 profiles D_{aF} "fast"
CEM I	148	–	$1.4 \cdot 10^{-14}$	$1.3 \cdot 10^{-13}$
	380	–	$0.7 \cdot 10^{-14}$	$1.5 \cdot 10^{-13}$
	1494	$1.5 \cdot 10^{-14}$	$1.1 \cdot 10^{-14}$	$3.5 \cdot 10^{-14}$
Average value		$\mathbf{1.5 \cdot 10^{-14}}$	$\mathbf{(1.1 \pm 0.3) \cdot 10^{-14}}$	$\mathbf{(1.0 \pm 0.6) \cdot 10^{-13}}$
CEM II	149	–	$2.2 \cdot 10^{-14}$	$6.1 \cdot 10^{-13}$
	380	–	$1.0 \cdot 10^{-14}$	$8.9 \cdot 10^{-13}$
Average value		**-**	$\mathbf{(1.6 \pm 0.6) \cdot 10^{-14}}$	$\mathbf{(7.5 \pm 1.4) \cdot 10^{-13}}$
CEM IV A	143	$2.3 \cdot 10^{-14}$	$1.2 \cdot 10^{-14}$	$7.9 \cdot 10^{-14}$
	378	$2.4 \cdot 10^{-14}$	$1.6 \cdot 10^{-14}$	$7.8 \cdot 10^{-14}$
	1494	$2.2 \cdot 10^{-14}$	$1.8 \cdot 10^{-14}$	$2.6 \cdot 10^{-14}$
Average value		$\mathbf{(2.3 \pm 0.1) \cdot 10^{-14}}$	$\mathbf{(1.5 \pm 0.3) \cdot 10^{-14}}$	$\mathbf{(6.1 \pm 3.5) \cdot 10^{-14}}$
CEM IV B	148	$1.8 \cdot 10^{-14}$	$1.2 \cdot 10^{-14}$	$5.0 \cdot 10^{-14}$
	380	$1.2 \cdot 10^{-14}$	$1.0 \cdot 10^{-14}$	$3.4 \cdot 10^{-14}$
	1494	$0.7 \cdot 10^{-14}$	$0.7 \cdot 10^{-14}$	$0.7 \cdot 10^{-14}$
Average value		$\mathbf{(1.2 \pm 0.6) \cdot 10^{-14}}$	$\mathbf{(1.0 \pm 0.3) \cdot 10^{-14}}$	$\mathbf{(3.0 \pm 2.3) \cdot 10^{-14}}$
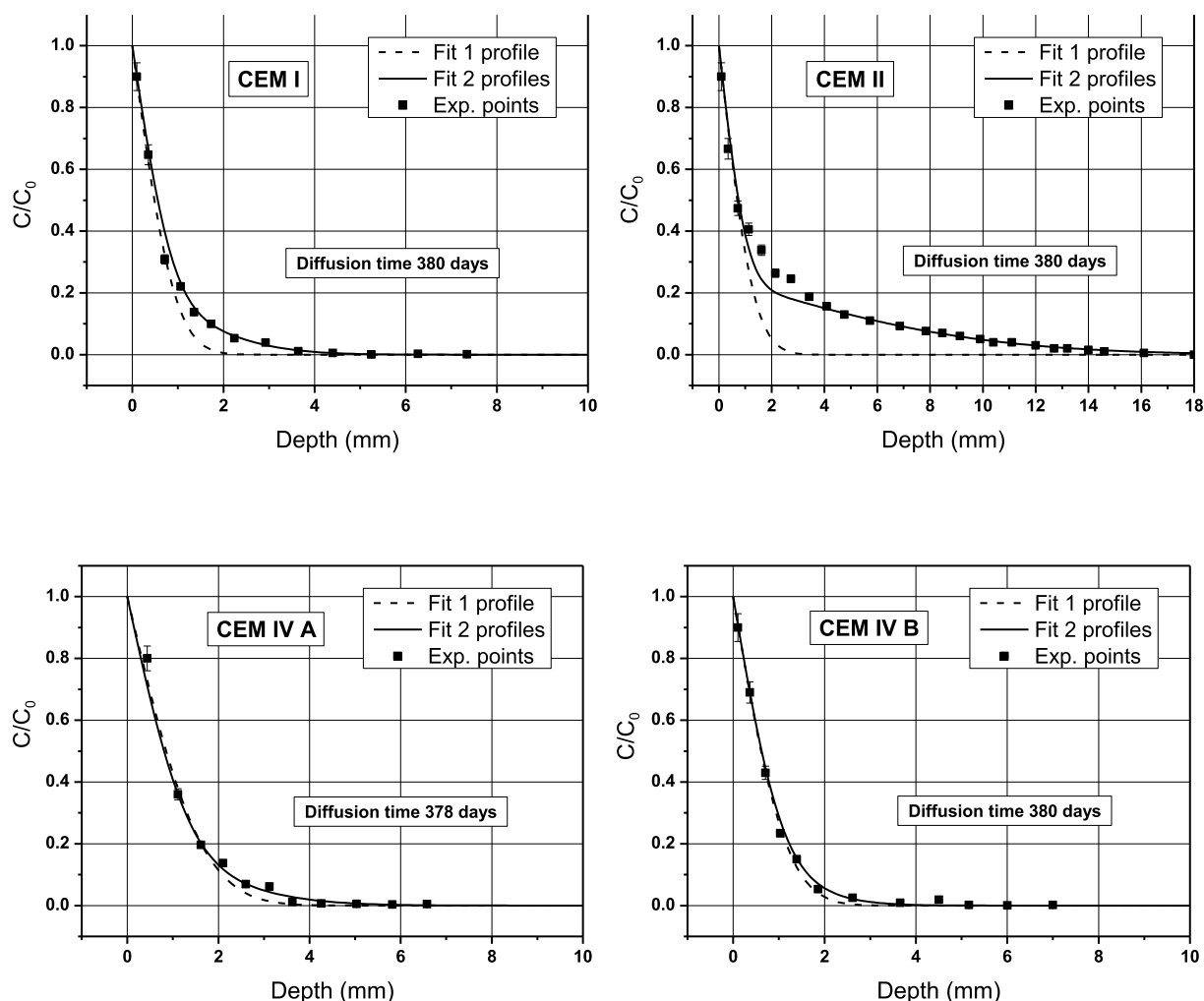
Fig. 3. Diffusion profiles obtained for CEM I, CEM II, CEM IVA and CEM IV B, for the second set of experiments. The fit considering one and two profile are shown.

the case of CEM II a distribution of 33% and 67% (instead of 25%/75%) could fit better the experimental profile. The fit of the data using Eq (3) is represented as a continuous line and the obtained Da are summarised in Table 3.

Fig. 4 shows the third set of diffusion experiments, which lasted approximately 4 years, for CEM I and CEM IV A and B. The profile of CEM II after 4 years is not presented because, after this time, Cs concentration was homogeneous throughout the sample, thus a diffusion profile cannot be obtained.

It is interesting noticing that diffusion profiles obtained with these long experiments could be quite satisfactorily simulated considering the simple approach of equation (1), and the quality of the fit was similar when using one profile (dotted line) or the superposition of two (continuous line).

This result clearly indicates that the diffusive behaviour of Cs at long times is much less affected by the material heterogeneities.

Therefore the observed "double porosity" it is most probably related only to the initial stages of the experiments, as the cement tend to evolve over time to a more homogeneous structure throughout the entire sample (thus not related to *shallow* of *deep* regions).

Most of the previous diffusion tests were carried out in a short experimental time (for example, Atkinson and Nickerson (1988) and Sato et al. (2015) used 21 and 16 days), therefore the material could have not attained its physical-chemical equilibrium (so being not fully representative of that existing in a repository).

As a main conclusion, the experimental time is very important for an adequate determination of D$_a$ to be used in performance assessment.

If the material is clearly evolving with time a realistic evaluation of diffusion coefficients can be done only if enough experimental time is adopted. Diffusion coefficients determined at short experimental times, are more affected by the diffusion in the fissure, they can be used as "conservative values" but they are not actually representative of the characteristics of the cement matrix.

All the apparent diffusion coefficients obtained in this work, Table 3, vary from $0.7 \cdot 10^{-14}$ to $8.9 \ 10^{-13} \ m^2/s$, for the faster path and from $0.7 \cdot 10^{-14}$ to $2.2 \ 10^{-14}$ for the slower one.

The largest difference in diffusion coefficients are observed in the faster path, whereas similar diffusion coefficients are measured for all the cements in the slowest path. In CEM I and CEM II, the transient fast contribution is more pronounced.

Table 3 also presents the average value and the standard deviation of the diffusion coefficients obtained taking in account all the experimental results. "Fast" apparent diffusion coefficients could be taken as a conservative value of D$_a$, even it will be less relevant at longer times, of interest for radionuclide waste repositories.

Amongst the four types of cement studied the most effective as a barrier to the diffusion of ^{137}Cs is, according to our laboratory test, the CEM IV, in its two modalities. A is a cement with 70% clinker and 29% fly ash and B is a cement with 58% clinker and 40% fly ash. Aggarwal et al. (2000) found that for Ordinary Portland cement with Pulverised Fuel Ash (PFA/OPC) sorb Cs more strongly than the OPC Blast Furnace Slag (BFS/OPC) cesium distribution coefficients, R$_d$, varies from 21 to 1 mL/g for PFA and BFS respectively.

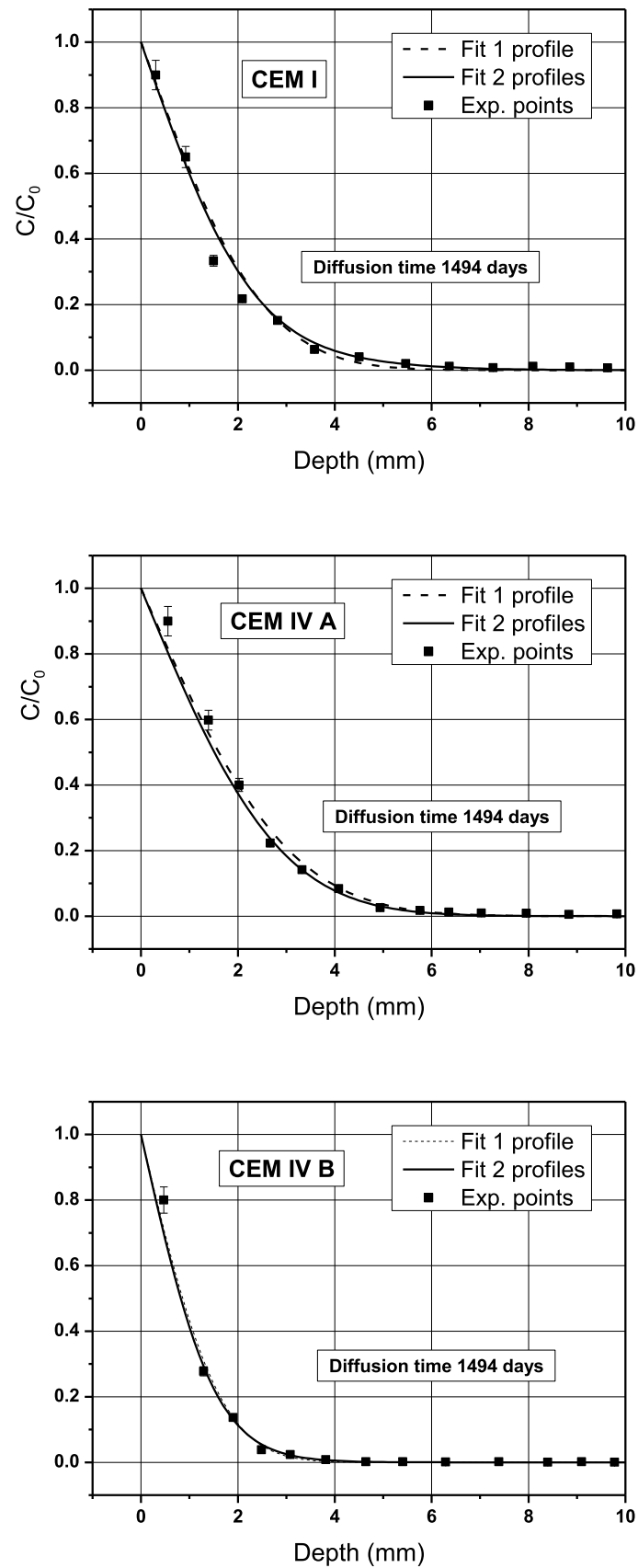Cement CEM II with blast furnace slag (12%) presents the highest

Fig. 4. Diffusion profiles obtained for CEM I, CEM IVA and CEM IV B, for the third set of experiments. The fit considering one and two profile are shown.

apparent diffusion coefficient that can be related to the highest pH, calcium content and electrical conductivity in the pore water. Furthermore, the presence of larger cation content (Ca) may affect Cs sorption due to competitive effects favoring transport.

4. Conclusions

Diffusion coefficients for Cs in mortars from different cements were determined by in-diffusion method.

For CEM IV A and B, apparent "fast" diffusion coefficient (the conservative value) is in the range of $(3–6) \cdot 10^{-14}\,\mathrm{m}^2/\mathrm{s}$; for CEM I an average value of $D_a = 1 \cdot 10^{-13}\,\mathrm{m}^2/\mathrm{s}$ was measured. The highest values, $7.5 \cdot 10^{-13}\,\mathrm{m}^2/\mathrm{s}$, was obtained for the mortar with CEM II.

Amongst the four types of cement studied the most effective as a barrier to the diffusion of $^{137}\mathrm{Cs}$ is, according to our laboratory test, the CEM IV.

It was shown that cesium concentration profiles in mortars cannot be adjusted with a single diffusion coefficient, especially in the experiments of less duration. If much longer experimental time is considered, this effect is attenuated and only one diffusion coefficient is enough to explain diffusion data.

Cement is not an inert material with a fixed pore distribution, when water or solute transport through the pore exist, alteration into its pore structure due to cementations process, can change the pores size and distribution. The experimental time is very important for an adequate determination of D_a to be used in performance assessment, considering that the material should have reached physical-chemical equilibrium. Diffusion coefficients determined at short experimental times depends basically on the existence of larger pores but they are not actually representative of the characteristics of the cement matrix.

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