

## LEACHING KINETICS OF Cs<sup>+</sup> AND Co<sup>2+</sup> UNDER DYNAMIC CONDITIONS

by

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The possibility of retaining Cs<sup>+</sup> and Co<sup>2+</sup> bound by immobilization processes in the cement matrix is defined as the subject of its investigation: the cement matrix formulation, the water/cement ratio, the amount of waste, and the porosity of such a structure. Implementing the standard leaching method by Hesse the possibility of comparing different authors' results was achieved. Diffusion and semi-empirical model were used to investigate the transport phenomenon in order to predict the leaching level for a long period of time. Leaching of Co<sup>2+</sup> and Cs<sup>+</sup> ions under dynamic conditions immobilized in the cement matrix dynamic conditions decreases with the increase of the sludge content, regarding porosity increase. The effects of the diffusion and surface washing are equalized, and the contribution of the matrix dissolution to the Cs<sup>+</sup> and Co<sup>2+</sup> transport in the cement porous media increases, on average, for one order of magnitude. The semi-empirical model gives a better approximation for Co<sup>2+</sup> and Cs<sup>+</sup> leaching process for the duration of the experiment while both models significantly approximate leaching results in dynamic conditions.

*Key words: radioactive waste, cement matrix, leaching, mathematical modeling*

### INTRODUCTION

The activity of the nuclear industry generates significant amounts of liquid and solid radioactive waste (RW). The RW is produced in the processes of the nuclear fuel cycle, the industry of exploitation, processing, and enrichment of uranium ore, the activity of reactors of nuclear power plants and research centers, application of radionuclides in research institutes, hospitals and industry [1-8]. The RW contaminants are mainly  $\beta/\gamma$  emitters that occur in the form of activated corrosive products (<sup>60</sup>Co and <sup>54</sup>Mn) or fission products (<sup>137</sup>Cs and <sup>90</sup>Sr). Regarding radiation and toxic effects as well as abundances in waste streams, the most important RW pollutants for analysis and treatment are <sup>60</sup>Co and <sup>137</sup>Cs [2]. In order to immobilize the RW of the low and middle level of activity, the solidification of the concentrate is accomplished by binding to inactive matrices: usually cement, bitumen, polymeric materials, and rarely glass. Even though cement has several unfavorable characteristics as a solidifying material, e. g., low volume reduction and relatively high leachability, it possesses many

practical advantages: good mechanical characteristics, low cost, easy operation and radiation, and thermal stability.

The radionuclide leaching process from the cement matrix can be viewed successively through three phases [9]:

- phase transformation of the dissolution component from the cement matrix composition in the solvent,
- dissolved component diffusion of the matrix to its surface, and
- matrix dissolved component migration to the surrounding solvent mass

The rate of radionuclide release from the cement matrix will be determined by the slowest process.

The possibility of retaining radionuclides bound by immobilization processes in the cement matrix is defined as the subject of its investigation: the cement matrix formulation, the water/cement ratio, the amount of waste, and the porosity of such a structure, based on the tests determining the level of their deposition. Implementing the standard leaching testing method by Hesse [10] accepted by the International Atomic Energy Agency (IAEA), the possibility of comparing different authors' results was achieved.

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**MATHEMATICAL MODELING OF CS<sup>+</sup> AND CO<sup>2+</sup> LEACHING BEHAVIOR FROM CEMENT MATRIX**

**Diffusion model**

The diffusion model is used to investigate the transport phenomenon in order to predict the leaching level for a long period of time, and in the past period, several mathematical interpretations of this problem have been published [11-13]. Common to the more recently set problem interpretations of linking the radionuclides diffusion coefficients, from the composition of the cement matrix and their leaching rate from the matrix surface, are basically starting from the Fick's law of diffusion.

The assumptions and boundary conditions that we must implement for the diffusion model application are [14, 15]:

- the mobility of Cs<sup>+</sup> and Co<sup>2+</sup> ions is limited by diffusion,
- the sample is semi-infinite in the direction of the negative *x* axis, ensuring that the waste cumulative leaching fraction does not exceed 20 %,
- the concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions is uniformly distributed to the waste, and their quantity in the center of the matrix does not change during the leaching period, and
- the concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions at the boundary phase is zero.

The corresponding differential equation and the boundary conditions that define this law can be presented as

$$D_e \frac{\partial^2 C}{\partial x^2} = \frac{\partial C}{\partial t}, 0 \leq t < \infty, 0 \leq x < \infty \quad (1)$$

$$C(x,0) = C_0 \quad (2)$$

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

$$\frac{\partial C}{\partial x} = 0 \text{ for } x = \infty \quad (4)$$

where *C* [mgdm<sup>-3</sup>] is the concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions, *x* [cm] – the distance, *t* [s] – the time, and *D<sub>e</sub>* [cm<sup>2</sup>d<sup>-1</sup>] – the effective diffusion coefficient.

The solution of eq. (1) is

$$C(x,t) = C_0 \operatorname{erf} \left( \frac{x}{2\sqrt{D_e t}} \right) \quad (5)$$

where

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-u^2} du = \frac{2}{\sqrt{\pi}} \left( x - \frac{x^3}{3!} + \frac{x^5}{5!} - \frac{x^7}{7!} + \dots \right) \quad (6)$$

when *x* = 0, eq. (6) became

$$\operatorname{erf}(0) = \frac{2x}{\sqrt{\pi}} \quad (7)$$

and

$$C(x,t) = C_0 \frac{x}{\sqrt{\pi D_e t}} \quad (8)$$

Flux, *j(t)*, of Cs<sup>+</sup> and Co<sup>2+</sup> ions, dimension [mgcm<sup>2</sup>d<sup>-1</sup>], in the time unit through the sample contact surface *S* [cm<sup>2</sup>], for *x* = 0 is therefore

$$j(t)_{x=0} = D_e \frac{\partial C}{\partial x} = C_0 \sqrt{\frac{D_e}{\pi t}} \quad (9)$$

The total concentration of radionuclides accumulated in the leaching solution for each *N*<sup>th</sup> sampling period over the total duration of the experiment *T*(*d*) is

$$C_n = \int_0^T S j(t)_{x=0} dt = \frac{SD_e C_0}{\sqrt{\pi D_e}} \int_0^T \frac{dt}{\sqrt{t}} \quad (10)$$

$$C_n = \frac{2SC_0}{V} \sqrt{\frac{D_e T}{\pi}} \quad (11)$$

where *C<sub>0</sub>* [mg dm<sup>-3</sup>] is the initial concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions in matrix, *V* [cm<sup>3</sup>] – the volume of the sample exposed to distilled water, *S* [cm<sup>2</sup>] – the total surface area exposed to distilled water, and *C* [mg dm<sup>-3</sup>] – the concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions in the leaching solution according to its *N*<sup>th</sup> variation.

Using the experimentally obtained values for *C<sub>n</sub>* [mg dm<sup>-3</sup>] a graph can be created

$$\frac{C_n}{C_0} \frac{V}{S} = f(\sqrt{t})$$

If the experimental data are obeyed by linear dependence, it can be assumed that diffusion is the dominant mechanism in the process of separating the radionuclide from the cement matrix and from the slope of the right, *m*

$$m = 2\sqrt{\frac{D_e}{\pi}} \quad (12)$$

the effective diffusion coefficient, *D<sub>e</sub>* [cm<sup>2</sup>d<sup>-1</sup>], can be calculated using the term

$$D_e = \frac{m^2}{2} \pi \quad (13)$$

**Semi-empirical model**

Using the leaching cumulative fraction as a function of time, we distinguish three different mechanisms: surface wash off, dissolution and diffusion [14].

A mathematical model involving all three leaching mechanisms is very complex, and the obtained results have very limited practical application.

In order to overcome this problem, the semi-empirical model is adopted, based on an orthogonal-polynomial equation, which describes the leaching of waste constituents immobilized in a solid matrix over a long period of time [16]. The leaching cumulative fraction is expressed according to this model

$$\frac{C_n}{C_0} = K_1 + K_2 t^{1/2} + K_3 t \quad (14)$$

where  $K_1$  is the constant representing the contribution of surface wash-out to radionuclide transport,  $K_2$  – the constant representing the contribution of diffusion to the radionuclides transport ( $s^{-1/2}$ ),  $K_3$  – the constant representing the contribution of a kinetically controlled solution to the radionuclides transport ( $s^{-1}$ ), and  $t$  [s] – the duration of the experiment.

## EXPERIMENTAL PART

### Preparation of cement matrix

The defined composition of the cement matrix, as well as the initial Cs<sup>+</sup> and Co<sup>2+</sup> ions concentration,  $C_0$  [mg dm<sup>-3</sup>], are listed in tab. 1. Materials of the cement matrix are:

- Portland cement, PC-35 (MPa), manufacturer Lafarge,
- Sand- “Moravac”, granulate 2 mm,
- Distilled water, electrical conductivity  $G = 1.5 \cdot 10^6 \text{ cm}^{-1}$  and pH = 6.2, and
- Sludge, density  $\rho_{\text{sludge}} = 1.075 \text{ gcm}^{-3}$

The materials were treated in a planetary mixer to achieve a degree of viscosity that ensures the transfer of the cement matrix to the prepared molds. Using the mixer with a planetary mixing method, cement matrixes of high structure homogeneity are obtained. The duration of mixing after the batch is limited to 15 minutes, usually 5 minutes after achieving the required degree of viscosity. The rotational speed of the mixer is limited to  $n_1 = 75 \text{ rpm}$  while the batch counter to  $n_2 = 35 \text{ rpm}$ .

### Investigation of leaching in dynamic conditions

Leaching investigation of Co<sup>2+</sup> and Cs<sup>+</sup> ions, immobilized in the cement matrix M1, M2, and M3 in dynamic conditions, was adapted to the standard ANS test 16.1 [17]. The shape and dimensions of the test sample, as well as the shape and dimensions of the vessels, the experimental temperature, are identical to the standard method by Hespe [10]. The sample was completely immersed in 0.5 liters of distilled water while

**Table 1. The initial concentration of Cs<sup>+</sup> and Co<sup>2+</sup> ions and the cement matrix composition for the production of the samples  $V = 1 \text{ dm}^3$**

Composition	CEMENT MATRIX		
	M1	M2	M3
Cement [g]	933	1000	1000
Sludge [g]	280	350	500
Sand [g]	747	700	600
H <sub>2</sub> O [ml]	93	40	0
Cs <sup>+</sup> [mgdm <sup>-3</sup> ]	457	572	817
Co <sup>2+</sup> [mgdm <sup>-3</sup> ]	819	1024	1463

the volume flow of the distilled water circulating around the sample was  $200 \text{ l day}^{-1}$ . The leaching solution is exchanged after 1<sup>st</sup>, 2<sup>nd</sup>, 3<sup>rd</sup>, 4<sup>th</sup>, 5<sup>th</sup>, 6<sup>th</sup>, 7<sup>th</sup>, 8<sup>th</sup>, 10<sup>th</sup>, 12<sup>th</sup>, 14<sup>th</sup>, 16<sup>th</sup>, 18<sup>th</sup>, 21<sup>st</sup>, 23<sup>rd</sup>, 25<sup>th</sup>, 28<sup>th</sup> day and any change in the leaching solution is an incremental contribution to the total concentration of ions release. The concentration of Co<sup>2+</sup> and Cs<sup>+</sup> metal ions was measured by the method of atomic absorption spectrometry using Perkin Elmer Analyst 200.

During the experiment, attention was paid to the reproducibility of the obtained results. The combined measurement uncertainty of the experimental procedure was expressed by a statistical analytical procedure and was less than 5 % [18-20].

## RESULTS AND DISCUSSION

Leaching of Co<sup>2+</sup> and Cs<sup>+</sup> ions immobilized in the cement matrix, M1, M2, M3 in the dynamic conditions decreases in the following series  $M3 > M2 > M1$ . The leaching increases with the increase of the sludge content, regarding porosity increase, figs. 1 and 2.

Leaching level of Co<sup>2+</sup> ions is more than two orders of magnitude smaller than leaching level of Cs<sup>+</sup> [21-25]. Due to the high alkalinity of the cement matrix (pH > 10), the Co<sup>2+</sup> ions react with the hydroxide ions, creating a cobalt-hydroxide which can formerly react with CO<sub>2</sub> and form insoluble cobalt-carbonate [24]. Cobalt can also be exchanged with calcium cations that are present in the C-S-H gel by forming analogous cobalt complex salts which, by crystallizing, become part of the cement matrix structure.

Conversely, cesium ions retention in the cement matrix is poor [21-24]. Cesium is an element of a large atomic radius and a weak electric field. Therefore, it does not hydrolyze in solution and is present in the free cationic form at all pH values. Moreover, precipitating cesium ions are rarely constituent of the complex salts that are retained in the matrix structure.

In the dynamic performance of the experiment, fluid circulation around a sample of the cement matrix is carried out with its constant depth depletion for the observed ion, thereby establishing a higher concentration gradient than in the static performance of the experiment [26]. Consequently, higher values of the effective diffusivity coefficients,  $D_e$ , as well as the higher values of the constants  $K_1$  and  $K_2$  in the semi-empirical leaching model are established.

The destructive action of the fluid circulation around the cement matrix sample leads to its dissolution. This phenomenon is confirmed by the semi-empirical model, where the dissolution constants  $K_3$  absolute values are significantly higher and much less distinctive than the diffusion constants  $K_2$  values in the static conditions.

Generally, in the dynamic conditions, the effect of diffusion and surface flushing is balanced, and the

contribution of matrix dissolution to the ions transport in the cement porous medium increases on average for one order of magnitude. In consideration of all this, it becomes clear why the leaching level of Co<sup>2+</sup> and Cs<sup>+</sup> ions in dynamic is incomparably greater than that in static conditions. [26-28]. As expected, the leaching for Cs<sup>+</sup> is almost three orders of magnitude greater than for Co<sup>2+</sup>.

Although diffusion model cannot describe the entire leaching process, it is very suitable for simulation due to its simplicity [29]. Based on the value of the regression coefficient, the semi-empirical model gives a better approximation for Co<sup>2+</sup> and Cs<sup>+</sup> leaching process for the duration of the experiment [29, 30]. Furthermore, both models significantly approximate leaching results in dynamic conditions.

In tab. 2 the effective diffusion coefficients values,  $D_e$ , and the regression coefficients,  $R$ , are presented, calculated by leaching diffusion model of Cs<sup>+</sup> and Co<sup>2+</sup> ions immobilized in the cement matrices M1, M2, and M3 under dynamic conditions.

In tab. 3 the constants  $K_1$ ,  $K_2$ ,  $K_3$  values and the regression coefficients,  $R$ , are presented calculated by semi-empirical leaching model of Cs<sup>+</sup> and Co<sup>2+</sup> ions immobilized in cement matrices M1, M2, and M3 under dynamic conditions.

**CONCLUSION**

Leaching of Co<sup>2+</sup> and Cs<sup>+</sup> ions immobilized in the cement matrix, M1, M2, M3 in dynamic conditions decreases in the following series M3 > M2 > M1, with the increase of the sludge content, regarding porosity increase. Cs<sup>+</sup> and Co<sup>2+</sup> leaching level in dynamic is three and two orders of magnitude greater than in static conditions, respectively.

In dynamic conditions, the effects of the diffusion and surface washing are equalized, and the contribution of the matrix dissolution to the Cs<sup>+</sup> and Co<sup>2+</sup> transport in the cement porous media increases, on average, for one order of magnitude.

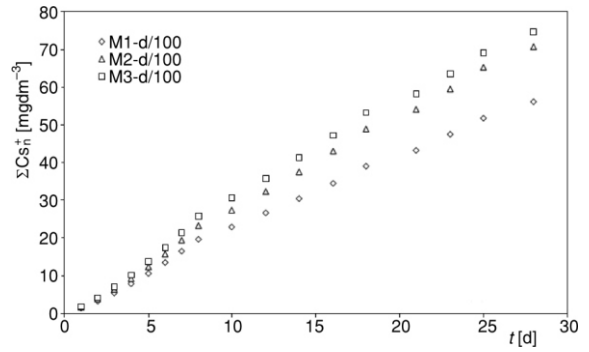
The semi-empirical model gives a better approximation for Co<sup>2+</sup> and Cs<sup>+</sup> leaching process for the duration of the experiment while both models significantly approximate leaching results in dynamic conditions.

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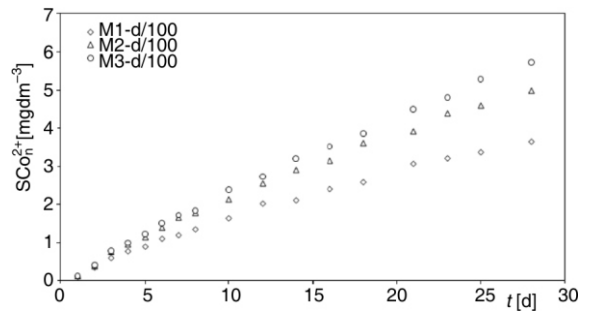
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**AUTHORS' CONTRIBUTIONS**

The experiments were carried out by S. D. Dimović. All authors analyzed results and participated



**Figure 1. Leaching of Cs<sup>+</sup> ions bound in 100 g of sludge, immobilized in the cement matrix M1, M2, and M3 under dynamic conditions**



**Figure 2. Leaching of Co<sup>2+</sup> ions bound in 100 g of sludge, immobilized in the cement matrix M1, M2, and M3 under dynamic conditions**

**Table 2. Effective diffusion coefficients values,  $D_e$  [ $\text{cm}^2\text{d}^{-1}$ ], and regression coefficients,  $R$ , calculated by leaching diffusion model of Cs<sup>+</sup> and Co<sup>2+</sup> ions immobilized in the cement matrices M1, M2, and M3 under dynamic conditions**

Diffusion molding parameters	CEMENT MATRICES		
	M1	M2	M3
$D_e(\text{Cs}^+)$	$2.94 \cdot 10^{-3}$	$4.74 \cdot 10^{-3}$	$5.36 \cdot 10^{-3}$
$D_e(\text{Co}^{2+})$	$3.63 \cdot 10^{-6}$	$6.78 \cdot 10^{-6}$	$8.81 \cdot 10^{-6}$
$R(\text{Cs}^+)$	0.993	0.990	0.992
$R(\text{Co}^{2+})$	0.996	0.995	0.993

**Table 3. Constants  $K_1$ ,  $K_2$  [ $\text{s}^{-1/2}$ ],  $K_3$  [ $\text{s}^{-1}$ ] vs. and regression coefficients,  $R$ , calculated by semi-empirical leaching model of Cs<sup>+</sup> and Co<sup>2+</sup> ions immobilized in cement matrices M1, M2, and M3 under dynamic conditions**

Parameters of semi-empirical model leaching	CEMENT MATRICES		
	M1	M2	M3
$K_1(\text{Cs}^+)$	$-3.70 \cdot 10^{-2}$	$-3.80 \cdot 10^{-2}$	$-5.57 \cdot 10^{-2}$
$K_1(\text{Co}^{2+})$	$-1.42 \cdot 10^{-3}$	$-1.89 \cdot 10^{-3}$	$-1.76 \cdot 10^{-3}$
$K_2(\text{Cs}^+)$	$2.97 \cdot 10^{-2}$	$2.70 \cdot 10^{-2}$	$4.28 \cdot 10^{-2}$
$K_2(\text{Co}^{2+})$	$1.59 \cdot 10^{-3}$	$1.94 \cdot 10^{-3}$	$1.61 \cdot 10^{-3}$
$K_3(\text{Cs}^+)$	$8.09 \cdot 10^{-3}$	$1.19 \cdot 10^{-2}$	$1.05 \cdot 10^{-2}$
$K_3(\text{Co}^{2+})$	$2.07 \cdot 10^{-4}$	$3.21 \cdot 10^{-4}$	$4.64 \cdot 10^{-4}$
$R(\text{Cs}^+)$	0.999	0.999	0.999
$R(\text{Co}^{2+})$	0.999	0.999	0.999

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### **КИНЕТИКА ИЗЛУЖИВАЊА Cs<sup>+</sup> И Co<sup>2+</sup> У ДИНАМИЧКИМ УСЛОВИМА**

Могућност задржавања Cs<sup>+</sup> и Co<sup>2+</sup> процесима имобилизација у цементном матриксу дефинише се преко формулације цементог матрикса, односа вода/цемент, количина отпада и порозности такве структуре. Уз помоћ имплементације стандардног метода излуживања према Хеспелу постигнута је могућност упоређивања резултата различитих аутора. За истраживање феномена транспорта коришћен је дифузиони и полуемпиријски модел како би се предвидео ниво излуживања за дужи временски период. Излуживање имобилисаих јона Cs<sup>+</sup> и Co<sup>2+</sup> у цементном матриксу у динамичким условима расте са повећањем садржаја муља и порозности. Ефекти дифузије и површинског спирања су изједначени, а допринос растварања матрикса при транспорту Cs<sup>+</sup> и Co<sup>2+</sup> повећава се, у просеку, за један ред величине. Полуемпиријски модел даје бољу апроксимацију процеса излуживања Cs<sup>+</sup> и Co<sup>2+</sup> током трајања експеримента, док оба модела добро апроксимирају резултате излуживања у динамичким условима.

*Кључне речи: радиоактивни отпад, цементна матрица, спирање, математичко моделовање*

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