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Assessment of the leaching characteristics of incineration ashes in cement matrix

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ABSTRACT

In this work, the effort is directed to assess the feasibility of immobilizing the ash produced from the incineration of solid radioactive wastes. Within this context, the ash was characterized to determine its chemical composition and physical properties. Immobilized cement-ash matrices have been prepared to investigate the influence of waste to cement ratio. To characterize the extent of the solidification process of the immobilized waste matrices, the mechanical strength test was conducted. The standard mass transfer leach test has been employed to test the extent of ^{137}Cs and ^{60}Co stabilization. Non-linear fitting of the experimental leach data to different mathematical models was conducted to evaluate the mechanisms those instigate the leaching phenomena and the leaching parameters were determined. The controlling leaching mechanism and leachability indices were calculated for the studied waste matrices. The results indicated that ^{137}Cs leaching is resulted from first-order reaction between the surface of the waste matrix and the leaching solution followed by diffusion through the studied matrices. The leaching of ^{60}Co was found to be as result of four subsequent mechanisms that include release of loosely bound ^{60}Co followed by first-order reaction the diffusion and finally dissolution. It was found that the studied immobilized waste matrices have acceptable mechanical performance. The values of the leachability indices indicate that the performance of the studied matrices in ^{137}Cs stabilization is not acceptable.

1. Introduction

Combustible solid radioactive wastes are generated during the operation of nuclear power plants, research centers and laboratories, hospitals, universities, radioisotope production facilities, reprocessing plants, and fuel fabrication plants or laboratories [1,2]. The composition of these wastes depends largely on the type of application that produces these wastes. Combustible solid radioactive wastes include; plastic objects, rubber, papers, activated charcoal, biological materials, wood, cotton and other cellulosic fabrics and ion exchange resins in the form of slurries, contained in filter cartridges [3]. The safe management of these wastes includes sorting, volume reduction, conditioning, transport and disposal.

Incineration is a proven volume reduction method that has been generally applied to combustible solid radioactive waste. It involves oxidation of burnable components of the waste, the end product of this process are inorganic ash residues and secondary wastes in the form of vapors, and gases. There are diverse available combustion technological options; Table 1 presents process comparison between different combustion technologies. The selection of the appropriate technology is bounded by administrative and technical factors. Administrative factors include; basic radioactive waste management principals, regulatory requirements, available resources, and compatibility with other elements in the waste management system [4–6]. The technical factors include:

(a) Capability to process a variety of non-homogeneous wastes of different chemical compositions, physical dimensions, densities, moisture contents and heat values;
(b) Displaying low sensitivity to incompatible items with the process-system design;
(c) Complete oxidation of the waste feed, including the combustible products of the waste thermal decomposition, within the boundary of the combustion part of the system;
(d) Consistent process parameters and consistent off-gas composition of the off-gas at the exit from the combustion part of the system; and
(e) Consistent quality of ash having desirable physical characteristics from the viewpoint of its ease of removal, transfer and immobilization.

Inorganic residues, containing radionuclides, are highly dispersive powders and their specific toxicity is higher than that of the primary wastes. It is recommended that these residues should be immobilized in an appropriate matrix to reduce the risk of radiation exposure to workers and public, produce a final structurally stable waste form, and to limit the environmental contamination

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Table 1
Comparison between different combustion technologies.

Combustion techniques	Temperature (°C)	Process	Output
Excess air incineration	800–1100	Allow an excess of oxygen during the primary combustion process so that both the gaseous and solid fractions can burn directly in one combustion chamber simplest combustion technique.	The excess air and the controlled air incineration leave a similar end product but contain less carbon.
Controlled air incineration (primary process)	800–1000	Limit the air supply in the primary combustion step to near or below the stoichiometric ratio a secondary combustion step is needed for the completion of the combustion of the gaseous fraction in an oxygen rich atmosphere.	
Pyrolysis (primary process)	500–600	A reducing atmosphere, usually maintained by restricting the air supply to much less than stoichiometric levels pyrolysis of organic materials causes their thermal degradation and a distillation of the volatile fraction, forming combustible liquids and vapors. Pyrolysis is an endothermic process and a continuous source of heat is required to maintain it. The pyrolysing incinerators employ secondary chambers where the ash and the gaseous products of pyrolysis are fully oxidized in an oxygen rich atmosphere.	The material remaining after pyrolysis is char, a charcoal-like substance consisting primarily of fixed carbon residue yields char containing some fixed carbon.
High temperature slagging incineration	1400–1600	The incinerators employed to produce the slag use relatively high process temperatures to burn the carbonaceous residue by receiving heat, typically from burning fuel, thus releasing an amount of heat energy sufficient to convert all non combustibles contained in the waste feed to molten slag.	Produces a glass-like aggregate containing very little or no fixed carbon, the main constituent being SiO_2.
Fluidized bed incineration	800	The incinerator vessel contains an inert bed of particles that are kept in suspension by fluidizing air flowing through the bed at a rate that is just rapid enough to sustain that condition The shredded waste ignites instantly upon introduction into the fluidized bed incinerator. The combustion of both the solid and gaseous fractions of the waste is accomplished in one chamber and the fly ash resulting from the process leaves the chamber with the off-gas.	Produce solid residues in the form of fly ash.

Table 2
Chemical composition of cement and ashes.

Chemical composition	OPC (wt.%)	Incineration ashes (wt.%)
CaO	63	24.6
SiO_2	20	30.0
Al_2O_3	6.0	5.4
Fe_2O_3	2.1	4.6
MgO	1.5	3.3
Na_2O	0.5	1.5
P_2O_5	NA	10.7
K_2O	NA	8.4
Others	6.9	11.5

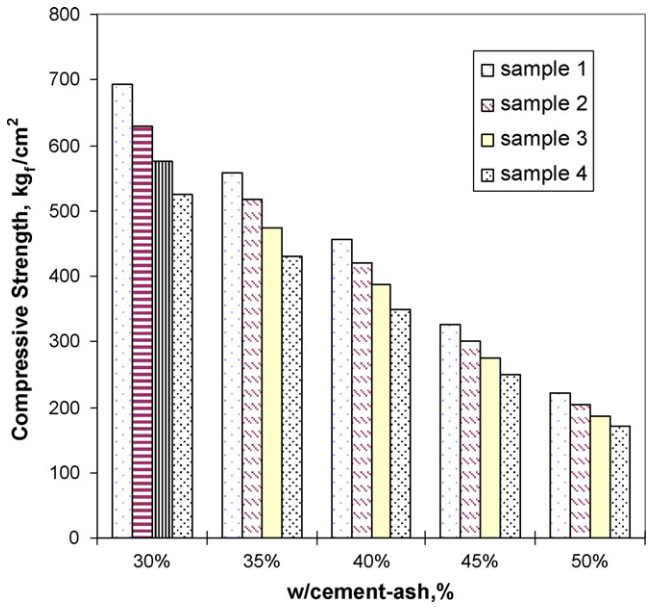

Fig. 1. Compressive strength of the studied samples as a function of water to cement-ashes content.

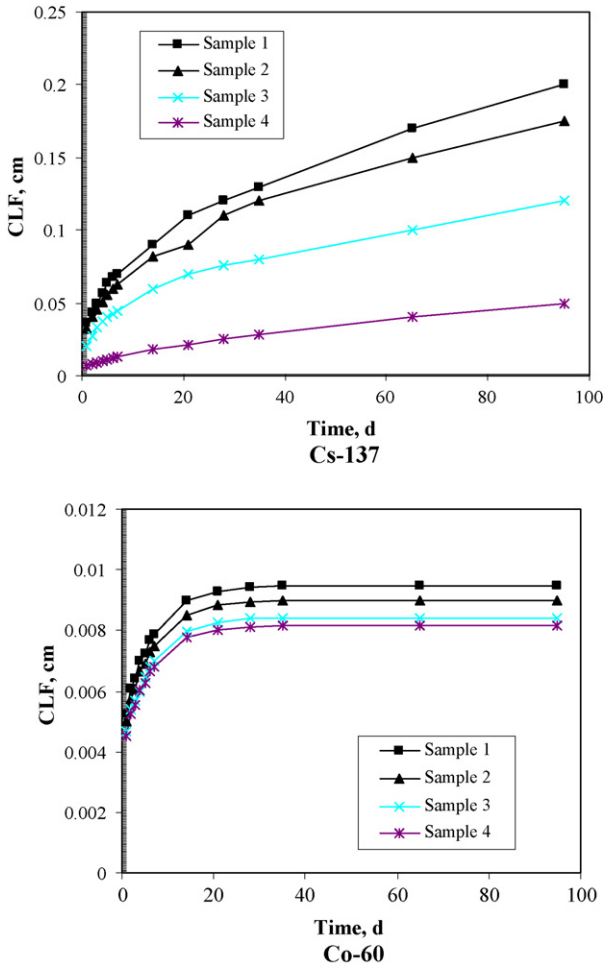

Fig. 2. CLF of the Cs and Co radionuclides from immobilized waste matrices.

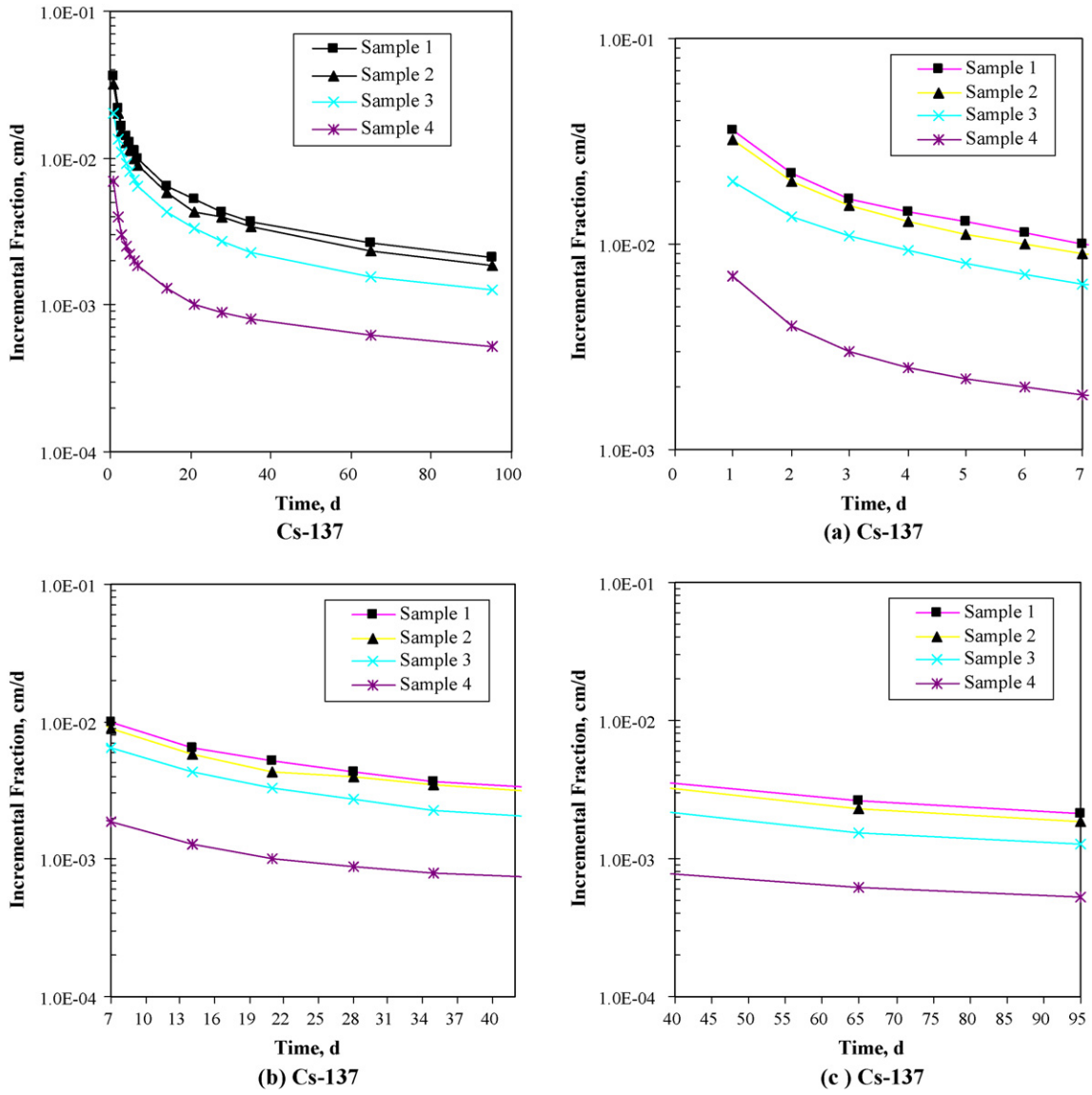

Fig. 3. Incremental leach fraction of Cs radionuclides as a function of time.

[7]. During immobilization two simultaneous processes occurred namely; stabilization and solidification. Inorganic residues are stabilized by converting the radionuclides to less mobile form as a result of chemical change, where solidifying these wastes aims to improve their mechanical performance [8].

A variety of immobilization techniques are available; these include cementation, bituminization and vitrification. Cementation of radioactive waste has been practiced for many years basically for immobilization of low and intermediate level radioactive waste [9]. The majority of cementation techniques rely on using Ordinary Portland Cement (OPC) as the primary binder. Other binders might be used to improve either the mechanical performance of the final waste matrix or to improve the retention of radionuclides in that matrix, these include fly ash, blast furnace slag, bentonite, zeolite and other materials [10–16].

To characterize the extent of stabilization and solidification processes, series of tests are conducted. Leaching tests are considered as the primary method to characterize the extent of stabilization, these tests could be classified as tests designed to simulate the release under specific environmental scenario, sequential chemical extraction tests and mass transfer tests that aims to evaluate the fundamental leaching parameters and to estimate the release rate values [17]. To evaluate the extent of the solidification process of the immobilized waste matrices, different tests are carried out to measure the unconfined compressive strength, wet–dry and freeze–thaw tests to evaluate the durability, and strength tests to estimate the long-term stability of the waste form.

In this work the feasibility of immobilizing the incineration ashes in cementatious matrix is investigated. Within this context, immobilized waste matrices were prepared with different ash to OPC ratios, and standard mass transfer leach test has been employed to study the leaching pattern of ^{137}Cs and ^{60}Co radionuclides. The leaching data were analyzed to evaluate the controlling leaching parameters and mechanisms. To provide a baseline comparison between the prepared matrices, from mechanical view point, the mechanical strength test was carried out to estimate the stability of the prepared matrices.

2. Evaluation of the leaching parameters

Leaching tests are used to measure the cumulative leach fraction (CLF) that represents the leaching rate of some radionuclides of potential concern from immobilized waste matrix under continuously saturated conditions that simulates the disposal conditions

under the most conservative scenario. The results of these tests are used to evaluate the leaching parameters by fitting the experimental data to mathematical models that represent different leaching mechanisms. In this section, some of these mathematical models will be presented. It should be noted that the equations used are valid only if the times of experiments are much shorter compared the half-life of radionuclides, otherwise more complex formulae should be used [18].

2.1. First-order reaction model (FRM)

This model is used to estimate the leaching parameters if the radionuclide leaching is controlled by the exchange kinetics between the surface of the waste matrix and leaching solution. The surface exchange rate could be governed by first-order reaction rate, so it will be proportioned to the amount of radionuclides in the waste matrix as follows [19]:

$$\frac{dQ}{dt} = kQ \tag{1}$$

where Q represents the amount of soluble radionuclides in the waste ($mg\,g^{-1}$) and k is a rate constant (s^{-1}). The CLF is given by:

$$CLF = Q_0(1 - \exp(kt)) \tag{2}$$

where Q_0 is the initial amount of soluble radionuclide in the matrix ($t = 0$) ($mg\,g^{-1}$). By fitting the experimental CLF to the model Eq. (2), Q_0 and k could be determined.

2.2. Constant first-order reaction model (CON FRM)

This model assumes the occurrence of two processes, in the first the loosely bound material would be leached instantaneously and easily washed away and the second is the first-order reaction exchange between the waste matrix and the leaching solution.

2.3. Diffusion model (DM)

To assess the leaching parameters of immobilized radionuclides, if their transport is controlled by diffusion, the solution of Fick's second law in semi-infinite medium and Fick's first law could be

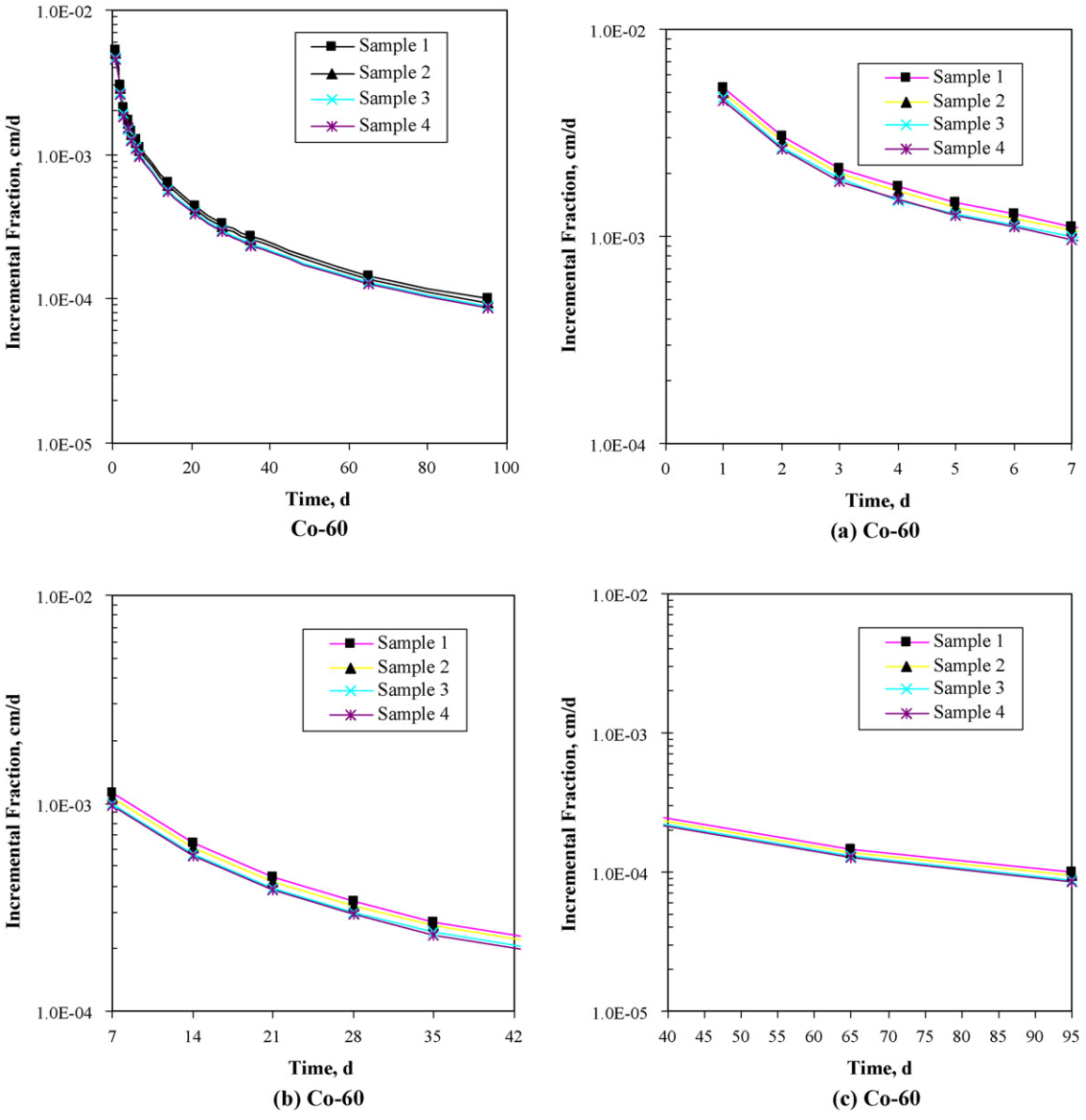

Fig. 4. Incremental leach fraction of Co radionuclides as a function of time.

used to obtain the flux of the diffusing materials $J(t)$ through the immobilized matrix as follows:

$$J(t) = -D\left.\frac{\partial C}{\partial x}\right|_{x=0} = -C_0\sqrt{\frac{D}{\pi t}} \tag{3}$$

where C_0 is the initial concentration in the waste matrix (Bq/m^3) and D is the diffusion coefficient (cm^2/s).

The leached activity from a unit surface area during time A_n (t) is expressed by:

$$A_n(t) = \int_0^t J(t)\,dt = 2A_0\sqrt{\frac{Dt}{\pi}} \tag{4}$$

From the above equation, the CLF out from the waste matrix can be expressed as:

$$\frac{\sum A_n}{A_0} = 2\left(\frac{S}{V}\right)\sqrt{\frac{Dt}{\pi}} \tag{5}$$

where $\sum A_n$ is the cumulative amount of radioactivity leached during cumulative time t_n.

The value of the apparent diffusion coefficient (D) can be calculated from the slope (m) straight line of the plot of $[\sum A_n/A_0]$ versus $(t_n)^{1/2}$, i.e.

$$D = \pi\left(\frac{mV}{2S}\right)^2 \tag{6}$$

2.4. First-order reaction/diffusion model (FRDM)

The leaching rates from immobilized waste matrix might be due a combination of the exchange kinetics between the surface of the waste matrix and leaching solution and bulk diffusion of radionuclides through the waste matrix. Therefore, the CLF could be obtained by combining Eqs. (2) and (5) yields:

$$\mathrm{CLF} = Q_0(1 - \exp(kt)) + 2\left(\frac{S}{V}\right)\sqrt{\frac{Dt}{\pi}} \tag{7}$$

The initial amount of soluble radionuclides, the rate constant, and the apparent diffusion coefficient could be evaluated by performing non-linear fitting.

2.5. Dissolution model (DIM)

If the leaching species is structurally a major component of the waste form, its release into leaching solution lead to a structural

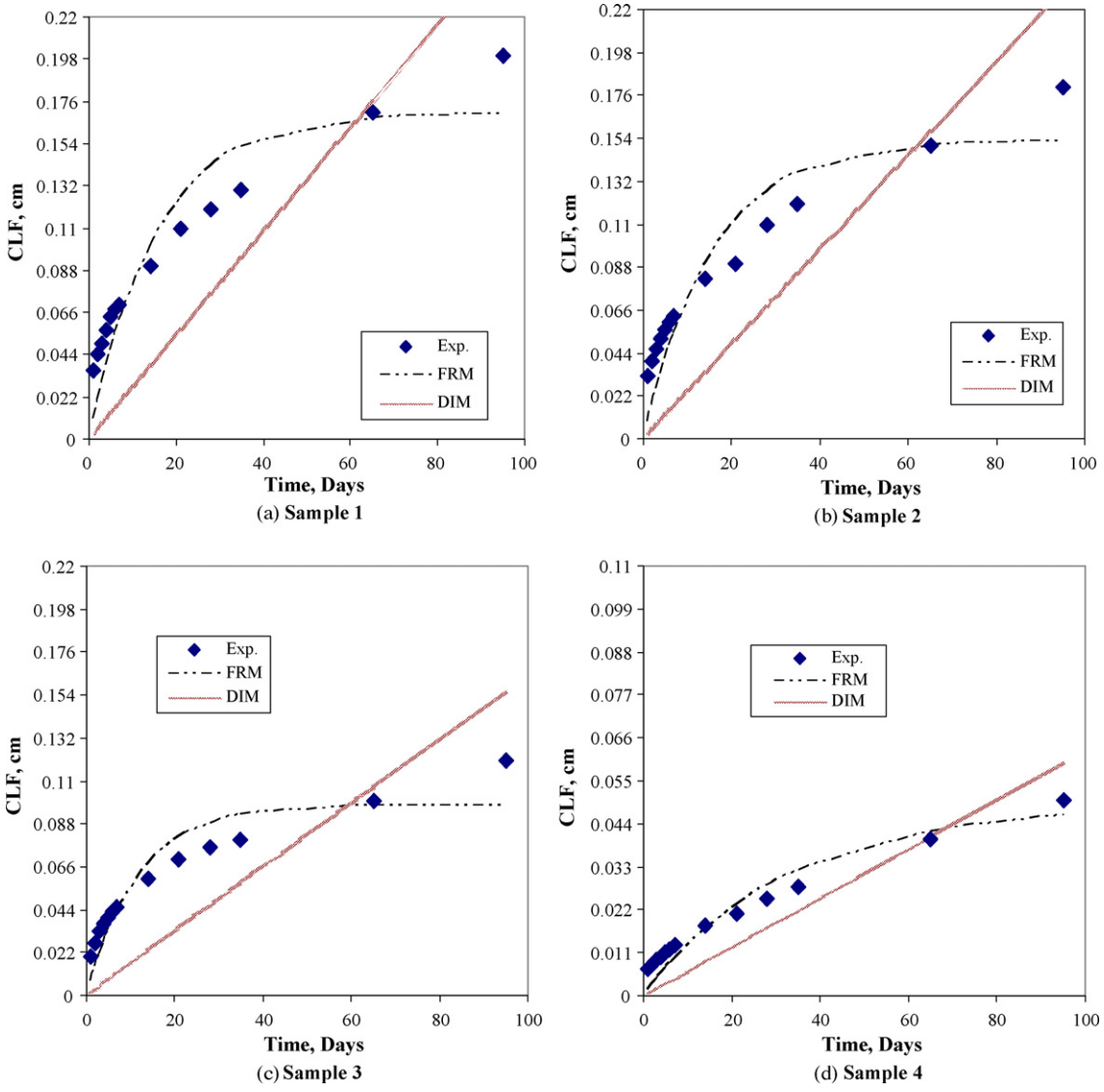

Fig. 5. (a–d) Non-linear fit of the Cs CLF to the FRM and DIM models; (e–h) non-linear fit of the Co CLF to the FRM and DIM models.

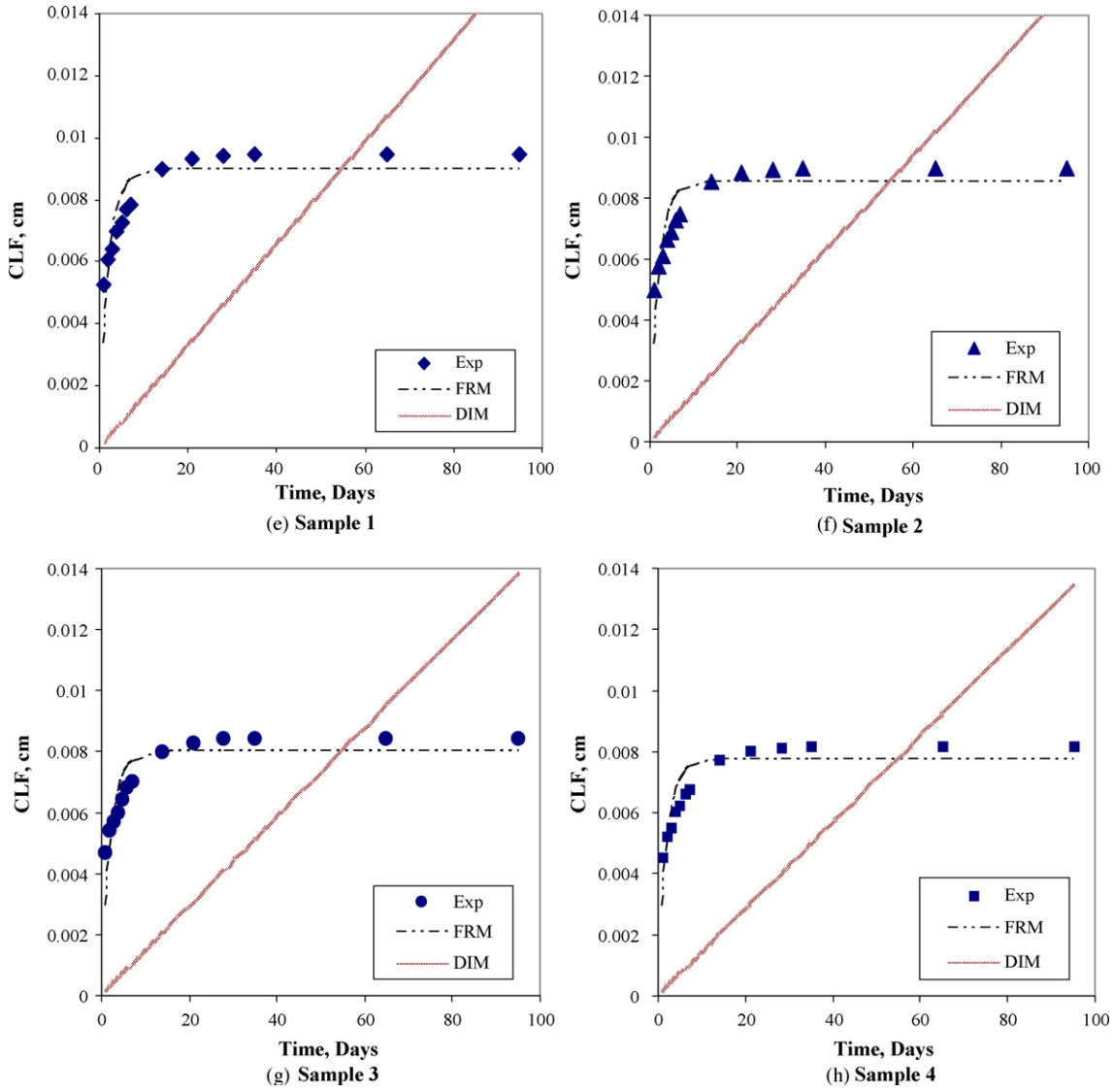

Fig. 5. (*Continued*).

breakdown of the matrix, a process which is referred to as dissolution. The kinetics of dissolution can be represented by a network dissolution velocity U, defined as the volume of solid material being dissolved per unit time and per unit surface area of solid exposed [20]:

$$U(t) = U_0 \left(1 - \frac{C^w(t)}{C^w_{\text{sat}}}\right) \tag{8}$$

where U_0 is the maximum network dissolution velocity and C^w_{sat} is the saturation concentration in the aqueous solution. For the simple case where $C^w_{\text{sat}} t \gg C_w(t)$, $U(t) = U_0$, and the cumulative fraction leached is obtained as:

$$\text{CLF} = \frac{S}{V} U_0 t \tag{9}$$

2.6. First-order reaction diffusion dissolution model (FRDDIM)

The leaching rates from immobilized waste matrix might be due superimposition of the FRM, DM, and DIM models, therefore the CLF could be obtained by:

$$\text{CLF} = Q_0(1 - \exp(kt)) + 2\left(\frac{S}{V}\right)\sqrt{\frac{Dt}{\pi}} + \left(\frac{S}{V}\right)Ut \tag{10}$$

Table 3
Correlation coefficients of different models for the leaching of ^{137}Cs.

Model	Sample 1	Sample 2	Sample 3	Sample 4
CONFRM	0.19	0.23	0.01	0.23
FRDM	0.98	0.98	0.98	0.99
FRDDIM	0.97	0.98	0.93	0.89
ALL	0.99	0.99	0.99	0.98

Table 4
Correlation coefficients of different models for the leaching of ^{60}Co.

Model	Sample 1	Sample 2	Sample 3	Sample 4
CONFRM	0.18	0.24	0.23	0.22
FRDM	0.48	0.48	0.47	0.48
FRDDIM	0.26	0.27	0.29	0.24
ALL	0.99	0.99	0.99	0.99

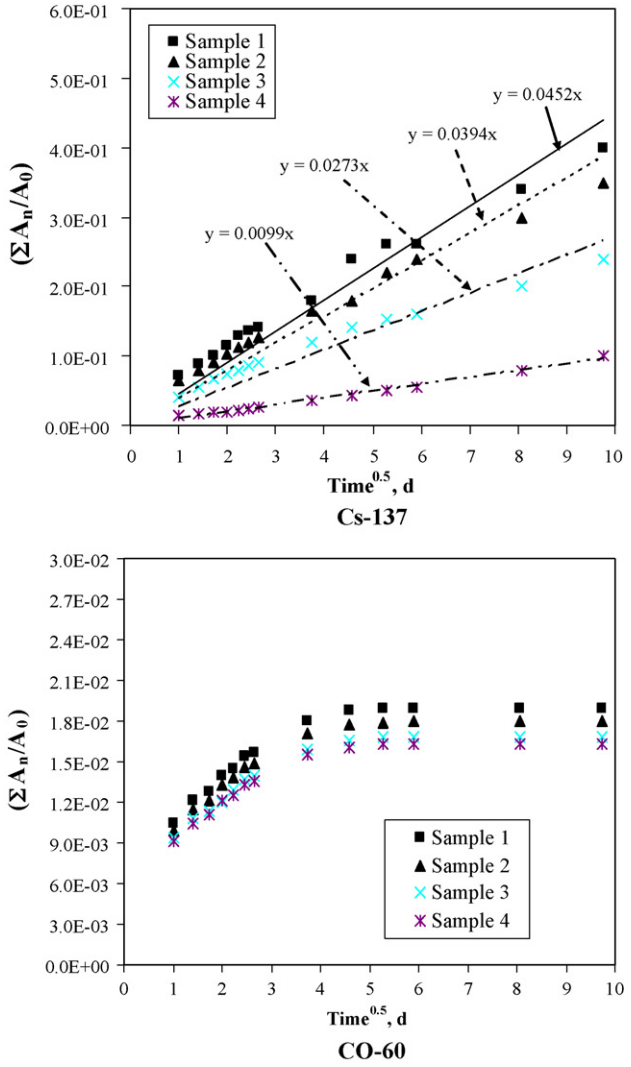

Fig. 6. Fitting the experimental data to diffusion model.

2.7. Collective model (ALL)

This model assumes that the leaching rate is occurred due to superimposition of CONFRM, DM, and DIM models.

3. Experimental

3.1. Materials

OPC was provided from Seuz Cement Company, Seuz, Egypt and its chemical composition is given in Table 2. The used incineration ashes were produced by open air incineration of dry solid wastes. The pH of water in contact with the ashes is 10.5 and the chemical composition of these ashes is also given in Table 2. Loss during calcinations at 900 °C was found to be 8.2 wt.%. BET surface area of the solid powder was measured after the thermal treatment for 2 h at 473 K, and was found to be 25.13 m^2/g.

3.2. Preparation of specimens

Cement pastes were prepared by mixing plain OPC with 6 wt.%, 12 wt.%, 20 wt.%, and 30 wt.% of ashes at different water to cement-ashes (w/c) ratio. The required amount of cement and ashes were 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 mixing using a helix grout mixer at a 1500 rpm speed for duration of 4 min. At the end of mixing, paste was directly poured into polyethylene cylindrical moulds with internal height/diameter ratio of 1.0 (2.0 cm diameter and height).

The 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 at 25 ± 2 °C.

3.3. Mechanical strength

For the determination of compressive strength, immobilized cement matrices containing the ashes were prepared. After curing for a period of 28 days, the compressive strength of the matrices was measured using a microprocessor based compression testing machine.

3.4. Static leaching test

Static leaching tests were preformed, using distilled water solution, to study the leaching of ^{137}Cs and ^{60}Co from hardened matrices of cement-ash matrices. The IAEA's standard test proposed by Hespe [21] was applied. Each cured specimen was immersed in beaker containing 300 ml distilled water, a sample withdrawn and analyzed using gamma spectrometer with 2 × 2 in. sodium iodide (NaI) crystal activated with thallium. The crystal was connected to a multi-channel analyzer which had 256 channels attached with preamplifier. The equipment was manufactured by the Nuclear Excellence in Nuclear Instrumentation, Model 800A. The CLF (cm) was calculated according to the following equation:

$$\mathrm{CLF} = \left(\frac{\Sigma A(t)}{A_0}\right)\left(\frac{V}{S}\right)$$

where $\sum A(t)$ 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).

4. Results and discussions

4.1. Mechanical strength

The results of the mechanical strength test as a function of the water to cement-ashes ratio for the prepared waste matrices after 28 days are illustrated in Fig. 1. From this figure, it is clearly shown that the compressive strength shows a decreasing behavior with increasing the water to cement-ash ratio. Also, the results indicate that increasing the ratio of the incineration ashes decreases the mechanical strength of the sample. The reason of the decrease in the compressive strength with increasing water could be attributed to existence of variable pore structure and visible holes in too wet mixtures [22]. Other reported studies in the literature indicate that when OPC is blended with fly ash, the increases in the fly ash to OPC ratio will lead to increase in the water demand of the mix and a decrease in the strength of the sample. They attributed the lower strength to the lower OPC ratio and much higher water contents [23–25]. The lowest measured compressive strength values were

Table 5
Results of the non-linear fitting of the experimental Cs leaching data.

Model	Sample 1	Sample 2	Sample 3	Sample 4
FRDM				
$Q\,(mg\,g^{-1})$	0.22	0.28	0.11	7.0E−3
$K\,(s^{-1})$	2.9E−3	2.1E−3	4.2E−3	5.3E−6
D	1.5E−4	1.2E−4	6.0E−5	4.8E−6
FRDDIM				
$Q\,(mg\,g^{-1})$	3.5E−3	3.5E−3	3.5E−3	3.5E−3
$K\,(s^{-1})$	0.03	0.03	0.03	0.03
D	1.3E−4	1.3E−4	5E−4	9.3E−7
U	−6E−5	−3.1E−4	2E−4	3.8E−4
ALL				
$Q\,(mg\,g^{-1})$	1.83	0.18	1.1E−3	1.6E−7
$K\,(s^{-1})$	5.9E−7	−5.7E−8	−1E−5	−2.648E−9
D	9E−5	7E−5	4E−5	7.2E−6
U	−1.1E−4	−1.1E−4	−1.8E−4	−5E−4
CON	0.016	0.014	0.008	−0.002

found to be higher than the acceptable regulation requirement for compressive strength (35 kg/cm^2). For the highest water to cement-ash samples the compressive strength was found to be in the range 221–170 kg/cm^2.

Table 6
Results of the non-linear fitting of the Co experimental data to ALL.

Model	Sample 1	Sample 2	Sample 3	Sample 4
$Q\,(mg\,g^{-1})$	2E−3	9E−3	8.9E−3	0.11
$K\,(s^{-1})$	0.002	2E−4	8E−4	1E−3
D	5.62E−7	4.87E−7	4.37E−7	4.02E−7
U	6E−5	6E−5	6E−5	5E−5
CON	3.9E−3	3.8E−3	3.5E−3	3.5E−3

4.2. Leaching charachterisitcs of ^{137}Cs and ^{60}Co radionuclides

The IAEA's standard leach test is designed to measure the cumulative leach fraction (CLF) from a monolithic immobilized waste matrix. The result of this test reflects the physical changes and chemical interactions that occurred within the tested matrix. The influence of increasing the ashes loading on the CLF of ^{137}Cs and ^{60}Co from the studied immobilized waste matrices are depicted in Fig. 2. The examination of this figure indicates that:

(a) The CLF is less than 20% for all the studied cases.
(b) Increasing the ash loading decrease the CLF for the studied radionuclides, and this can be attributed to the high value of

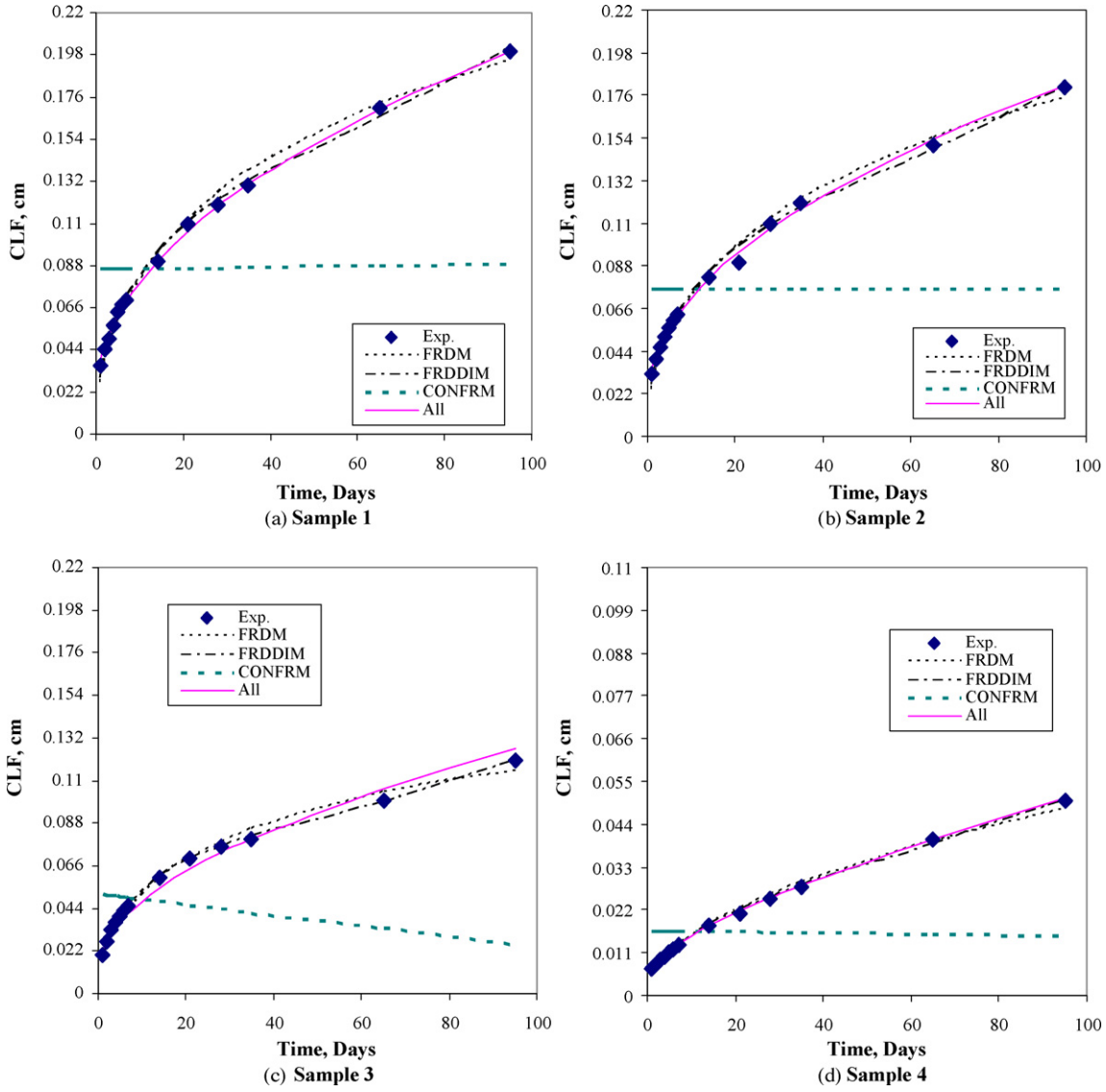

Fig. 7. (a–d) Fitting the Cs experimental data to the combination of different mechanisms; (e–h) fitting the Co experimental data to the combination of different mechanisms.

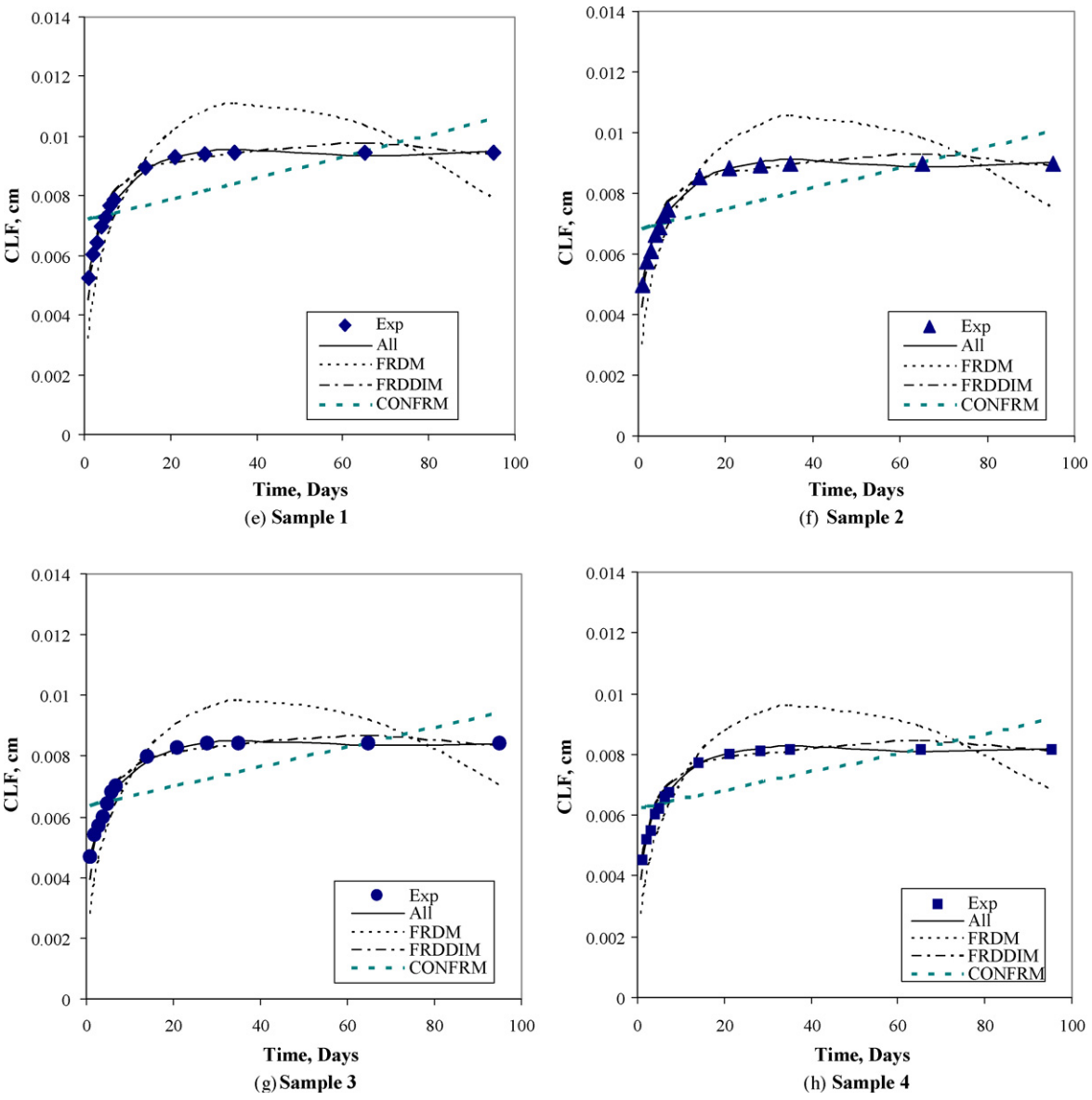

Fig. 7. (*Continued*).

surface area of the ashes that can increase the available site for sorption when compared with that of lower ash ratio matrix.

(c) ^{60}Co has smaller leachability than ^{137}Cs, this could be attributed to the low field strength of ^{137}Cs that keeps it substantially soluble in the high pH environment [26].

4.3. Leaching parameters and mechanism

The examination of the plots of incremental leach fractions form the studied immobilized waste matrices loaded by ^{137}Cs and ^{60}Co expressed as cm/day on log scale versus time (Figs. 3 and 4), indicate that the leaching pattern can be divided into three regions. Region I (Figs. 3a and 4a), shows initial rapid release of radionuclides within the first 7 days, then a drastic reduction in the release take place over a longer period of time (40 days). In region III, the leach rate of the radionuclides is further lowered and this trend continues up to 90 days (Figs. 3c and 4c). These results are consistence with some reported studies in the literature that found that leaching of cementatious matrices divided into three regions [15,26–28].

To evaluate the mechanisms those instigate the leaching phenomena of the studied waste matrices, the experimental CLF data were fitted to the non-linear form of first-order reaction model (FRM) and dissolution model (DIM) (Eqs. (2) and (9)) as illustrated in Fig. 5 and linearly fitted to the diffusion model (DM) (Eq. (6)) as in Fig. 6. From the visual examination of Fig. 5, it could be concluded that for all the studied matrices the dissolution model cannot represent the experimental data adequately either at short or long times. The pattern of the experimental data for ^{137}Cs leaching from all the studied matrices is different from that of the FRM, where ^{60}Co pattern could be represented with this model although it underestimate the release of ^{60}Co from the studied matrices. The examination of Fig. 6 indicates that ^{137}Cs leaching could be represented by the diffusion model where ^{60}Co leaching cannot be presented by the diffusion model. From the above mentioned results, it could be concluded that the leaching phenomena of both ^{137}Cs and ^{60}Co cannot be adequately represented by a single mechanism but it could be resulted from a combination of different mechanisms. These mechanisms could be estimated by further fitting the experimental data of ^{137}Cs and ^{60}Co release to the combination of different models, namely (CONFRM, FRDM, FRDDIM, ALL) as presented in Fig. 7. Form the visual examination of this figure, it is clearly shown that for all the studied waste

matrices the CONFRM model cannot represent the experimental data well where the rest of the models fit the data well. To assure the convergence of the fitted data the Chi^2 values, which represent the difference between two subsequent stages of the fitting, were set to be smaller than 10^{-5}. The correlation coefficients, which are the statistical measure of how well the experimental data points fit the regression curve, are shown in Tables 3 and 4. Fitting the experimental data to CONFRM model resulting in a very low correlation coefficients which confirm that the leaching of both radionuclides from the studied matrices are not resulted from the release of loosely bound radionuclides followed by first-order reaction exchange between the surface of waste matrix and the leaching solution. From the examination of the values of the correlation coefficients for ^{137}Cs leaching it was found that the data could be represented by FRDM, FRDDIM, and ALL where ^{60}Co leaching could be only well represented by ALL. The leaching parameters obtained from the non-linear fitting of the experimental data of ^{137}Cs and ^{60}Co are presented in Tables 5 and 6. Negative values of leaching parameters are observed for fitting the ^{137}Cs experimental data to ALL model lead to the violation of the model equation, so it could be concluded that this model cannot be used to represent the leaching phenomena. Also, the respectively low correlation coefficients for ^{137}Cs leaching from samples 3 and 4 and the negative values of samples 1 and 2 indicate that FRDDIM model is not the best to describe the leaching phenomena. This mean that ^{137}Cs leaching from all studied samples is attributed to first-order reaction exchange between the surface of waste matrix and the leaching solution followed by diffusion of ^{137}Cs. Where ^{60}Co leaching is resulting from four subsequent leaching mechanisms that include the release of loosely bound ^{60}Co radionuclides followed by first-order reaction exchange between the surface of waste matrix and the leaching solution then diffusion of ^{60}Co and finally dissolution. By comparing the order of the leaching parameters values for ^{60}Co leaching as listed in Table 6, it was found that the order of the values of CON and Q are much higher than those of D and U which indicate that surface mechanism (leach of loosely bound radionuclides and FRM) might control ^{60}Co leaching from the studied matrices.

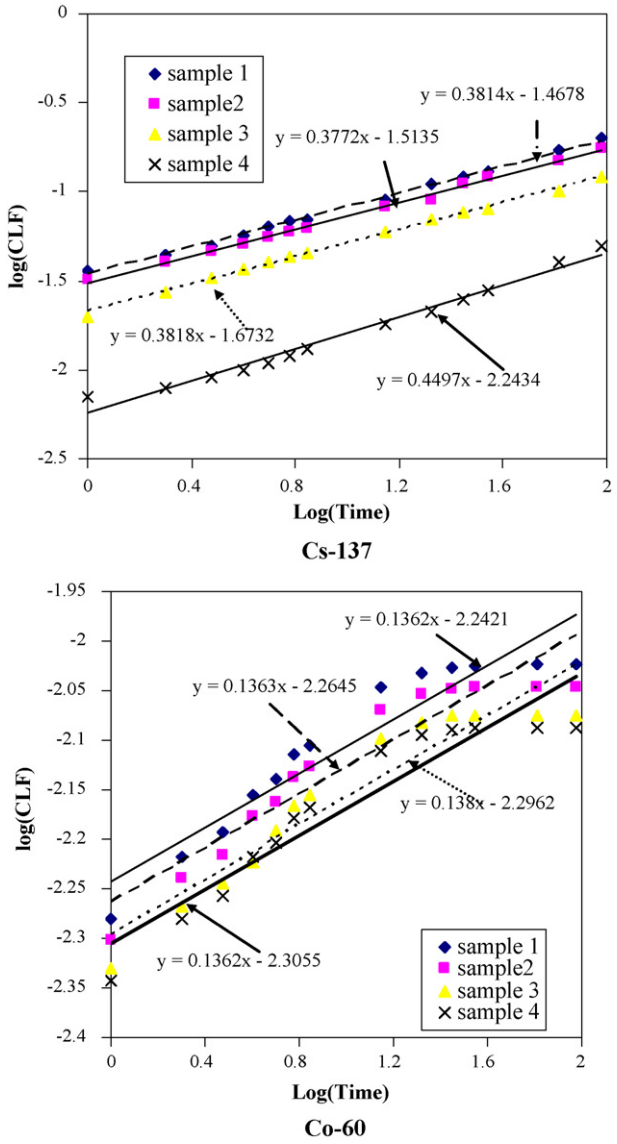

Fig. 8. Determination of the controlling leaching mechanism.

Table 7
Slope of the linear regression of log(CLF) versus log(time).

Radionuclide	Sample 1	Sample 2	Sample 3	Sample 4
Cs slope	0.38	0.38	0.38	0.45
Cs R^2	0.99	0.99	0.99	0.98
Co slope	0.14	0.14	0.14	0.14
Co R^2	0.89	0.89	0.89	0.89

Table 8
Lecahbility index of ^{137}Cs and ^{60}Co.

Radionuclide	Sample 1	Sample 2	Sample 3	Sample 4
Cs-137	3.82	3.92	4.22	5.32
Co-60	6.25	6.31	6.36	6.39

Various reported studies indicate that the determination of the controlling leaching mechanism could be conducted based on the slope of the linear regression of the logarithm of CLF versus the logarithm of time. If the slope is less than 0.35 the controlling leaching mechanism will be the surface wash-off, for the slope values ranging from 0.35 to 0.65 the controlling mechanism will be the diffusion, and higher slope values represent the dissolution mechanism [29]. The plots of the log(CLF) versus log(time) are illustrated in Fig. 8. The result of the linear regression for ^{137}Cs and ^{60}Co leaching data are listed in Table 7, it is clearly shown that the values of the correlation factor (R^2), have high values for ^{137}Cs leaching. The values of the slops for ^{137}Cs are in the range from 0.35 to 0.65 which indicate that the diffusion is the controlling leaching mechanism. Where for ^{60}Co radionuclides the correlation factors have low values and the slop values are less than 0.35 which indicate that surface wash-off control the leaching of ^{60}Co radionuclides which confirm the results obtained from Table 6.

The leachability index is a material parameter of the leachability of diffusing species, which used to catalogue the efficiency of a matrix material to solidify a waste and is defined as $L = -\log(D)$ [10,15]. The value of 6 is the threshold to accept a given matrix as adequate for the immobilization of radioactive wastes. As shown in Table 8, the leachability index of ^{137}Cs from the studied matrices is lower than 6 where those of ^{60}Co are higher than 6. These values indicated that all studied matrices can be catalogued as efficient materials for immobilizing incineration ashes containing ^{60}Co but not efficient for the immobilization of ^{137}Cs.

5. Conclusion

^{137}Cs and ^{60}Co leachability from different immobilized waste matrices were evaluated. The specific conclusions pertaining to the results presented herein can be drawn as follows:

(1) From the mechanical strength test, the extent of solidification of the studied waste matrices is acceptable based on a comparison with the compressive strength threshold of 35 kg/cm^2.
(2) The CLF of the studied radionuclides were significantly reduced by increasing the incineration ash in the cement grouts and this is due to the low porosity of the ash.
(3) By fitting the experimental data to FRM, DIM, and DM models it was found that the leaching phenomena of the studied waste matrices cannot represented by single mechanism.
(4) The leaching of ^{137}Cs was found to be as a result of first-order reaction exchange between the surface of waste matrix and the leaching solution followed by diffusion of ^{137}Cs through the waste matrix.
(5) The leaching of ^{60}Co was found to be as a result of four subsequent leaching mechanisms that include the release of loosely bound ^{60}Co radionuclides followed by first-order reaction exchange between the surface of waste matrix and the leaching solution then diffusion of ^{60}Co and finally dissolution.
(6) The surface wash off mechanisms was found to control ^{60}Co leaching where diffusion was found to control ^{137}Cs leaching.
(7) The efficiency of the studied waste matrices in immobilizing ^{137}Cs and ^{60}Co was tested by evaluating the leachability index, it was found that these matrices composition can efficiently immobilize ^{60}Co but cannot immobilize ^{137}Cs.
(8) It is recommended to improve the performance of ^{137}Cs stabilization in cement matrix to increase the waste loading (ash ratio) or add a material that have high sorption capacity towards ^{137}Cs but greater care should be give to the reduction in mechanical performance of these matrices.
(9) It is recommended when driving mathematical tool to predict the long-term behavior of the immobilized waste matrices to include surface leaching mechanisms such as leaching of loosely bound radionuclides and FRM in addition to the diffusion model.

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