Abstract – To assess the safety for disposal of radioactive mortar-waste composition, the leaching of $^{137}$Cs from a waste composite into a surrounding fluid has been studied. Leaching tests were carried out in accordance with a method recommended by IAEA. Determination of retardation factors, $K_F$ and coefficients of distribution, $k_d$, using a simplified mathematical model for analyzing the migration of radionuclides, has been developed. In our experiment we have achieved the lowest leaching values after 60 days in samples. Results presented in this paper are examples of results obtained in a 20 year mortar and concrete testing project, which will influence the design of the engineered trenches system for a future central Serbian radioactive waste storage center.

1. INTRODUCTION

In order to prevent widespread dispersion of radionuclides into the human environment, radioactive waste produced in nuclear facilities has been incorporated in several kinds of matrices. The objectives of immobilization of radioactive waste is to convert the waste into forms which are: Leach resistant so that the release of radionuclides will be slow even though they may come into contact with flowing water; and mechanically, physically and chemically stable for handling transport and disposal.

Cement and concrete are widely used in low-level waste management both as a means of solidifying waste and for containment of dry or liquid wastes. At present there is also widespread interest in the use of near-surface concrete trench system for the disposal of radwaste materials. Typical mortar is a mixture of cement, sand and water in various proportions, that together determine the structural properties and tightness of the poured material. Water content is one of the critical parameters and must be carefully controlled during pouring and setting; to a large extent it will determine the porosity of the resulting material. Cement is porous, continuously hydrating material whose actual surface area greatly exceeds its geometric surface area. In leaching, the rate of dissolution varies as a function of phase chemistry and this dissolution exposes or enlarges pores; thus the leaching behavior must be related to pore structure and the composition of the pore solution.[1,2,3,4]

2. RADIONUCLIDE MIGRATION THROUGH POROUS MATERIALS

The dispersion of radionuclides in porous materials, such as grout or concrete, is described using a one dimensional dimodel.[2,4,5,6,7]

\[
D \frac{\partial^2 A}{\partial X^2} - V_Y \frac{\partial A}{\partial X} \left(1 + \frac{1-f}{f \rho_T k_D} D \right) \frac{\partial A}{\partial t} = 0
\]  

or

\[
D \frac{\partial^2 A}{\partial X^2} - V_Y \frac{\partial A}{\partial X} - K_F \frac{\partial A}{\partial t} = 0
\]

(1)

where:

$K_F$ - retardation factor (= 1)

D - diffusion coefficient (cm$^2$/d) or (cm$^2$/s)

A - concentration in liquid (mol/l) or (Bq/l)

X - length (cm)

$V_Y$ - velocity of leachant fluid (cm/d)

f - porosity (= 1)

$\rho_T$ - bulk density (g/cm$^3$)

$k_d$ - distribution coefficient (ml/g)

t - time variable (d).

Using Laplace transformation method, Eq.(1) becomes:

\[
\frac{A_R}{A_0} = \frac{1}{2} \text{erf} z \left| \frac{\frac{V_Y X}{4 D_{cf}}}{\sqrt{\frac{V_Y t}{K_F X}}} \right|
\]

(2)

from which we can calculate a retardation factor, $K_F$. The coefficient of distribution, $k_d$, can be calculated:

\[
k_d = \frac{(K_F - 1)f}{(1-f) \rho_T} (\text{ml/g})
\]

(3)

in which: $V_Y$, X, $\rho_T$, t and $A_0$ are known. $A_n$ and $D_c$ can be determined experimentally using a leaching test procedure [3].

3. DETERMINING THE EFFECTIVE COEFFICIENTS OF DIFFUSION

For the interpretation of the results of leach tests shown in the following tables, leach coefficient D, is used, and it is defined as:

\[
D = \frac{\pi}{4} m^2 \frac{V^2}{S^2} (\text{cm}^2/\text{d})
\]

(4)

where:

D - leach coefficient (diffusion) (cm$^2$/d) or (cm$^2$/s); $m - (\Sigma A_0/A_0) / (1/\sqrt{\Sigma t})$, slope of the straight line (d$^{-1/2}$); $A_n$ - initial sample activity at time zero (Bq); $A_c$ - activity leached out of sample after leaching time t, (Bq); t - duration of leaching renewal period (d); (1,2,3,4,5,6,7,15,30,60)
V - sample volume (cm$^3$);
S - sample surface (cm$^2$).

4. MATERIALS AND CONCRETE COMPOSITION

Concrete samples were made of:
- Portland cement PC-20-Z-45 MPa;
- Sand, fraction 0-2 mm;
- Water;
- Additive, Superfluidal;

More than 100 different formulations of mortar formulations were examined to optimize their mechanical and sorption properties. In this paper we discuss four representative formulations. Mortar composition are shown in Table I.

Table I Mortar compositions (calculated as grams for 1000 cm$^3$ of concrete)

<table>
<thead>
<tr>
<th>Formula</th>
<th>M$_1$</th>
<th>M$_2$</th>
<th>M$_3$</th>
<th>M$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement (Portland), gr</td>
<td>750</td>
<td>700</td>
<td>650</td>
<td>600</td>
</tr>
<tr>
<td>Sand, 0-2 mm</td>
<td>1250</td>
<td>1310</td>
<td>1325</td>
<td>1375</td>
</tr>
<tr>
<td>Water, ml</td>
<td>250</td>
<td>235</td>
<td>230</td>
<td>225</td>
</tr>
<tr>
<td>Additives, ml</td>
<td>12</td>
<td>13</td>
<td>14</td>
<td>15</td>
</tr>
<tr>
<td>Initial activity, A$_0$(kBq)</td>
<td>50,0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

5. RESULTS

The results are obtained after 60 days. Using equation (4), coefficients of diffusion are calculated for four experimental samples.

Using equation (2) and (3), retardation factors, K$_F$(=1), and distribution coefficients, k$_d$(ml/g) are calculated. Table II gives $^{137}$Cs, leach coefficients in different mortar samples.

Table II Leach coefficients $D_c$(cm$^2$/s) in different mortar samples after 60 days, using Eq.(4)

<table>
<thead>
<tr>
<th>Formula</th>
<th>M$_1$</th>
<th>M$_2$</th>
<th>M$_3$</th>
<th>M$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Leach coeff. $D_c$$^{137}$Cs</td>
<td>$1.45 \times 10^{-13}$</td>
<td>$3.44 \times 10^{-13}$</td>
<td>$6.34 \times 10^{-13}$</td>
<td>$7.78 \times 10^{-13}$</td>
</tr>
</tbody>
</table>

6. CONCLUSION

The analysis of the results presented in Table II and Table III shews that the values of retardation factors and coefficients of radionuclides $^{137}$Cs, are similar to the literature data, and prove that the one-dimensional model can be used for calculating parameters of the migration process. The main conclusion from this experiment is that biggest quantity of water gives higer Retardation factors, K$_F$ and determine the porosity in samples, and the diffusion coefficient are the critical and major contribution parameters compared to any other mechanisms in analyzing the mortar samples.

REFERENCES


