



Leaching of ^{226}Ra and ^{40}K from cement based matrices

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Abstract

A study of the leaching behavior of immobilized contaminated soil, as radioactive waste, has been performed using an IAEA recommended semi-dynamic leaching test. Different radioactive waste matrices were prepared by solidifying soil contaminated by ^{226}Ra and ^{40}K radionuclides, with cement and bentonite as additive. The experimental data of this test were fitted to a semi-empirical mathematical model based on simple leaching rate mechanisms to evaluate the mechanisms that instigate the leaching phenomena. The results indicated that the controlling leaching mechanism is the diffusion but the leaching phenomenon of both radionuclides is the result of a combination of first order reaction and diffusion mechanisms.

Keywords: leaching mechanism; radioactive waste; IAEA semi-dynamic test; ^{226}Ra , ^{40}K ; semi-empirical model.

1. Introduction

The immobilization of radiocontaminants could be carried out with different matrices i.e. cementitious, polymeric, metallic, and glass-based materials. Among these matrices, cementitious materials are the most commonly used for immobilizing Low and Intermediate Level Radioactive Wastes (LILRW) [1, 2-4]. Conventional cementitious waste matrices consist of waste, cement, additives, and water [4, 5]. The additives are used to enhance the mechanical performance of the waste matrix and/or reduce the leachability [6-9]. Bentonite has been tested for its potential use as additive [6, 8, 10-12]. Limited studies in the literature investigated the immobilization of radio contaminants in cement-bentonite matrices. These studies assessed the leaching and mechanical performances of the produced matrices and estimated the diffusion coefficients [6, 12, 13]. The leaching of radioactive substances from wastes incorporated in cement is of great interest in waste management from the view

of environmental safety [14] since the leaching of radio nuclides can be reduced considerably by mixing the waste with cement mixture that causes it to solidify[15, 16].

This paper presents the results of a study that evaluate the leachability of a waste form using a semi-dynamic leaching test recommended by the International Atomic Energy Agency (IAEA) [17].

The waste form evaluated in this study was produced by solidifying a soil contaminated by NORM (Naturally Occurring Radioactive Materials) of Algerian oil fields. The cement-sand mixture with and without bentonite, were used. The radioactive contaminants of interest were radium (^{226}Ra) and potassium (^{40}K) natural radionuclides contained in NORM.

A semi-dynamic leaching test was carried out for 92 days on a waste form. Regression analyses were conducted on the cumulative fraction of radium and potassium leached from the solidified wastes, using a semi-empirical mathematical model based on simple rate leaching mechanisms. The resulting model parameter values give insights into the various leaching processes that occurred in the waste forms[15].

The cumulative leach fraction was calculated according to the following equation [18]:

$$\text{CLF} = \left(\frac{\sum(A_n)}{A_0} \right) \left(\frac{V}{S} \right) \quad (1)$$

Where:

$\sum(A_n)$: Cumulative radioactivity leached (Bq), A_n : radioactivity loss during the leaching period n (Bq), A_0 : initial radioactivity present in specimen (Bq), V: volume of specimen (cm^3), S: exposed surface area of specimen (cm^2).

2- Semi-empirical 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 [19].

Several studies [20-22] have shown that under mild leaching conditions, diffusion within the solid matrix usually controls the transfer of contaminants from the solid to the surrounding liquid phase. A diffusion model is often used to study the kinetics of leaching in order to predict long-term leachability of the waste components. An expression based on Fick's diffusion theory is often used to describe the release of pollutants from the solidified waste, with the aid of the following assumptions [23, 24]:

- 1- The mobility of a contaminant is limited by diffusion;
- 2- The specimen behaves as a semi-infinite medium, provided that the cumulative fraction leached does not exceed 20% [25];
- 3- The concentration of a contaminant at the specimen surface is approximately zero.

The diffusion model can be expressed as:

$$\text{CLF} = 2 \left(\frac{De}{\pi} \right)^{0.5} t_n^{0.5} \quad (2)$$

Where t_n : time (end of the leaching period) (s), De : effective diffusion coefficient (cm^2/s).

Assuming that the leaching rate is linear over a leachant renewal period n , the following solution can also be derived [26]:

$$\left(\frac{A_n}{A_0}\right) \left(\frac{V}{S}\right) \left(\frac{1}{\Delta t_n}\right) = \left(\frac{De}{\pi}\right)^{0.5} \frac{1}{T_n^{0.5}} \quad (3)$$

Where Δt_n : duration of the leaching period (s), T_n : time (middle of the leachant period) (s).

Equations (2) and (3) provide two ways of calculating the effective diffusion coefficient, De . With Equation (2), De can be calculated from the slope (m) of a plot of $\frac{\sum(A_n)}{A_0}$ versus the square root of time, ($t_n^{0.5}$). De is then expressed as [6, 13, 16, 18, 19]:

$$De = \frac{\pi}{4} m^2 \left(\frac{V}{S}\right)^2 \quad (4)$$

From Equation (3), an average value of De can be derived from the incremental fraction (A_n / A_0) leached at each period n [16].

The American Nuclear Society (ANS) [27] has proposed that the results can be a function of a coefficient called Leachability Index (LI). It can be interpreted as the negative logarithm of the effective diffusion coefficient, provided that leaching data support the hypothesis that diffusivity controls leachability.

This index can be used to compare the relative mobility of different contaminants on a uniform scale that varies from 5 ($De= 10^{-5} \text{ cm}^2/\text{s}$, very mobile or rapid diffusion) to 15 ($De=10^{-15} \text{ cm}^2/\text{s}$, immobile or very slow diffusion) [16, 28].

The value of LX is given by:

$$LI = \frac{1}{m} \sum_{n=1}^m \log\left(\frac{\beta}{De}\right) \quad (5)$$

Where β : constant ($1 \text{ cm}^2/\text{s}$), m : number of leaching periods [16, 23].

Several authors [16] have determined that, if the calculated coefficient De remains constant over the entire leaching period, it is possible to conclude that diffusion is the primary transport mechanism responsible for leaching. However, results obtained from some leaching tests [21, 29, 30] showed that mechanisms other than diffusion were also affecting the leaching process.

During migration, chemical species will react with other dissolved elements. They will also interact with solid surfaces via surface reactions and ion exchanges. Furthermore, some of these species will participate in precipitation/dissolution reactions [16].

The development of a model that takes into account all the leaching phenomena is mathematically very complex and the resultant expression is not likely to have practical application. To overcome this problem, authors [15] have adopted a semi-empirical method that combines the general mathematical forms of different conceptual models that include diffusion and various chemical reactions (i.e. adsorption or surface dissolution) to obtain a model that describes the leaching characteristics of a waste component. The resulting model parameter values give insights into the various leaching processes that occurred in the waste forms [16].

In this model, the cumulative leached fraction of contaminant is expressed by:

$$CLF = k_1(1 - \exp(-k_2t)) + k_3 t^{0.5} + k_4t \quad (6)$$

k_1 is a constant representing the immediate dissolution (instantaneous wash-off);

k_2 is a rate constant and the term $k_1(1 - \exp(-k_2t))$ describes the kinetics of exchange of species between the surface of the solid waste form and solution (pore water or leachant).

In the case where the initial surface exchanges are fast relative to the time span of the data being analyzed, this term can be reduced to the constant k_1 .

k_3 is a constant representing the diffusion controlled transport mechanism and k_4 is a constant representing leaching from a slow chemical reaction, such as corrosion or structural breakdown of the waste form matrix [16, 31].

2. Experimental

2.1. Materials

The studied matrixes were prepared using commercial Ordinary Portland Cement (OPC) with sand as aggregate with or without bentonite as additive. The OPC was provided from a cement Algerian company and the bentonite from an Algerian Mining Company.

Soil contaminated by NORM was taken from an Algerian oil and gas field. The chemical composition of the OPC, bentonite and contaminated soil are given in Table 1.

Before used, the contaminated soil considered as radioactive waste, was dried with infrared lamp and grinded to fine powder. This powder was placed into polyethylene containers, sealed during at least three weeks in order to ensure the secular equilibrium between ^{226}Ra and its gas daughter ^{222}Rn , before carrying out the spectrometric measurements.

Table 1: Chemical composition of OPC, bentonite and contaminated soil (%)

Chemical component	OPC	Contaminatedsoil	Bentonite
SiO ₂	20.93	28.21	55.7
CaO	64.51	11.85	0.23
Na ₂ O	0.28	7.78	0.486
Al ₂ O ₃	5.23	6.37	14.5
Fe ₂ O ₃	2.59	3.29	1.25
K ₂ O	0.50	1.51	0.800
TiO ₂	0.251	0.32	0.075
BaO	-	0.61	0.044
Total activity of ²²⁶ Ra (Bq)		508 – 574	
Total activity of ⁴⁰ K (Bq)		130 - 146	

2.2. Preparation of specimens

The immobilization matrices were prepared by mixing cement-sand with respectively 0, 5 and 10 wt% of bentonite. Each matrix was loaded by 15wt% of radioactive waste. Matrix with wt 0% bentonite was used as reference one. Water was added to the mixture at a ratio of range 0.3-0.4. Each mixture was placed in the cylindrical moulds (4.4 cm in diameter and 1.5 cm high). These moulds were kept in air for 24 h and then were demoulded and cured. The curing time of the specimen was ranged from 28 to 30 days in a humid atmosphere at ambient temperature.

Each matrix was prepared in triplicate and the average of the results was taken in account.

Initial total activities (A_0) of the ²²⁶Ra and ⁴⁰K contained in matrices are reported in Table 1.

2.3. Leaching test

A semi-dynamique leaching test was performed, using distilled water as leachant (pH= 6.6), to study the leaching of ²²⁶Ra and ⁴⁰K from hardened cement-waste forms.

The recommended IAEA’s standard test [17]was applied. Each cured specimen was immersed in beaker, so that the volume of the leachant to the waste matrix surface area equal 10 cm.

The leachant was renewed at frequent intervals and the activities of the radionuclides leached during each interval were measured.The duration of leaching renewal period was 92 days. The leachate was totally replaced with fresh water after 1, 2, 3, 6, 9, 16, 37, 62 and 92 days.The leachate sample was analyzed using Ge(HP) gamma spectrometer with a relative efficiency of 30% and a resolution of 1.8

keV, for the photon of 1332 keV of the ^{60}Co . The measurement of the background radiation was performed periodically. The efficiency of the equipment was calibrated.

The measurement of the ^{40}K was performed directly by its own-ray at 1460.8 keV, while the measurement of the ^{226}Ra was performed through the detection of the gamma emission of ^{214}Bi at 609 keV and ^{214}Pb at 352 keV and the mean of their activities was taken as results.

To obtain meaningful statistics in the measurements, the time of spectrometry for each sample was between 24 and 48 h.

3. Results and discussion

The result of the IAEA leach test reflects the physical changes and chemical interactions that occurred within the tested matrix and the magnitude of the cumulative leach fraction is an index for the release of the nuclide from the cement matrix.

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 [19, 32].

The variation of cumulative leach fractions of ^{226}Ra and ^{40}K radionuclides incorporated in cement and cement mixed with bentonite are depicted in Figure 1 and Figure 2 respectively. The examination of the CLF versus time indicated that the addition of 5 and 10% bentonite is accompanied by a reduction in CLF [13]. In addition, in all studied cases, ^{40}K has the highest values of cumulative leach fraction than ^{226}Ra .

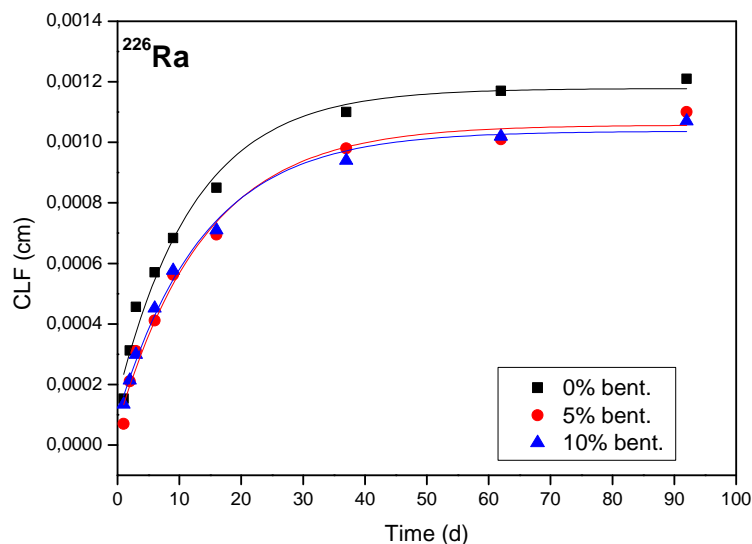


Figure 1: Regression lines and experimental data for the CLF of ^{226}Ra

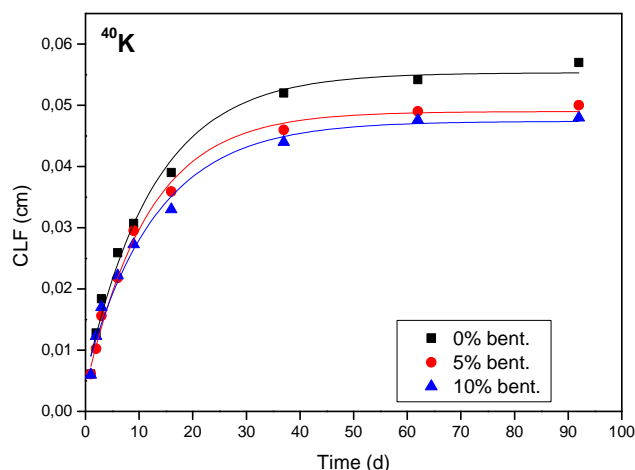


Figure 2: Regression lines and experimental data for the CLF of ^{40}K

Several studies showed that alkaline metal salts, such as potassium, are generally very soluble species[33], contrary to radium which is better retained by minerals and especially by clays minerals as bentonite [13, 34].

It is known that bentonite has a high surface area capable of reducing the volume of large pores and capillaries that are normally found in cement pastes. Its high Cation Exchange Capacity (CEC) can also play a role in the retention way of radionuclides [13, 18].

Other studies results showed that both radium and potassium could be sorbed on bentonite. Sorption might be dominated by ion exchange for radium and by surface complexation reaction for potassium [35].

So, this decrease in time of CLF can be justified by a slow but progressive fixation of the radionuclides by bentonite and this fixation is slower for ^{40}K than for ^{226}Ra . This result has been observed by authors [34] when they studied the behavior of these radionuclides with respect to mineral clays in sediments.

The theoretical leaching model described in paragraph above (Equation (6)), considered the behavior of a single species, not taking into account the transport and reactions of different species present in the solid, which could change the chemical environment [16].

The derivation of the model was also based on simple boundary conditions:

It was assumed that the specimen behave as a semi-infinite media provided that the cumulative fraction leached ($\frac{\sum(A_p)}{A_0}$) does not exceed 20% [15]. This is the case for the two radionuclides. The maximum cumulative fractions leached of ^{226}Ra and ^{40}K is found less than 0.3 % and 14%, respectively.

In previous studies [20-22], it was shown that under the most conservative approach, the radiocontaminants transport from the waste matrix is much faster than diffusion within the waste matrix. This leads to the assumption that the concentration at the waste matrix edge equal zero [15, 16].

Various reported studies [13, 32, 36] 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 slope values higher than 0.65 represent the dissolution mechanism.

Table 2: Slope of the linear regression of log(CLF) versus log(time).

Radionuclide	0 wt% Bent.	5 wt% Bent.	10 wt% Bent.
²²⁶ Ra Slope	0.42	0.54	0.46
²²⁶ Ra R ²	0.89	0.87	0.94
⁴⁰ K Slope	0.46	0.46	0.43
⁴⁰ K R ²	0.91	0.93	0.92

The plots of the log(CLF) versus log(time) are illustrated in Figure 3 and Figure 4. The calculated slopes in Table 2 were found to lie in the range 0.35–0.65, which indicates that the controlling leaching mechanism for both radionuclides of interest is diffusion.

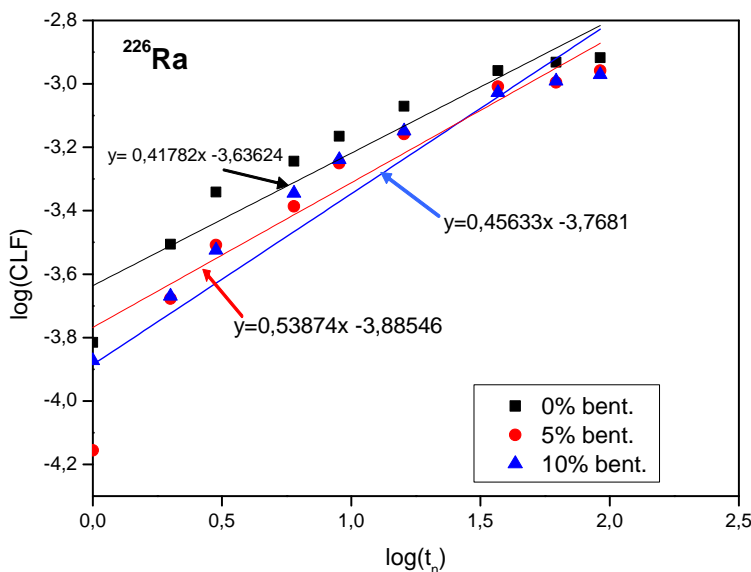


Figure 3: Determination of the controlling leaching mechanism of ²²⁶Ra

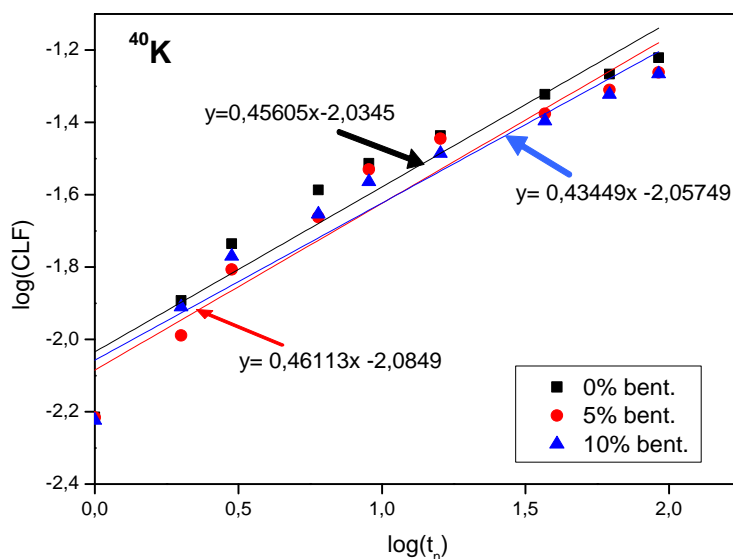


Figure 4: Determination of the controlling leaching mechanism of ^{40}K

Thus, the assumptions made to derive Equation (6) appear to be applicable in this study and the parameter values in Table 3 can be used to gain insights into the various leaching mechanisms that occurred in the waste forms.

Regression analyses using Equation (6) were conducted on the CLF of both radionuclides leached from the waste forms and the regression lines are compared to the observed data in Figure 1 and Figure 2.

For all matrices, a linear relationship between the CLF and time was not observed. It was therefore concluded that the dissolution rate was not a controlling factor. Then, parameter k_4 is not significant in the linear regression analysis. Conversely, the surface phenomena term, $(k_1(1 - \exp(-k_2t))$ which is rate-limiting for small values of time, is present through the values of k_1 and k_2 of Table 3 [15].

In order to determine the effective diffusion coefficient De , $(\frac{\sum(A_n)}{A_0})$ was represented against the square root of time $((t_n)^{0.5})$ [13, 32] (Figure 5 and Figure 6). By examining the figures, it was found that in all plots, there is an initial fast leaching during the first period followed by slow leaching in subsequent periods. Most of the researchers calculated the diffusion coefficient, according to Equation (4), by fitting the fast and slow leaching to single line and the mean values of slopes of these lines were utilized to estimate the diffusion coefficients [6, 8, 18, 19, 37-41]. Effective diffusion coefficients calculated and expressed as $pDe = -\log De$ are reported in Table 3.

Table 3.Leaching mechanism parameters

Radionuclide	Controlling mechanisms			Chemical reaction or corrosion (k_4)	pDe ^b	LI	
	Surface phenomena ($k_1(1-\exp(-k_2t))$)	Diffusion ($K_3t^{0.5}$)					
²²⁶ Ra	k_1^a	k_2^a	k_3^a	k_4^a			
	($\times 10^{-3}$)	($\times 10^{-2}$)	($\times 10^{-4}$)				
	0 wt% Bent.	1.02	8.01	1.55	-	12.85	13.10
	5 wt% Bent.	0.99	7.06	0.66	-	12.89	13.21
10wt% Bent.	0.94	7.23	0.97	-	12.91	13.19	
⁴⁰ K	($\times 10^{-2}$)	($\times 10^{-2}$)	($\times 10^{-3}$)				
	0 wt% Bent.	5.00	7.84	5.33	-	9.50	9.78
	5 wt% Bent.	4.54	8.60	3.57	-	9.57	9.85
	10wt% Bent.	4.14	7.60	6.00	-	9.61	9.86

pDe = - log De.

^aObtained from Eq. (6).

^bCalculated from Eq. (4).

^cCalculated from Eqs. (3) and (5).

A dash (-) indicates that the value of k_4 was no significant.

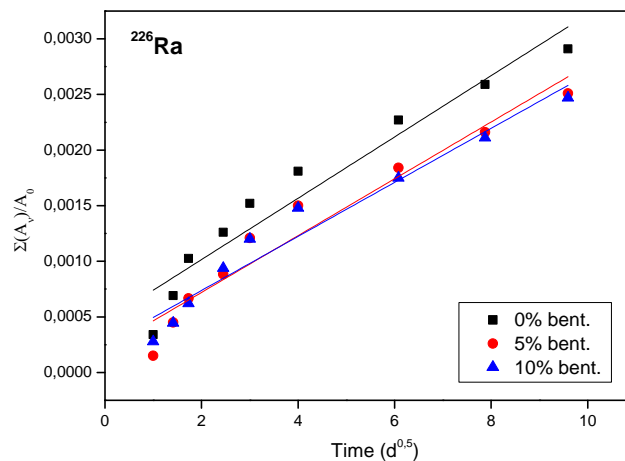


Figure 5: Variation of ²²⁶Ra fraction leached from cement and cement mixed with bentonite vs. square root of time.

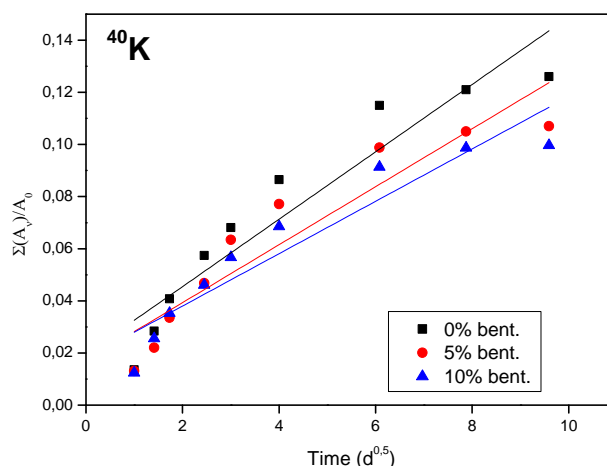


Figure 6: Variation of ⁴⁰K fraction leached from cement and cement mixed with bentonite vs. square root of time.

The experimental data were also used to calculate the leachability index (LI) of Equation (5). The value of LI was calculated from Equation (3). The arithmetic average of LI obtained with all the leaching periods is presented in Table 3. Examination of these values leads to the conclusion that each radionuclide has a similar leaching behavior for all matrices, since similar values of pDe and LI were obtained. According to the parameters presented in Table 3, the leaching mechanism results in the first order reaction and diffusion primarily whereas the dissolution mechanism has negligible contribution to the leaching, if existed.

In addition, the ⁴⁰K has a high mobility (LI < 10) while the ²²⁶Ra has a low mobility (LI > 13) and taking into account that the leaching index of ⁴⁰K is more than three units lower than the index of ²²⁶Ra (Table 3), reflects that ⁴⁰K has greater mobility than ²²⁶Ra.

Following the results of some researchers who found that the higher the LI and lower the De, the better the retention of contaminant in the matrix [42], we can say that the ²²⁶Ra is better retained in the cement and cement-bentonite.

Taking into account that the leaching index obtained with the immobilized waste samples greater than 12, suggests that immobilization process is acceptable [43], we can say that the process is acceptable for the ²²⁶Ra and less for the ⁴⁰K, in this case.

Conclusion

The behavior of ²²⁶Ra and ⁴⁰K immobilized in cement and cement bentonite matrices was studied. This study was carried out by applying a semi-empirical model to the cumulative fraction of radioactivity leached and model parameters were estimated.

Experimental data showed that the addition of bentonite allowed a better retention of radionuclides since the CLF were lower.

The leachability index and diffusion coefficients for ²²⁶Ra and ⁴⁰K were calculated provide a measure of the mobility of the contaminants in the immobilized waste matrices.

Both radionuclides have similar behaviors for all matrices studied (with or without bentonite). However, ^{40}K presents greater mobility than ^{226}Ra (LI of ^{40}K is 1.34 units lower than LI of ^{226}Ra).

Finally, despite the controlling leaching mechanism was found to be diffusion for all the studied matrices, the leaching phenomenon for both radionuclides, was found to occur as a result of first order reaction and diffusion mechanism primarily.

References

1. R. O. Abdel Rahman, D. H. A. Zin El Abidin, H. Abou-Shady, *Chem. Eng. J.* 245 (2014) 276-287.
2. IAEA (International Atomic Energy Agency), *TECDOC-1255*, Vienna (2001).
3. IAEA (International Atomic Energy Agency), *TECDOC-1701*, Vienna (2013).
4. Z. Drace, I. Mele, M. Ojovan, R. O. Abdel Rahman, *MRS Proceedings* 1475 (2012).
5. F. P. Glasser, Application of inorganic cements to the conditioning and immobilisation of radioactive wastes. *Handbook of Advanced Radioactive Waste Conditioning Technologies*. In M. Ojovan (Eds.) 67–135 (2011).
6. R. O. Abdel Rahman, A. M. El-Kamash, A. A. Zaki, *J. Hazard. Mater.* 45 (2007) 372–380
7. I. B. Plecas, *Ann. Nucl. Energy.* 30 (2003) 1899-1903.
8. S. Goni, A. Guerrero, M.P. Lorenzo, *J. Hazard. Mater.* 137 (2006) 1608-1617
9. H. Matsuzuru, N. Moriyama, Y. Wadachi and A. Ito, *Heal. Phys.* 32 (1977) 529-534
10. A. Guerrero, S. Goni, *Waste Manag.* 22 (2002) 831-836.
11. A. Fernandez-Jimenz, D. E. Macphee, E. E. Lachowsk and A. Dalomo, *J. Nucl. Mater.* 346 (2005) 185–193.
12. A. E. Osmanlioglu, *Waste Manage.* 22 (2002) 481-483.
13. R. O. Abdel Rahman, D. H. A. Zin El Abidin, H. Abou-Shady, *Chem. Eng. J.* 228 (2013) 772-780.
14. H. Matsuzuru, A. Ito, *Ann. Nucl. Energy.* 4 (1977) 465-470.
15. P. L. Côté, T. W. Constable, A. Moreira, *Nucl. Chem. Waste Manage.* 7 (1987) 129-139.
16. A. André, I. Ortiz, J. R. Viguri, A. Irabien, *J. Hazard. Mater.* 40 (1995) 31-42.
17. E. D. Hespe, *At. Energy Rev.* 9 (1971) 195-207.
18. A. M. El-Kamash, A. M. El Dakroury, H. F. Aly, *Cem. Concr. Res.* 32 (2002) 1797-1803.
19. A. M. El-Kamash, M. R. El-Naggar, M. I. El-Dessouky, *J. Hazard. Mater.* 136 (2006) 310-316.
20. H. W. Godbee, O. U. Anders, R. M. Jr. Neilson, *Nucl. Chem. Waste Manage.* 1(1980) 29-35.
21. P. L. Côté, D.P. Hamilton, Leachability comparison of four hazardous waste solidification processes. *Proceeding of the 38th Conference on Industry Waste*, Purdue Univ., 395-401, 1983.
22. T. M. Brown, P. L. Bishop, D. L. Gress, Use of an upflow column leaching test to study the release patterns of heavy metals from stabilized/solidified heavy metal sludges in Lorenzen D., Conway R.A., Jackson L.P., Hamza A., Perket CL. and Lacy W.J. (Eds.), *ASTM STP 933, ASTM, Philadelphia*, 6 (1986) 79-90.
23. H. W. Godbee, D. S. Joy, *Oak Ridge National Laboratory Report N° ORNL/TM-4333*, Tennessee (1974).
24. P. L. Bishop, *Hazard. Waste and Hazard. Mater.* 5(2) (1988) 129-143.
25. M. W. Abdel Raouf, H. G. Nowier, *J. Environ. Eng.* 130 (5) (2004) 499.
26. P. L. Côté, D. Isabel, 3rd Symposium on Hazardous and Industrial Waste Management and Testing, in Jackson L.P., Rohlik A.R. and Conway R.A. (Eds.), *ASTM STP 851*, Philadelphia (1984) 48-60.
27. ANSI/ANS-16.1, in *American Nuclear Society* (Ed.), Illinois (1986).
28. J.S. Nathwani, C.R. Phillips, *Water Air and Soil Pollution*, 14 (1) (1980) 389-402.

29. P.L. Bishop, in *Emerging Technologies in Hazardous Waste Management II, ACS Symp. Ser.*, 15 (1991) 302-315.
30. E. Zamorani, F. Lanza, G. Serrini, H. Blanchard, *Nucl. Chem. Waste Manage.* 6 (1986) 197-202
31. R. J. Serne, R. O. Lokken, L. J. Criscenti, *Waste Manage.* 12 (1992) 271-287.
32. R. O. Abdel Rahman, A.A. Zaki, *Chem. Eng. J.* 155 (2009) 698–708.
33. L. R. T. Barna, R. Barna, P. Moszkowicz, *Environ. Scie. Technol.* 35 (2001) 149-156.
34. R. A. Ligeró, F. Fera, M. Casas-Ruiz, C. Corredor, *J. Environ. Radioact.* 87 (2006) 325-334.
35. Y. Tachi, T. Shibutani, H. Sato, M. Yui, *J. Contam. Hydr.* 47(2001) 171-186
36. D. H. Moon, D. Dermatas, *Eng. Geol.* 85 (2006) 67–74.
37. K. G. Papadokostaki, A. Savidou, *J. Hazard. Mater.* 171(2009) 1024-1031.
38. G. Bar-Nes, A. Katz, Y. Peled, Y. Zeiri, *Cem. Concr. Res.* 38 (2008) 667–674.
39. D. Anji Reddy, S. K. Khandelwal, R. Muthiah, A. G. Shanmugamani, P. K. Sinha, *Ann. Nucl. Energy*, 37 (2010) 934-941.
40. P. K. Sinha, A. G. Shanmugamani, K. Renganathan, R. Muthiah, *Ann. Nucl. Energy*, 36 (2009) 620-625.
41. R. O. Abdel Rahman, A.A. Zaki, *Chem. Eng. J.* 173 (2011) 722-736.
42. D. Singh, V.R. Mandalika, S.J. Parulekar, A.S. Wagh, *J. Nucl. Mater.* 348 (2006) 272-282.
43. X. N. Yang, W. Y. Zhou, R. S. Wang, *J. Radioanal. Nucl. Chem.* 198 (2) (1995) 287-294.