

Immobilization of cesium and strontium radionuclides in zeolite-cement blends

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Received 5 September 2005; received in revised form 29 November 2005; accepted 12 December 2005

Available online 18 January 2006

Abstract

Zeolites are preferred ion exchange materials for the removal of radioisotopes from aqueous nuclear wastes because of their selectivity, radiation and temperature stability, and good compatibility with the cement matrix. Loaded materials, which are not regenerated, are preferably embedded in a solid matrix prior to ultimate storage in a repository. The aim of the present study is to investigate the possibility of solidifying exhausted synthetic zeolite A, loaded with ^{137}Cs and/or ^{90}Sr radionuclides, in Ordinary Portland Cement (OPC). Several factors affecting the characteristics of the final solidified waste product towards safe disposal such as mechanical strength and leaching behavior of the radionuclides have been studied. A simplified mathematical model based on diffusion mechanism for cylindrical geometry waste matrix has been simulated to predict the release rates of the investigated radionuclides from cement matrix. The predicted values are discussed in relation to experimentally observed leach rates to confirm the proposed mechanism in the model. The obtained results showed that the presence of zeolite A in the final cemented wastes improve the mechanical characteristics of the solidified cement matrix (mechanical strength and setting times) towards the safety requirements and reduce considerably the radionuclides leach rates.

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Keywords: Immobilization; Radioactive wastes; Portland cement; Zeolite; Mathematical modeling

1. Introduction

Low and intermediate level radioactive wastes are produced from diverse applications of radionuclides in industry, medicine, radioisotope production facilities and fuel processing plants. These wastes need treatment to reduce the quantities of radioactive contaminants to the level, which allows safe discharge of the decontaminated liquid to the environment and safe disposal of the concentrated radionuclides according to the international requirements and national regulations [1]. The objectives of the liquid waste processing are to immobilize the radioactive elements and to reduce the volume to be stored. The solidified product must be non-dispersible, insoluble and with good mechanical and structural stability. Portland cements are met the acceptance criteria for immobilizing radioactive wastes because of their low cost, high density, durability and amenability to simple processing techniques [2].

Immobilized waste forms can be safely isolated from the biosphere for a long time in a radioactive waste repository. However, despite the engineering and natural barriers of radioactive waste repositories, the radioactive waste forms would eventually be in contact with ground water, and the releases of radioactive species from the waste forms would occur by the leaching mechanism [3]. Hence, there is a considerable interest in understanding the diffusive transport of radionuclides through cement matrix. Several leaching studies [4–6] have been reported using different methods to study the temporal distribution of radionuclides in the leachant medium. Also, a significant number of works dealing with the immobilization of different radionuclides with cement and cement mixed with different additives have been carried out [7–13]. In our previous work [14], the International Atomic Energy's (IAEA's) standard leach method [15] has been employed to study the leach pattern of cesium and cobalt radionuclides solidified in cement and cement mixed with two different ratios of locally produced silica fume and ilmenite. The results indicated that addition of up to 15% silica fume and/or ilmenite to the final cement matrix decreases the leaching rate of each studied nuclide. In the present work,

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immobilization of spent zeolite A, loaded with ^{137}Cs and/or ^{90}Sr radionuclides, with OPC have been carried out. Several properties such as the effect of zeolite addition percent and water type immersion on the mechanical strength of the solidified waste package were examined. The leach characteristics of the investigated radionuclides from a simple cement waste form and a waste form consisting of granules of contaminated zeolite ion exchange in a cement matrix have also been studied using IAEA's standard leach method. Release rates, diffusion coefficients, and leachability indexes of both radionuclides were determined using diffusion release models.

2. Experimental

2.1. Equipments

2.1.1. Load compression machine

Load compression machine, Wykeham France Eng., model WF was used to perform the compression test according to the British Standard. The compression machine has a rigid frame assembly and provides the reaction of the loading of a movable piston against the sample produced to this equipment.

2.1.2. Vicat needle apparatus

Vicat apparatus with different needles was used to determine some of the physical properties as setting times of the studied samples.

2.1.3. Total X-ray fluorescence instrument

Quantitative elemental analysis of the used synthesized zeolite A was accomplished by total X-ray fluorescence instrument of type Philips, Vacuum-Spectrograph, Italy, using Ga solution as internal standard for analysis measurements.

2.1.4. Gamma spectrometer

Gamma spectrometer was used to measure the gamma energy emitted from ^{137}Cs and/or ^{90}Sr radionuclides. The gamma spectrometer used has $2'' \times 2''$ sodium iodide (NaI) crystal activated with thallium. The crystal was connected to a multichannel analyzer which had 256 channels attached with preamplifier. The equipment was manufactured by the Nuclear Excellence in Nuclear Instrumentation, Model 800A.

2.2. Materials

OPC was provided from Suez Cement Company, Suez, Egypt and its chemical composition was given in Table 1. Zeolite A was synthesized in its sodium form from mixture consisting of sodium aluminosilicate substrate contacted with sodium hydroxide. In 1 L polypropylene bottle supplied with a magnetic stirrer, 7.88 g SiO_2 and 104.63 g H_2O was mixed. Then, 10.5 g NaOH was added and exactly 22.8 g NaAlO_2 was also added drop wise for about 2 h. The resultant product was aged over night at 353 K for completion of the reaction. The inorganic amorphous product was collected on Buchner funnel, washed with 3 L of distilled water for removal of unreacted species

Table 1
Chemical composition of ordinary Portland cement

Chemical	Wt. %
CaO	63
SiO_2	20
Al_2O_3	6
Fe_2O_3	3
MgO	1.5
SiO_3	2
Na_2O	0.5
K_2O	0.5
Others	1.0

and partially dried. The crystallinity, morphology as well as chemical composition of the prepared material were measured using X-ray diffraction (XRD); X-ray fluorescence (XRF), differential thermal analysis (DTA), thermal gravimetric analysis (TGA), and Scanning Microscope. The results have been presented elsewhere [16]. Based on these results, the oxide chemical composition of the synthetic zeolite A can be expressed via the formula: $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 1.85 \cdot \text{SiO}_2 \cdot 5.1 \text{H}_2\text{O}$. The theoretical cation exchange capacity (CEC) of synthetic zeolite A in Na^+ form equals 5.45 mequiv./g as calculated from the chemical formula. The quantitative elemental analysis as given in Table 2, indicated that the Si/Al ratio was about 0.82, which is in the characteristic range of zeolite A [17]. The BET surface area of the solid powder, measured after the thermal treatment for 2 h at 473 K, was $634.52 \text{ m}^2/\text{g}$. The radioactive tracers were delivered by Amersham Radioanalytical Center (England).

2.3. Loading of zeolite A with radionuclides

Inactive zeolite A was wet-sieved to provide uniform fractions. Cesium and strontium were exchanged onto zeolite A by contacting 1 g of zeolite A with 50 ml of 0.04 M CsCl and/or SrCl_2 solutions (labeled with ^{137}Cs and/or ^{90}Sr) for 3 days. Detailed description of the ion exchange procedure needed to prepare loaded zeolites was presented elsewhere [18–20].

2.4. Preparation of specimens

2.4.1. Mixing

Cement pastes were prepared by mixing plain OPC with different ratios (2, 4, 6, 8, and 10 wt.%) of loaded zeolite A at a water/cement (w/c) ratio of 0.35. The required amount of cement

Table 2
Quantitative elemental analysis of synthesized zeolite A using X-ray fluorescence spectroscopy

Element	Concentration (%)
Na	27.79
Al	39.43
Si	32.14
Ca	0.076
Ti	0.026
Mn	0.008
Fe	0.015

was placed on a smooth non-absorbent surface, and a crater was formed in the center. The required amount of mixing water was poured into the crater by the aid of a trowel. The mixing operation was then completed by continuous vigorous mixing using an helix grout mixer at a 1500-rpm speed for duration of 4 min. At the end of mixing, paste was directly poured into moulds.

2.4.2. Moulding and curing

Polyethylene cylindrical shape moulds with internal height/diameter ratio of 2.0 (2.0 cm diameter and 4.0 cm height) was used to prepare cement pastes. Paste was placed in the moulds in two approximately equal layers. Each layer was compacted and passed along the surface of the moulds until homogenous specimen was obtained. After the top layer was compacted, the moulds was then vibrated for 2 min to remove any air bubbles and to have a better compaction of the paste and even the top surface of the mould was smoothed by the aid of thin edged trowel. Immediately after moulding, moulds were kept in air for 24 h. The moulds were demoulded and were cured during 28 days in a humid atmosphere (100% RH) at $25 \pm 2^\circ\text{C}$.

2.5. Mechanical strength

The mechanical strength test, given as applied maximum load divided by a cross-sectional area of a specimen, was performed to confirm the integrity of waste forms. The specimen of a cylindrical shape with the height/diameter ratio of 2.0 (2.0 cm diameter and 4.0 cm height) would be desirable for the test because the restriction effect, the increase effect of apparent strength by virtue of friction between loading plate and specimen, could be reduced to a minimum. The criterion of mechanical strength for cement waste form is >500 Psi (3.4 MPa) as recommended by the US Nuclear Regulatory Commission (NRC) Standard [21].

2.6. Water immersion

In order to study the effect of water immersion on the mechanical strength, different specimens of plain OPC and that mixed with 4 wt.% synthetic zeolite A were prepared and immersed in three different leachant; tap water; distilled water and ground water. The ground water used as immersion medium is obtained from Abu Zaabal well (well no. 202), which is one of the nearest ground water well to Inshas nuclear site. The chemical composition of the ground water (Table 3) was investigated using suppressed ion chromatographic system (Dionex-2000 i, USA) coupled with conductivity detection mode except for bicarbonate concentration, which was investigated by titration method using phenolphthalein as indicator.

Table 3
Chemical composition of ground water

TDS (ppm)	pH	Soluble cations (ppm)				Soluble anions (ppm)		
		K ⁺	Na ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	HCO ₃ ⁻
1050	7.2	23	149	13	74	137	317	272

2.7. Static leaching test

Static leaching tests were performed, using ground water solution, to study the leaching of cesium and strontium ions from hardened blocks of cement and cement mixed with 4 wt.% synthetic zeolite A. The IAEA's standard test proposed by Hesse [15] was applied. All prepared samples (2.0 cm diameter and 2.0 cm height) were stored in laboratory at ambient temperature ($25 \pm 2^\circ\text{C}$) for 28 days during curing time. Each sample was immersed in beaker containing 50 ml ground water for certain time intervals and 1 mL of leachant was taken, dried, and counted using a gamma spectrometer. The cumulative leach fraction (cm) was calculated according to the following equation:

$$\text{cumulative leach fraction} = \left(\frac{\Sigma A(t)}{A_0} \right) \left(\frac{V}{S} \right)$$

where $\Sigma A(t)$ is the cumulative radioactivity leached, A_0 the initial radioactivity present in specimen, V the volume of specimen, cm^3 , and S is the exposed surface area of specimen, cm^2 .

3. Mathematical model

Radionuclides from the cement block are transferred to the surrounding water by dissolution to the interstitial water, diffusion through interstitial water to the surface of the cement block, and eventual release to the water. Since the geometry of the cement block is well defined, the following equation is used to represent the migration of nuclides from the cylindrical cement block to the surrounding water

$$\frac{\partial C}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(rD \frac{\partial C}{\partial r} \right) \quad (1)$$

where C is the concentration of radionuclide in cement block, mol/L, D the diffusion coefficient, cm^2/s , r the radial distance from the center of the block, cm, and t is the time, s.

The initial and boundary conditions for Eq. (1) are given by

$$C(r, 0) = C_0, \quad |r| \leq R \quad (2)$$

$$C(R, t) = 0 \quad (3)$$

$$\frac{\partial C}{\partial r} \Big|_{r=0} = 0 \quad (4)$$

where C_0 is the initial concentration of radionuclide in the cement block and R is the radius of the cement block.

Using initial and boundary conditions, the solution for Eq. (1) can be written as follows:

$$C = C_0 \sum_{n=1}^{\infty} \frac{2J_0(r\alpha_n/R)}{\alpha_n J_1(\alpha_n)} \exp[-(D\alpha_n^2 t/R^2)] \quad (5)$$

where J_0 and J_1 are the first kind of Bessel functions of order 0 and 1, respectively, and α_n are the roots of $J_0(\alpha) = 0$.

The leaching flux $F(t)$ from the surface of the cement block can be evaluated from the relation $F(t) = -SD(\partial C/\partial r)$ at $r = R$ as

$$F(t) = \frac{2C_0SD}{R} \sum_{n=1}^{\infty} \exp[-(D\alpha_n^2 t/R^2)] \quad (6)$$

where S is the surface area of the cement block.

The cumulative radioactivity, A (Bq) in the leachant can be evaluated by integrating Eq. (7) with respect to t . Since $C_0 = A_0/V$, where A_0 is the initial radioactivity added to the cement block, and V is the volume of the cement block, the fraction of the activity released to the leachant can be obtained as

$$\frac{A(t)}{A_0} = \sum_{n=1}^{\infty} (4/\alpha_n^2) [1 - \exp(-D\alpha_n^2 t/R^2)] \quad (7)$$

4. Results and discussion

4.1. Mechanical characteristics

The incorporation of radioactive waste into cement is the most widely used technique for its practical, technological and economical advantages. Mechanical properties are one of the important factors to be taken into consideration in evaluating the final solidified products. It is worth mentioning that certain minimum values of mechanical properties are required for safe handling of the immobilized waste forms during transportation and final disposal [22].

4.1.1. Effect of zeolite additive percent

The results of the mechanical strength of the hardened cement pastes and cement pastes mixed with different weight percentages of synthetic zeolite A are given in Table 4. Results indicated that addition of up to 4 wt.% zeolite A leads to increase in the value of the mechanical strength from 56.2 MPa in plain samples to 72.7 MPa and beyond this percent (4 wt.% zeolite A), the mechanical strength is decreased. On the other hand, the initial and final setting times of the plain cement paste decrease with the addition of 4 wt.% zeolite A from 4.0 to 3.5 and from 2.3 to 1.5 h, respectively. This increase in mechanical strength and lowering in initial and final setting time values may be attributed to the increase in silicates present in OPC due to the addition of zeolite A that in consequence increase the rate of hydration and strength the final solid product. Adding more than 4 wt.% zeolite A to the cement paste decreases the mechanical strength as reported in Table 4. This result could be attributed to the

Table 4
Effect of zeolite additive percent on mechanical strength

Addition percent	Mechanical strength (MPa)
Plain cement	56.18
2 wt. %	60.1
4 wt. %	72.7
6 wt. %	64.4
8 wt. %	47.8
10 wt. %	39.5

Table 5

Compressive strength of plain OPC and OPC mixed with 4% zeolite A after immersion in three types of leachants

Time	Composition	Mechanical strength (MPa)		
		Ground water	Tap water	Distilled water
30 days	OPC	97.7	81.0	74.8
	OPC + 4% zeolite A	108.0	91.4	87.3
90 days	OPC	133.0	108.0	95.6
	OPC + 4% zeolite A	141.3	120.5	112.2

increase in the alkali metal concentrations in the prepared mixture at the expense of aluminosilicates, which could affect the whole integrity of the pastes.

4.1.2. Effect of water immersion type

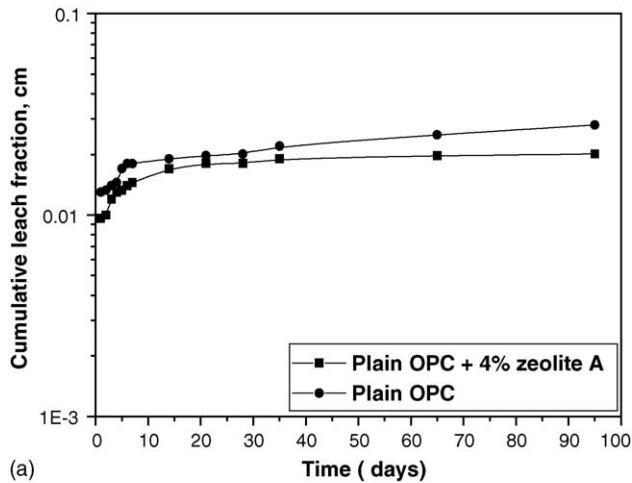
The effect of water immersion type on the compressive strength values of plain OPC and that mixed with 4 wt.% synthetic zeolite A was also examined and the obtained results are given in Table 5. Samples were immersed in ground water, tap water and distilled water. The data revealed that OPC mixed with 4 wt.% synthetic zeolite A has higher compressive strength value than plain OPC. This could be attributed to that zeolite A contains silica and alumina which react with calcium hydroxide during the hydration of cement to form calcium silicate hydrate. Compressive strength was also increased with increasing the time of immersion from 1 to 3 months. Samples immersed in ground water have the highest compressive strength which may be attributed to the presence of sulphate ions that activate the hydration of granulated slag. As a result of this activation, calcium sulphoaluminate and calcium silicate hydrate are formed filling up the available pore volumes and consequently the compressive strength increased [13].

4.2. Long term static leaching test

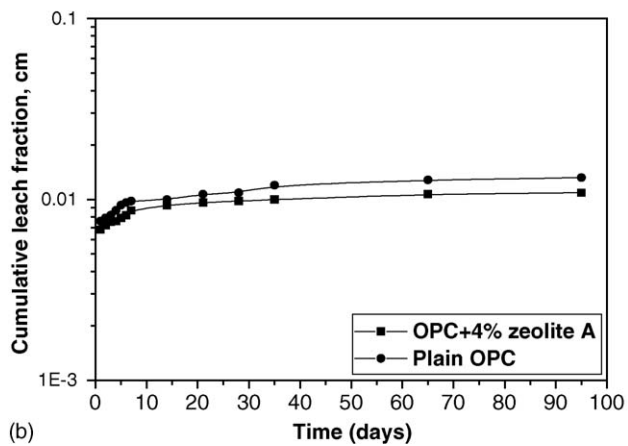
4.2.1. Leaching characteristic of ^{137}Cs and ^{90}Sr radionuclides

The shallow land burial waste forms contain fairly high concentrations of short-lived radionuclides and very low concentrations of long-lived radionuclides. Various radionuclides and salts make chemical bondage with cement components, or they exist dispersively in the state of sole crystals in concrete. When the cemented waste forms come in contact with water, the transfer of soluble materials from the waste to the surrounding water is caused by dissolution or chemical reaction with chemical components of water. In this study, the radionuclides chosen for the leach test are intended to represent the desorption (leaching) behavior of some of the typical radionuclides encountered in low-level solid waste forms.

The variations of the cumulative leach fractions of ^{137}Cs and ^{90}Sr radionuclides incorporated in cement and cement mixed with 4 wt.% zeolite A are depicted in Fig. 1. The results showed that, addition of 4 wt.% zeolite A decreased the leaching rates of both cesium and strontium radionuclides from cement matrix and this may be attributed to the extreme finesse and high surface



(a)



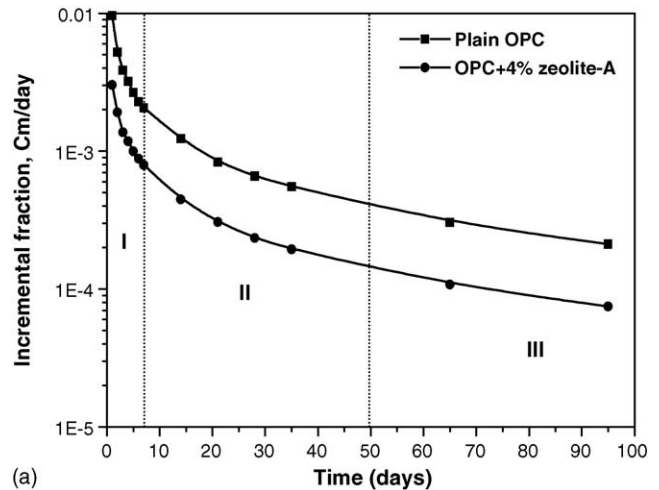
(b)

Fig. 1. Cumulative leach fractions of (a) ^{137}Cs and (b) ^{90}Sr radionuclides from plain cement and cement mixed with 4% zeolite A.

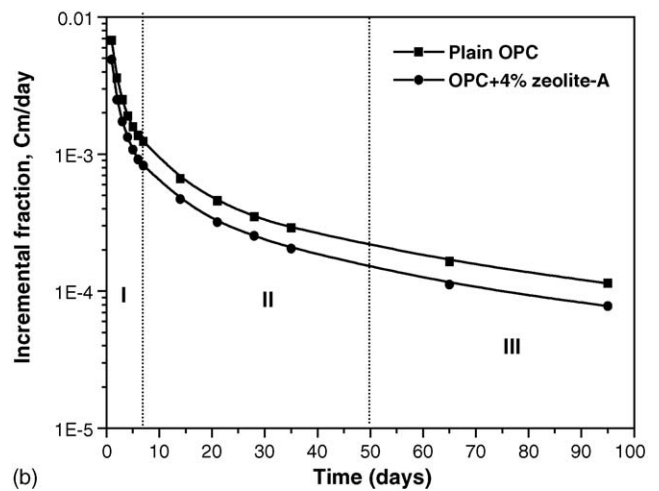
area of zeolite A which are capable to reduce the volume of large pores and capillaries founded in cement pastes. The magnitude of the cumulative leach fraction is an index for the release of the nuclide from the cement matrix. In both studied cases, cesium has highest values of cumulative leach fraction than strontium radionuclide.

4.2.2. Incremental fraction

Data of incremental fractions for plain OPC and OPC mixed with 4 wt.% zeolite A loaded with caesium and strontium radionuclides, expressed as cm/day on log scale against leach time in days, are graphically depicted in Fig. 2. The entire leaching period can be broadly divided into three regions. Region I from 1 to 7 days, region II from 8 to 50 days and region III from 51 to 90 days. Region I shows the initial leaching within the first 7 days, i.e. leaching from superficial surfaces of the block. Region II shows that after initial leaching within first 7 days, there is a drastic reduction in release of both radionuclides from the blocks which is maintained over a longer period of time (50 days). In region III, the leach rates of both radionuclides are further lowered and this trend continues up to 90 days. The leach behavior of the blocks can be explained as a combination of the



(a)



(b)

Fig. 2. Variation of incremental fraction of (a) ^{137}Cs and (b) ^{90}Sr radionuclides from solidified cement pastes as a function of time.

two processes: (i) surface wash-off mechanism and (ii) diffusion stage mechanism. Region I shows the maximum leaching of the ions because of the surface wash-off process. In this process a rapid equilibrium is established between ionic species present in the surface pores of the OPC block and ions in the leachant solution. When each ion has been leached out from the surface of the block, it migrates by longer pathways from the bulk through diffusion-controlled stage, which determines the long term leaching behavior of the block [23].

4.2.3. Effective diffusion coefficient

Leaching is assumed to be diffusion-controlled process. The mechanism of this diffusion could not be completely studied due to complex microstructure of composite and the presence of multi variables which affect the rate of leaching such as matrix composition, temperature, chemical nature of leaching solution and chemical nature of the element diffused out. Several methods are used to measure leaching data and IAEA suggested that diffusion coefficients may be used to compare leaching data [24], where the quantity of radionuclide leached out from a unit

surface area during time, t_n is given by

$$A_n = 2A_0 \left(\frac{Dt_n}{\pi} \right)^{1/2} \tag{8}$$

where A_n is the activity leached out after time t_n , Bq , A_0 the initial activity in the composite, Bq , and D is the diffusion coefficient, cm^2/s .

From the above equation, the cumulative fraction leached out from the composite can be expressed as

$$\left[\frac{\Sigma A_n}{A_0} \right] \left[\frac{S}{V} \right] = \frac{2SA_0(D\Sigma t_n)^{1/2}}{A_0V} \tag{9}$$

$$\frac{\Sigma A_n}{A_0} = 2 \left(\frac{S}{V} \right) \left(\frac{D\Sigma t_n}{\pi} \right)^{1/2} \tag{10}$$

where ΣA_n is the cumulative amount of radioactivity leached during cumulative time t_n .

Thus a plot of $[\Sigma A_n/A_0]$ versus $(t_n)^{1/2}$ should give a straight line if D remains constant. The value of D can be calculated from the slope m of the line, i.e.

$$D = \frac{\pi m^2 V^2}{4S^2} \tag{11}$$

Fig. 3 represent the plotting of the fraction leached of cesium and strontium radionuclides from both studied samples versus square root of leaching time. As can be seen from these plots, for all studied leaching tests, the results indicated an initial fast leaching during the first period followed by slow leaching in the subsequent periods. This behavior suggests the presence of two different values of diffusion coefficient for the fast and slow components. So, the calculated diffusion coefficients for both nuclides, as presented in Table 6, are the average values of their diffusion coefficients. These data indicated that ^{137}Cs have largest values of diffusion coefficients in both waste matrices compared to ^{90}Sr nuclide.

The American National Standards Institute (ANSI) defines a material parameter of the leachability of diffusing species. This parameter is called the leachability index, L , and is defined as [25]

$$L = \log \left(\frac{\beta}{D} \right) \tag{12}$$

where β is a defined constant ($1.0\text{ cm}^2/s$) and D is the diffusion coefficient of the species (cm^2/s) assumed to be independent of time during the course of leaching.

The corresponding values of the leachability index (L) for all studied samples calculated using Eq. (12), as listed in Table 6,

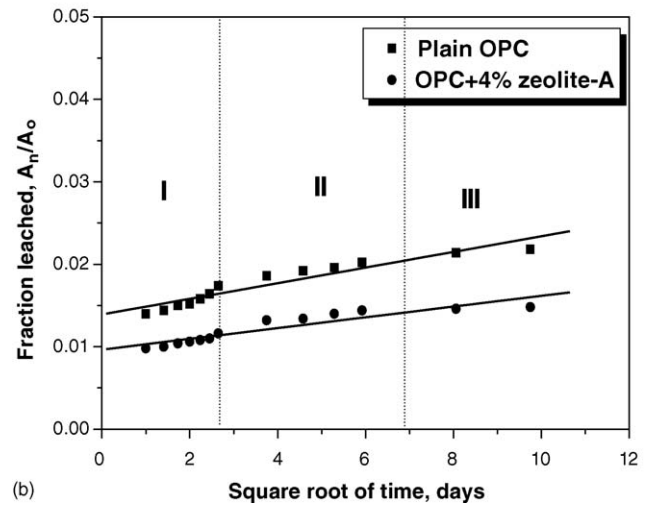
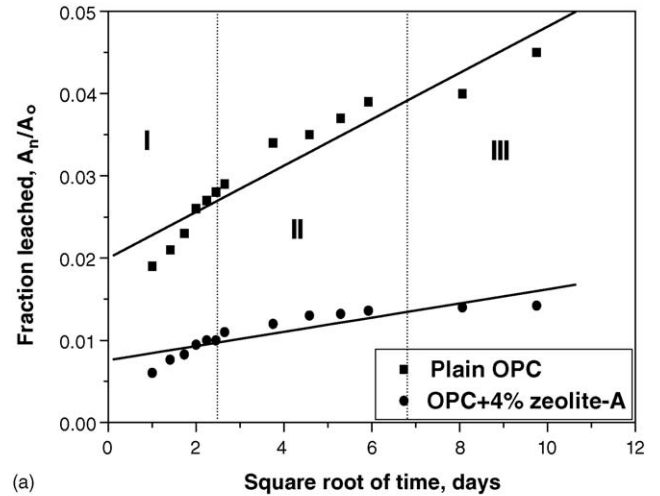


Fig. 3. Variation of fraction leached of (a) ^{137}Cs and (b) ^{90}Sr radionuclides from cement and cement mixed with 4% zeolite vs. square root of time.

exceeded the value of 6, which is the minimum value for acceptance as a low level waste form [25].

4.3. Prediction of time-profile leach fraction

Temporal profiles of leach fractions of ^{137}Cs and ^{90}Sr from OPC waste form measured under laboratory conditions (as previously mentioned in Section 4.2.1) are shown in Fig. 4. The predicted curves by the simulated model (Eq. (7)) using the evaluated diffusion coefficients are also presented in this figure. The simulation gives an acceptable representation of experimental data; thereby demonstrating that the proposed mechanism in the model can represent the leaching kinetics of studied radionu-

Table 6
Diffusion coefficient and leachability index values of cesium and strontium radionuclides leached from solidified cement matrices

Cement matrix	Diffusion coefficient (cm^2/s)		Leachability Index, L	
	^{137}Cs ($\times 10^{-8}$)	^{90}Sr ($\times 10^{-8}$)	^{137}Cs	^{90}Sr
Plain OPC	46.00	5.40	6.34	7.27
OPC + 4% zeolite A	5.70	3.75	7.25	7.42

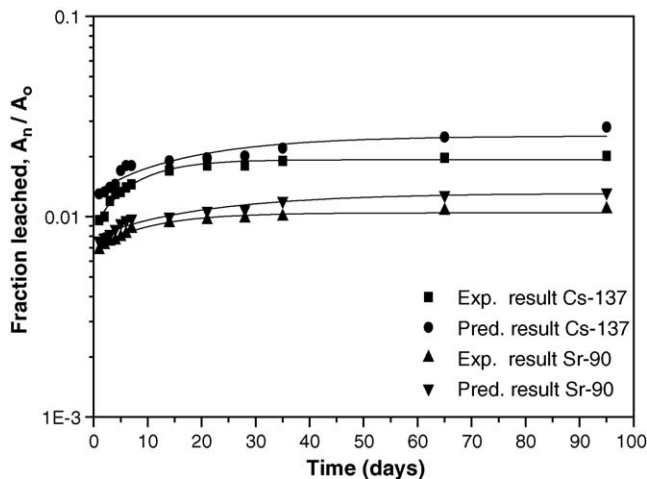


Fig. 4. Experimental and predicted values of the fraction leached of ^{137}Cs and ^{90}Sr radionuclides from Portland cement matrix vs. leaching time.

clides from cylindrical geometry cement matrix. The observed difference in data can be related to the discrepancies in calculating the values of diffusion coefficients. Further work, is under investigation to rectify this model.

5. Conclusion

This paper presented results of immobilization of cesium and strontium radionuclides loaded zeolite A ion exchange material in a mixture with OPC. The results showed that addition of up to 4 wt.% contaminated zeolite A leads to increase the compressive strength of the final solid matrix form 56.2 MPa in plain samples to 72.7 MPa and beyond this percent the compressive strength is decreased. Also, the leaching rates for both studied radionuclides from cement matrices were decreased with the addition of zeolite A and this may be attributed to the extreme fineness and high surface area of zeolite A that capable of reducing large pores and capillaries founded in cement pastes. The leachability index measured for each of the samples was in the range of 7.42 to 6.34 and exceeded the required value of 6, the minimum value for acceptance as a low level waste form. The corresponding diffusion coefficient was in the range of 3.75×10^{-8} to $46 \times 10^{-8} \text{ cm}^2/\text{s}$, this range is low that measurements could only be accomplished by leaching very thin sections of the waste form for long times.

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