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Accelerated Leach Test for Low-level Radioactive Waste Forms in the Hungarian NPP Paks

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Abstract

An accelerated leach test method was used for low-level radioactive waste forms in the Hungarian NPP Paks. These experiments were performed using cylinders prepared form Hungarian cement type CEM I 32,5 LH and CEM III/B 32, N-LH/SR. Each cylinder was made using cement or cement plus additive using radioactive waste water. The cemented radioactive material was evaporator bottom residue or sludge as well as evaporator cleaning acid solution, spent ion exchange resin, decontamination solution from NPP Paks, containing $^{134}C_s$, $^{137}C_s$ and $^{60}C_o$ as main radioactive components. Leach tests were performed according to ASTM C 1308-08 standard. A computer program (ILT15) associated with the accelerated leach test was developed based on the ASTM C 1308-08 standard. Literature test and measured leaching data were analyzed to assess whether the model for diffusion from a finite cylinder describes leaching from cement based waste forms. In this paper some of the experimental and modeling work used to validate the test method are presented.

Keywords: Radioactive waste; Leach test; Cement; Computer program; Diffusion

Introduction

For a number of years increasing attention has been given in Hungary to the management of the low and medium level radioactive wastes (LLW, MLW) being produced in Paks nuclear power plant.

Some of these wastes, for example, evaporator bottom concentrates, pond sludge and spent ion exchange media are produced in relatively large volumes. In addition to national programs on the development of immobilization processes, the European Community commissioned programs on the immobilization of LLW and MLW. These wastes are immobilized by incorporating them into cement. In order to optimize these immobilization processes, for example with respect to waste loading, it was necessary to characterize the products with respect to such properties as density, strength, dimensional stability, leach resistance and so on. In this article we report about an accelerated leach test and the developed computer program.

Experimental

Accelerated leach tests

An accelerated leach test method was used for low-level radioactive waste forms in the Hungarian NPP Paks. The leach test method was designed to minimize experimental artifacts that could be misinterpreted as release mechanisms such as effects of increased ionic concentrations in the leachate. This is particularly important when the data is used for mechanistic interpretations and long-term extrapolations.

The test method we used was a semi-dynamic leach test that is the leachant is changed at predefined intervals. The test requires a volume of distilled water that is 10 times the surface area of the sample. The leachant was changed twice on the first day and then daily for 11 more days.

These experiments were performed using cylinders of 2.93 cm diameter and 4.96 cm height, prepared form Hungarian cement type CEM I 32,5- LH and CEM III/B 32, N LH/SR. Each cylinder was made using cement or cement plus additive using radioactive waste water, with a surface to liquid volume ratio of 10. The cemented radioactive material we used was evaporator bottom residue or sludge as well as evaporator cleaning acid solution, spent ion exchange resin, decontamination solution from NPP Paks, containing $^{134}C_s$, $^{137}C_s$ and $^{60}C_o$ as main radioactive components. Leach tests were performed according to ASTM C 1308-08 standard. The leachant was distilled water with a conductance of less than 5 μS/cm. The leached radioactivity was determined using gamma spectrometry. Experiments run at 20°C. Results of the leach tests were expressed as cumulative fraction leached (CFL) and an effective diffusion coefficient (De) was determined. A model for bulk diffusion from a finite cylinder is used as a guide to determine if diffusion is the rate controlling transport mechanism during leaching. A value for the optimum De is obtained by first estimating De using the semi-infinite medium approach. Then for CFL values higher than 0.2 iterations were performed with the finite cylinder model to obtain the De value that gives the lowest sum of the residuals for the entire set of data. The goodness-of-fit between the data and the model result is evaluated by expressing the sum of the residuals as a percentage of the maximum CFL of the experimental data. If the error was less than 0.5%, then it is taken to mean that diffusion is the dominant leaching mechanism.

The ILT15 program

A computer program (ILT15) associated with the accelerated leach test was developed, based on the ASTM C 1308-08 standard (Figure 1). The

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computer program allows the user to test experimental results against a model for diffusion from a finite cylinder. If the model and the data fit within preset criteria then the diffusion model can be used to make projections of releases. Leaching data were analyzed to assess whether the model for diffusion from a finite cylinder describes leaching from cement based waste forms. In this paper some of the experimental and modeling work used to validate the test method are presented.

The ILT15 program contains four mathematical models that can be used to represent the data.

The mathematical models describing leaching mechanisms are as below:

1- Diffusion through a semi-infinite medium (for low fractional releases),

2- Diffusion through a finite cylinder (for high fractional releases),

3- Diffusion plus partitioning of the source term,

4- Solubility limited leaching.

The program was written in C++ in the Borland C++ Builder programming environment. The detailed description of this computer program will be described elsewhere.

Results and Discussion

The developed ILT15 accelerated leach test computer program was tested with leach test data available from literature. Data were used from the US ASTM C-1308-08 standard (Table 1 Test1, Test2, Test3) [1], and from the cesium leaching data of the Users' Guide for the Accelerated Leach Test Computer Program ALT [2-5] (Cs-137).

The ASTM leach data (Test1, Test2 and Test3) are shown in Table 1, and the Cs-137 data in Table 2.

Based on these data (Cs-137, Test1, Test2 and Test3) we fitted all the four leaching models using the ILT15 program. The fitting results had the same diffusion coefficients, percent relative errors as in the earlier ALT computer program. For example in case of Cs-137 leach data series the inputted leach test data and measured (blue) and calculated (red)

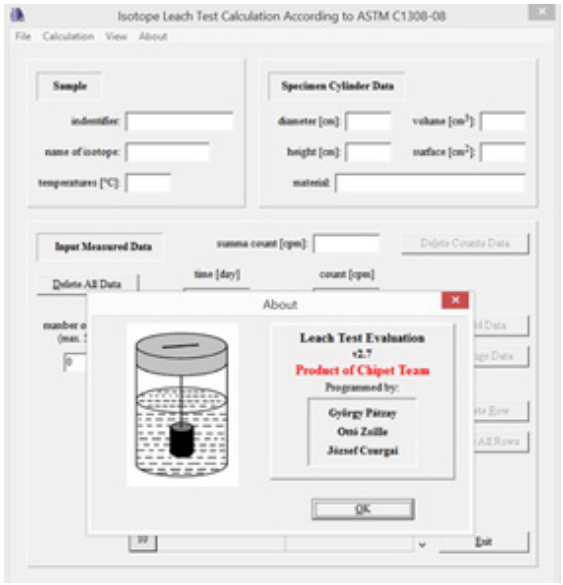

Figure 1: The starting picture of the ILT15 computer program Cs-137 [2].

Time (days)	Test1		Test2		Test3	
	IFL	CFL	IFL	CFL	IFL	CFL
0.083	0.0612	6.12000E-2	0.0606	6.06000E-2	0.0609	6.09000E-2
0.29	0.0582	1.19400E-1	0.0413	1.01900E-1	0.0396	1.00500E-1
1.0	0.104	2.23400E-1	0.0743	1.76200E-1	0.089	1.89500E-1
2.0	0.0827	3.06100E-1	0.0661	2.42300E-1	0.0745	2.64000E-1
3.0	0.0551	3.61200E-1	0.0468	2.89100E-1	0.061	3.25000E-1
4.0	0.0398	4.01000E-1	0.0385	3.27600E-1	0.0395	3.64500E-1
5.0	0.0337	4.34700E-1	0.0358	3.63400E-1	0.0345	3.99000E-1
6.00	0.0245	4.59200E-1	0.0248	3.88200E-1	0.0245	4.23500E-1
7.0	0.0245	4.83700E-1	0.0248	4.13000E-1	0.025	4.48500E-1
8.0	0.0214	5.05100E-1	0.022	4.35000E-1	0.0265	4.75000E-1
9.0	0.0184	5.23500E-1	0.022	4.57000E-1	0.02	4.95000E-1
10.0	0.0153	5.38800E-1	0.0193	4.76300E-1	0.0225	5.17500E-1
11.0	0.0153	5.54100E-1	0.0193	4.95600E-1	0.017	5.34500E-1

Table 1: Leaching data from the ASTM C-1308-08 standard [1].

Time (days)	Cs-137	
	IFL	CFL
0.083	0.0709	7.09000E-2
0.29	0.0639	1.34800E-1
1.0	0.13	2.64800E-1
2.0	0.103	3.67800E-1
3.0	0.0642	4.32000E-1
4.0	0.054	4.86000E-1
5.0	0.0358	5.21800E-1
6.00	0.0351	5.56900E-1
7.0	0.0289	5.85800E-1
8.0	0.0223	6.08100E-1
9.0	0.0206	6.28700E-1
10.0	0.0143	6.43000E-1
11.0	0.0172	6.60200E-1

Table 2: Leaching data of 137 Cs from cement/sodium sulfate specimen at 20°C [2].

leaching curves using the diffusion plus partition model are shown in Figures 2 and 3.

In case of Test1 [1] leach data series the inputted leach test data and measured (blue) and calculated(red) leaching curves using the diffusion plus partition model are shown in Figures 4 and 5.

While testing the ILT15 computer program we found that it is suitable for modeling and extrapolation of the measured accelerated leach test data in the NPP Paks. Therefore we use the ILT15 computer program to evaluate the measured accelerated leach test data at the NPP. As an example we investigated leaching characteristics of a specimen prepared form CEM I 32,5- LH containing cemented evaporator bottom residue from tank 02TW80B003 of NPP Paks shown in Table 3.

The calculated leach modeling data and curve are shown in Figures 6 and 7 for isotope Cs-137 and Figures 8 and 9 for Cs-134 [3,6-8].

According to modeling results of the test data and the measured

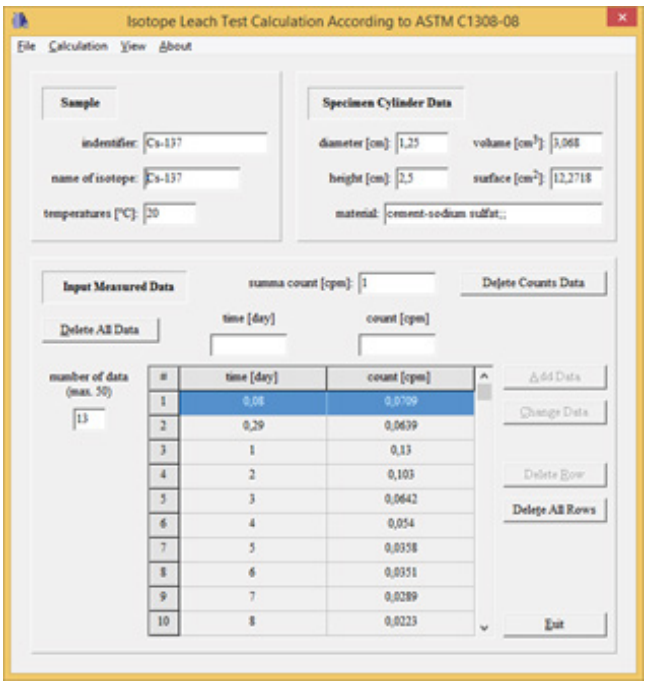

Figure 2: Test data Cs-137 [2] input.

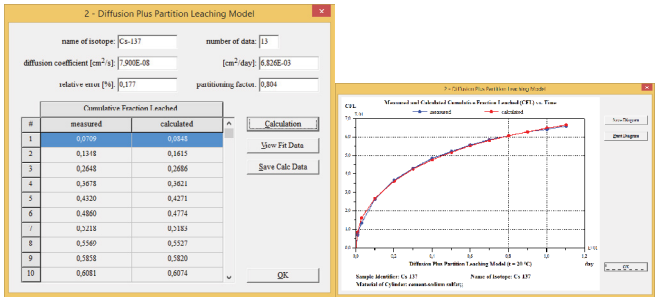

Figure 3: Test data Cs-137 [2] fitting results with model diffusion plus partition leaching Test1 [1].

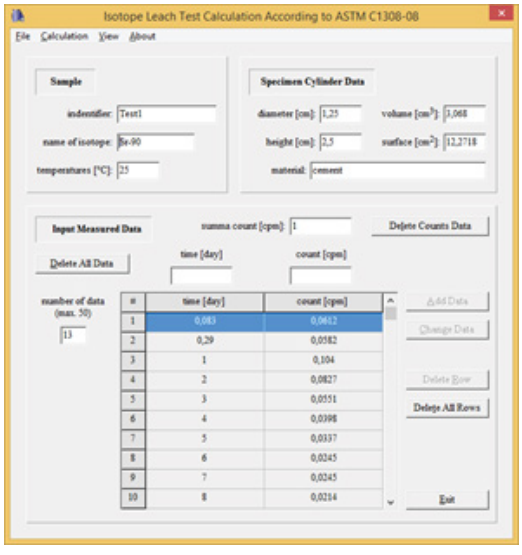

Figure 4: Test data Test1 [1] input.

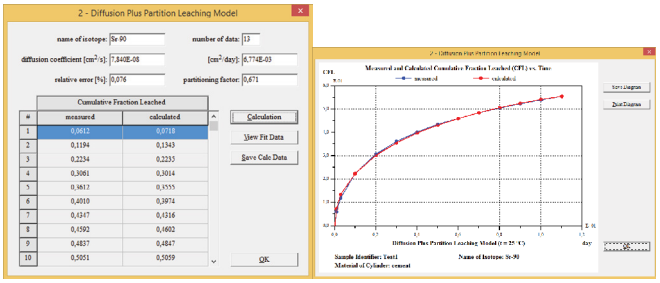

Figure 5: Test data Test1 [1] fitting results with model diffusion plus partition leaching.

Figure 6: The measured leach test data for CEM I specimen and isotope Cs-137.

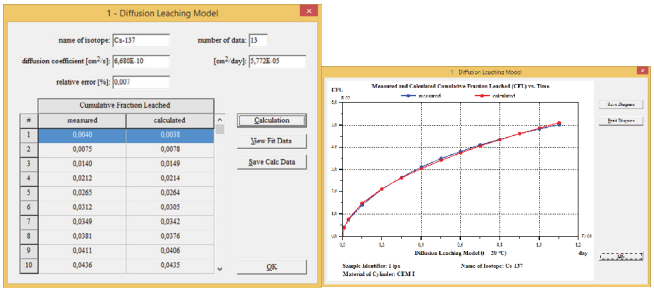

Figure 7: The calculated leach test data for CEM I specimen and isotope Cs-137 using the diffusion leaching model.

Characteristics	Value
mass	57.62 g
diameter	2.85 cm
height	4.80 cm
volume	30.671 cm^3
surface	55.74 cm^2
liquid volume	557 cm^3
total ^{137}Cs activity in the specimen	63.747.83 Bq
total ^{134}Cs activity in the specimen	451.21 Bq
total ^{60}Co activity in the specimen	2664.63 Bq

Table 3: Leaching characteristics of a specimen prepared form CEM I 32,5- LH containing cemented evaporator bottom residue from tank 02TW80B003.

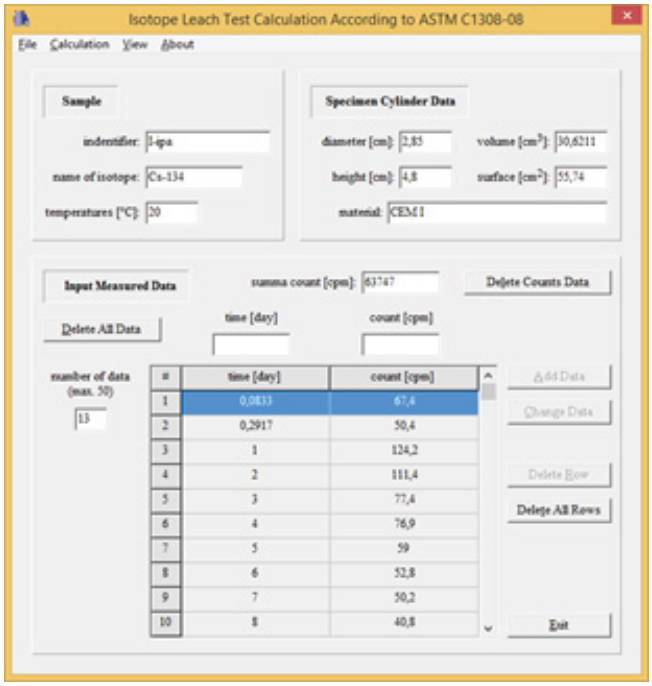

Figure 8: The measured leach test data for CEM I specimen and isotope Cs-134.

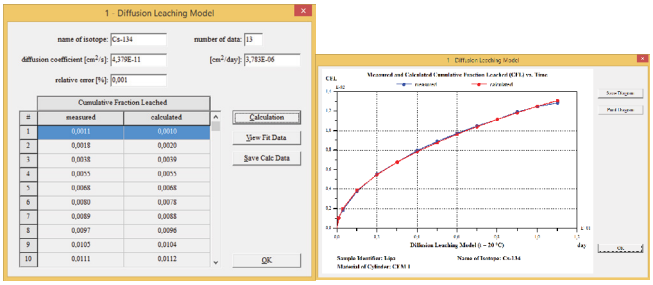

Figure 9: The calculated leach test data for CEM I specimen and isotope Cs-134 using the diffusion leaching model.

at the NPP accelerated leaching test data we concluded, that the ILT15 computer program, constructed according to ASTM C 1308-08 standard is a suitable too, to evaluate the leach test characteristics of cemented speciments. Best fittings were found using the diffusion and the diffusion plus partitioning models. The fitting errors were between 0.001-0.007% [9-15].

Conclusion

An accelerated leach test method was used for low-level radioactive waste forms in the Hungarian NPP Paks. These experiments were performed using cylinders of 2.93 cm diameter and 4.96 cm height, prepared form Hungarian cement type CEM I 32,5 LH and CEM III/B 32, N-LH/SR. Each cylinder was made using cement or cement plus additive using radioactive waste water, with a surface to liquid volume ratio of 10. The cemented radioactive material was evaporator bottom residue or sludge as well as evaporator cleaning acid solution, spent ion exchange resin, decontamination solution from NPP Paks, containing $^{134}C_s$, $^{137}C_s$ and $^{60}C_o$ as main radioactive components. Leach tests were performed according to ASTM C 1308-08 standard. The leachant was distilled water with a conductance of less than 5 μS/cm. The leached radioactivity was determined using gamma spectrometry.

A computer program (ILT15) associated with the accelerated leach test was developed based on the ASTM C 1308-08 standard. The computer program allows the user to test experimental results against a model for diffusion from a finite cylinder. If the model and the data fit within preset criteria then the diffusion model can be used to make projections of releases. Literature test and measured leaching data were analyzed to assess whether the model for diffusion from a finite cylinder describes leaching from cement based waste forms. In this paper some of the experimental and modeling work used to validate the test method are presented.

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