# Co-60 LEACHING FROM VARIOUS FORMULATIONS OF CONCRETE IN SHALLOW LAND DISPOSAL SYSTEM

I. B. Plećaš, J. D. Drljača,
A. M. Kostadinović, A. D. Perić
Boris Kindrič Institute of Nuclear Sciences-Vinča, Belgrade
Yugoslavia

Lj. Gradišar
Nuclear Power Plant Krško
Krško, Yugoslavia

## ABSTRACT

Determination of retardation factors and coefficients of distribution using simplified mathematical model for analysing the migration of leachate and radioactive material contained in radioactive waste burial concrete trenches system, has been developed.

Results show that engineer trench system secure radionuclide preservation in solidified medium much longer than 300 years. These results will be used for future Yugoslav radioactive waste storing centers.

## INTRODUCTION

In coming decades, nuclear energy will have a prominent position in energy development programs of many states, including Yugoslavia. Radioactive waste is an unavoidable by-product in nuclear energy production. After volume reduction and valuable components recovery, waste materials have to be conditioned for transport, storage and disposal. Conditioning is the waste management step in which radioactive wastes are immobilized and packed (1).

The objectives of immobilization are to convert the waste into forms which are:

* Leach resistant so that the release of radionuclides will be slow even though flowing water may contact them,

* Mechanically physically and chemically stable for handling, transport and disposal.

The immobilization processes involve conversion of the waste to solid forms that reduce the potential for migration or dispersion of radionuclides from the wastes by natural processes during storage, transport and disposal. The immobilization processes involve the use of various matrices of non-radioactive materials such as cement, bitumen and polymers, to fix the wastes as monoliths, usually directly in the waste containers used for subsequent handling (2).

Primary barriers for confinement and limiting the release of radionuclides are the following material properties:

* low solubility,
* mechanical strength,
* low permeability,
* compatibility with the waste,
* stability during storage,
* resistance to heat and radiation,
* resistance to external agents.

Concrete solidification process still is the most widespread process for conversion of low and intermediate level radioactive waste into concrete forms for final storing in predetermined locations.

The main reasons for using cement are:

* high mechanical strength of cement products,
* relative simplicity of handling,
* large experience in civil engineering,
* availability of raw material,
* relatively low cost,
* high density (shielding effect),
* compatibility of water with the matrix material.

## SHALLOW LAND DISPOSAL SYSTEM

According to safety regulations, intermediate level wastes must be put in concrete arrangements. To meet this specification, trenches are dug below the ground level above the water-table, a concrete pad is made and constitutes the bottom of the pill. Then the waste containers are placed in the trench. After each level in the trench is filled, it is backfilled with concrete. When the trench is full and reaches the ground level, the top is carefully sealed and covered with a layer of bitumen making a tight pad above which a tumulus will be built.

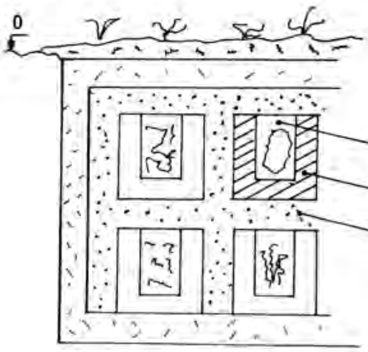
Engineering trenches system provides three biological protection barriers (Fig. 1).

These three kinds of concrete, which have totally different composition and function, make the whole technological unit.

### Mortar for Container Filling

Mortar for container filling has to:

- enable its penetration into cavities of solid radioactive waste and thus fix it permanently,
- permit no leakage of radionuclides (use of special cement with dross and pozzolane which have excellent absorption properties),
- provide primary biological protection.




1. Mortar for immobilizing the waste and filling the concrete containers.
2. Concrete container.
3. Concrete for filling trenches.

Fig. 1. Scheme of Engineering Trenches System.

Concrete Container

The function of the concrete container is:

- to store and enable transport of low and intermediate level radioactive waste,
- to provide secondary biological protection,
- to provide safe keeping of radioactive waste for 300 to 500 years.

Concrete for Filling Trenches

Concrete for filling trenches has to:

- provide tertiary biological protection,
- provent leakage of radionucledes which have penetrated the second barrier.

The concrete container, being the most important element of the system, has a very specific function as well as material choice. Granulometric composition calculation and rigorous control of its main physicomechanical characteristics, such as:

1. compresive strength
2. permeability
3. leakage test

are exceptionally important (3).

RADIONUCLIDE MIGRATION THROUGH POROUS MATERIALS

The dispersion of radionuclides in porous materials, such as concrete, is described using one-dimensional differential model (4,5).

$$D\frac{\partial^2 C}{\partial X^2} - V_v\frac{\partial C}{\partial X} - \left(1+\frac{1-f}{f}\cdot\rho_T\cdot k_d\right)\frac{\partial C}{\partial t} = 0 \qquad (1)$$

or

$$D\frac{\partial^2 C}{\partial X^2} - V_v\frac{\partial C}{\partial X} - K_f\frac{\partial C}{\partial t} = 0 \qquad (1a)$$

where are:

$K_f$ - retardation factor (=)1

$$K_f = \frac{\text{velocity of radionuclides}}{\text{velocity of conveying fluid}}$$

$D$ - diffusion coefficient ($cm^2/s$)
$C$ - concentration in liquid (mol/l)
$X$ - length (cm)
$V_v$ - velocity of conveying fluid(cm/d)
$f$ - porosity (=)1
$\rho_T$ - bulk density ($gr/cm^3$)
$k_d$ - distribution coefficient (ml/gr)

$$k_d = \frac{\dfrac{\text{quantity of adsorbed material}}{\text{mass of soil}}}{\dfrac{\text{quantity of dissolved material}}{\text{volume of water}}}$$

$t$ - time variable (d)

Using Laplass transformation method Eq.(1) becomes:

$$\frac{C_n}{C_o} = \frac{1}{2}\,\mathrm{erf}\,z\left[\sqrt{\frac{V_v\cdot X}{4\cdot D_e}}\cdot\frac{1-\dfrac{V_v\cdot t}{K_f\cdot X}}{\sqrt{\dfrac{V_v\cdot t}{K_f\cdot X}}}\right] \qquad (2)$$

wherefrom we can calculate retardation factor $K_f$, that is coefficient of distribution $k_d$:

$$k_d = \frac{(K_f-1)\cdot f}{(1-f)\cdot\rho_T}\ (=)\ \text{ml/gr} \qquad (3)$$

in which are known: $V_v, X, \rho_T, t$ and $C_o$. $C_n$ and $D_e$ can be determined experimentaly.

Determining the Effective Coefficients of Diffusion

Leaching experiments worked out by Hespe (6) with many types of solidified wastes including cement, containing waste solid, have shown that elution of radioactive ions often follows Fick's diffusion equation.

The proposed test consists of exposing one face of a cylindrical test specimen of immobilized waste to an aqueous solvent. It is assumed that the rate of loss of radionuclides to the solvent can be described by Fick's first and second laws, and that the diffusion process is characterized by: diffusion coefficient (or leaching factor), D, ($cm^2/s$) which is independent of time, position and concentration of the diffusing species.

When this is true a plot of versus is a straight line and for the case of a semi-infinite slab the diffusion coefficient D is expressed by:

where m is the slope of the straight line from a plot of versus (Fig. 2). Symbols used in Eq. (4) and on (Fig. 2) are:

V - volume of specimen ($cm^3$)
F - exposed surface area of specimen ($cm^2$)

$t_n$ - duration of leachant renewal period (d)
$C_n$ - radioactivity lost during leaching period (Bq)
$C_0$ - initial radioactivity (Bq)

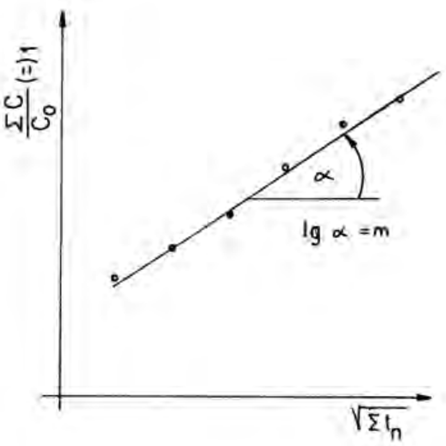



Fig. 2. Cumulative Fraction Leached Ploted Against the Square Root of Time.

EXPERIMENTAL EQUIPMENT FOR LEAKAGE RATE MEASUREMENT

Apparatus for concrete leakage test has been constructed in Institute of Nuclear Science "Boris Kidrič" - Vinča. This original method provides a virtual image about the ability of concrete to prevent "washing out" of solidified waste materials by underground flows. There is no suggested IAEA standard procedure worded out by E. Hespe (6).

The apparatus made possible simulation of real process with concrete disks (diameter 10 cm, height 1 cm), (Fig. 3). Leakage test results enable time calculation with a great degree of approximation after which radionuclide washing out from real system can be expected (6,7).

Materials and Concrete Compositon for Containers

Concrete samples have been made of:

- concrete PC-55 MPa (manufactured by Beočin Cement Mill)
- sand and granulate "Moravac" fraction 0-2 mm; 4-8 mm; 8-15 mm (manufactured by "Standard Beton", Belgrade)
- water attested according to Yugoslav standard
- additives: Super fluidal (manufactured by "Chromos", Zagreb)
- 55,5 MBq $Co^{60}$/per apparatus ($C_0$).

Ten formulas of concrete have been mixed for every combination of cement and additive, according to civil engineering regulations on "Granulometric composition of aggregate mixture". All formulas for container have been developed after extensive research and conforming IAEA rules.

Concrete compositon formulas are shown in Table I.

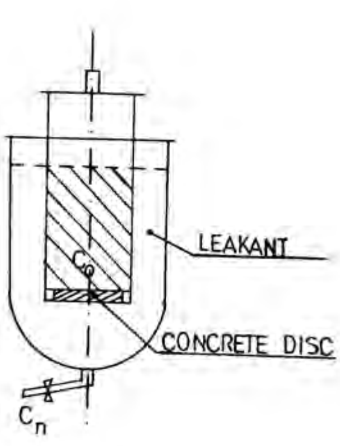



Fig. 3. Scheme of the Experimental Equipment for Leakage Rate Measurement.

TABLE I

Container Concrete Compositions (calculated as grams for 1000 $cm^3$)

| Formula No. | $K_3$ | $K_5$ | $K_9$ |
|---|---|---|---|
| Cement (gr) | 400 | 400 | 400 |
| Sand 0-2 mm | 625 | 692 | 750 |
| Granulate 2-4 mm | 69 | 75 | 83 |
| Granulate 4-8 mm | 437 | 423 | 417 |
| Granulate 8-15 mm | 853 | 794 | 734 |
| Water (ml) | 140 | 140 | 140 |
| Additive (ml) | 8 | 8 | 8 |

RESULTS

Data for three concrete compositions are plotted in Figs. 4,5, and 6.

The results are obtained after 120 days. During the measuring of the leaching factor, the pH values were controlled and measured, after every renewal period. The pH (values) were always varying from 7,5 - 8,5.

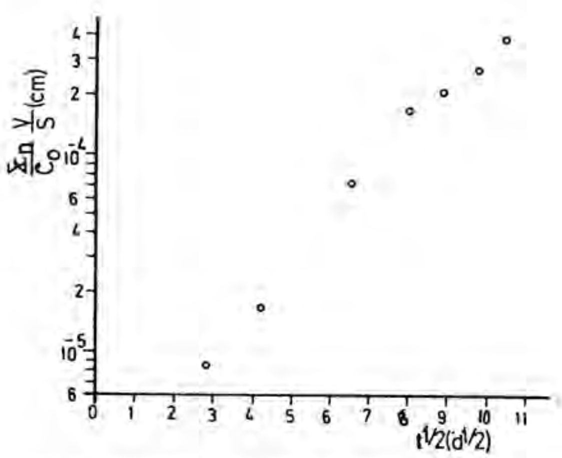



Fig. 4. Cumulative Fraction Leached Plotted Against Time, for Concrete $K_3$ After 120 days.

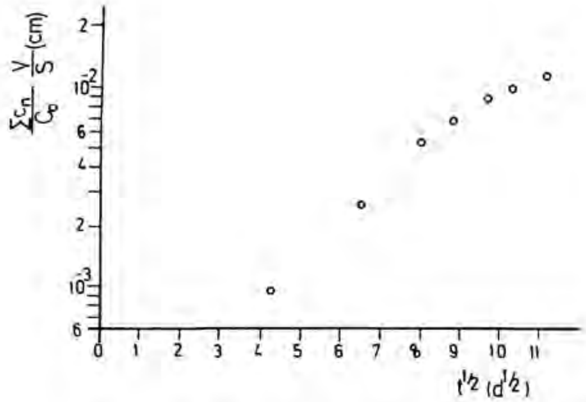



Fig. 5. Cumulative Fraction Leached Plotted Against Time, for Concrete $K_5$ After 120 Days.

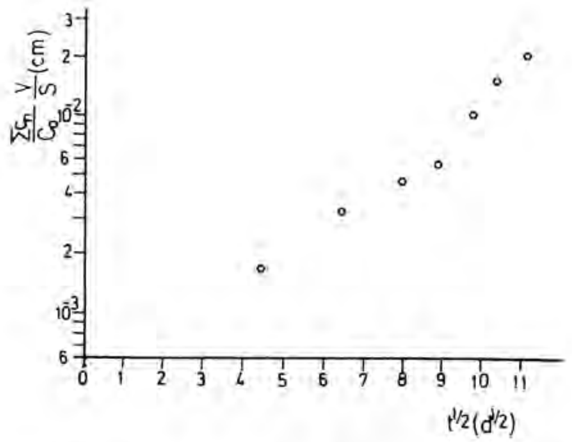



Fig. 6. Cumulative Fraction Leached Plots Against Time, for Concrete $K_9$ After 120 Days.

In tables II, III and IV are shown calculated coefficients of diffusion, retardation factors and coefficients of distribution.

TABLE II

Using Eq. (4), Coefficients of Diffusion are Calculated for Three Experimental Concretes

| Formulation | Slope $m(cm\cdot d^{-1})$ | $D_e(cm^2/s)$ |
|---|---|---|
| $K_3$ | $4{,}75\cdot10^{-5}$ | $2{,}04\cdot10^{-14}$ |
| $K_5$ | $1{,}35\cdot10^{-3}$ | $1{,}67\cdot10^{-11}$ |
| $K_9$ | $2{,}15\cdot10^{-3}$ | $4{,}22\cdot10^{-11}$ |

## CONCLUSION

Analysing results presented in Tables II-IV it is noticed that calculated values of retardation factors of radionuclides, $K_f$, and coefficients, of distribution, $k_d$, are similar to the literature data (7), what proves that one-dimensional model Eq. (1) can be used for calculating of parameters of migration process. That also proves that the system of engineering trenches permits secure preservation of radionuclides for more than 300 years.

TABLE III

Retardation Factors $K_f$(=)1 are Calculated From Eq. (2) and Data From Table II, for Three Experimental Concretes

| Formulation | $K_f$(=) 1 |
|---|---|
| $K_3$ | 46,4 |
| $K_5$ | 42,2 |
| $K_9$ | 41,0 |

TABLE IV

Effective Distribution Coefficients $k_d$(ml/gr) are Calculated From Eq. (3) and Porosity f = 0,10 - 0,25 For Three Experimental Concretes

| Formulation | $k_d$(ml/gr) |
|---|---|
| $K_3$ | 2 - 6 |
| $K_5$ | 2 - 5 |
| $K_9$ | 2 - 5 |


## ACKNOWLEDGEMENT

The authors are gratefull to the staff of Nuclear Power Plant "KRsKO" for constructive help.


## REFERENCES


1. I. Plećaš, "Comparative Study of French and Yugoslav Radioactive Waste Forms", Proc. Waste Management, Tucson, Arizona, Feb. 27 - March 3, 1983, Vol. 2, p. 186 (1983).

2. I. Plećaš, "Some Important Characteristics of Concrete Containers in Radioactive Waste Package", Proc. XI Regional Conf. of IRPA, Vienna, 1983, Vol. 2, p. 206 (1983).

3. I. Plećaš, "optimization of Concrete Containers Composition in Radioactive Waste Technology", RWMADW, 6(2), 161 (1985).

4. I. Hashimoto, "Peclet Numbers and Retardation Factors for Ion Exchange Columns", IEC Fundamentals, 3, 213 (1964).

5. A. H. Lu, "Modeling of Radionuclide Migration from a Low-Level Radioactive Waste Burial Site", Health Physics, 34, 39 (1978).

6. E. D. Hespe, "Leach Testing of Immobilized Radioactive Waste Solids", Atomic Energy Rev., 9, 195 (1971).

7. E. Peltonen, S. Vuori, M. Katka, "The Role of Waste Conditioning as Regards the Safety of Low and Intermediate-Level Waste Disposal" Proc. Int. Conf. of Atomic Energy, 10, 419 (1983).