# PROPOSED PROGRAM AND OBJECTIVES FOR SHALLOW LAND DISPOSAL SYSTEM OF LOW - LEVEL RADIOACTIVE WASTE IN YUGOSLAVIA

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## ABSTRACT

Results presented in this paper represent the nowadays status of the radioactive waste management, especially quality testing methods, which are in common with radwaste solidification processes, performing in Institute of Nuclear Sciences - Vina in Belgrade.

Incorporation of the low and intermediate level wastes into cement is the most frequently applied technique worldwide. Although cement has several unfavorable characteristics as a solidifying material, it also possesses many practical advantages such as good mechanical characteristics, low cost, easy of operation, and radiation and thermal stability.

Results presented in this report represent the part of the most important findings drawn from a 10 years mortar and concrete testing project, and it's radwaste mixtures investigations. The data are intended for use during the design process of proposed central radioactive waste repository in Yugoslavia.

## INTRODUCTION

The continuation of activities in the Yugoslav atomic energy program requires the solution of problems related to disposal of radioactive wastes from different sources, such as Institute of Nuclear Sciences Vinca.

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 (1,2). The objectives of immobilization are to convert the wastes 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 interim storage up to the time of actual disposal (3,4).

The immobilization processes involve conversion of the wastes 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 whole radioactive waste, of low and intermediate level activity in Yugoslavia, is stored in the area of the Institute of Nuclear Sciences - Vinca and controlled by Radiation Protection Department.

Solid radioactive waste is conditioned in 6000 metal barrels (of 200 l each) and stocked in two 600 $m^2$ huge, hangers. Liquid radioactive waste of low activity produced while working on Experimental Reactor is stored in three underground tanks (350 $m^3$ each). That liquid radioactive waste is waiting to be treated in plant that is being built nowadays. According to the fact that location for conditioning radioactive waste is temporary, project team was set up to select and characterize the site, prepare a design, and build a low - and intermediate - level waste repository.

### Shallow Land Disposal System

The yugoslav concept of final disposal of LILW in shallow ground has been adopted, taking into account the following factors:

- Experience in countries with developed nuclear programs,
- Experience in developing countries, and
- Technical and economic aspects of the back and of the nuclear fuel cycle.

In accordance with the above factors, simple technology for LILW disposal have been proposed:

- "Radioactive waste, properly conditioned and packed in concrete containers, is placed on a concrete pad in a trench, near the surface or on it, provided with a drainage system and hydroinsulation. The space is filled with gravel, sand and clay" (5,6).

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, and concrete pad, that constitutes the bottom of the pill is made. Then the waste containers are placed in the trench (after filling the each level in the trench, 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 (4). Engineering trenches system provides three protection barriers of the environment:

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

These three kinds of mortar and concrete, which have totally different composition and function, make the whole technological unit presented on Fig. 1.

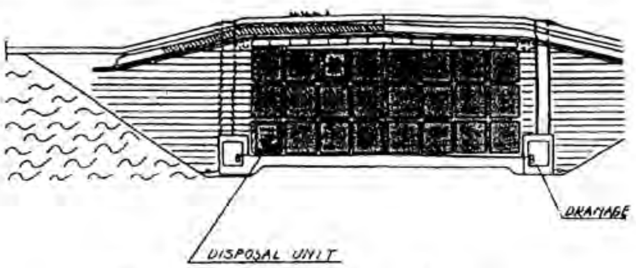



Fig. 1. Shallow land disposal system.

### Materials Used in the Solidification Process of Radwaste Materials

Concrete and mortar samples have been made of:

- two kinds of Portland cements, PC 45 MPa and PC 55 MPa,
- sand and granulate fractions 0-2 mm; 2-4 mm; 4-8 mm; 8-15 mm,
- water attested according to Yugoslav standard,
- additives: fluidal VX-OC, Super fluidal,
- bentonite clay.

### Quality Testing Methods Used in Radioactive Waste Management

The testing method, which are applied in cement and its radwaste mixtures properties examination, are:

- Testing of mechanical characteristics,
- Permeability measurement,
- Leakage test,
- Radionuclide migration in static semi-real conditions,
- Radionuclide migration in dynamic conditions.

### Mechanical Characteristics

Testing of concrete and mortar compressive strength is a classical method in civil engineering practice. Cube shaped concrete samples 10x10x10 cm were used. Compressive strength (M) is expressed in MPa.

### Permeability

We used permeability measurements as a method which defines property of a porous material which characterizes the easiness with which a fluid may be made to flow through the material by an applied pressure gradient. Permeability is the fluid conductivity of the porous material. In aim to define permeability in terms of measurable quantities we used Darcy's law equation.

Each concrete sample was subjected to nitrogen permeability measurement on absolute pressure of 1 MPa and temperature of 20°C. Samples have been conserved for 28 days on 20°C and 65% humidity. Apparatus for permeability measurement is shown on Fig. 2. Flow of a gas through concrete follows Darcy's law with great degree of approximation. By integrating Darcy's law over height and cross section (for cylinder shaped samples) permeability coefficient K(cm$^2$) is calculated:

$$K = \frac{Q \cdot H \cdot \eta}{S\,(P - P_o)};\quad Q = \frac{V \cdot d \cdot P_i}{P_m \cdot t} \qquad \text{(Eq. 1)}$$

whereas:

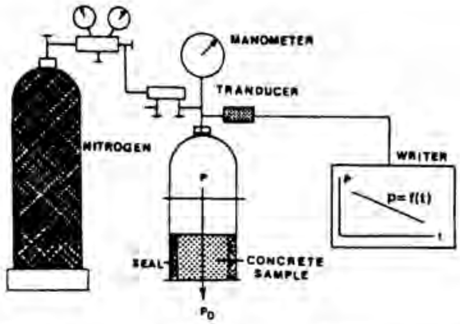



Fig. 2. Scheme of the experimental equipment for permeability measurement.

Q = gas flow (cm$^3$/s)
K = permeability coefficient (cm$^2$)
S = sample cross section (cm$^2$)
$P_o$ = absolute pressure (Pa)
$\eta$ = gas viscosity (Pa·s)
H = sample height (cm)
t = time (s)
P = starting pressure (Pa)
$dP_i$ = actual pressure (Pa)
$P_m$ = mean pressure (Pa)

Concrete samples have been cylindrical in shape (diameter 10 cm, height 10 cm). Testing of mechanical characteristics and permeability of concrete was done with each of forty formulations. Experimental results for 40 concrete mixture formulas for containers are shown in Table I.

### Leakage Test

Apparatus for concrete and mortar leakage test has been constructed in Institute of Nuclear Sciences - Vinca. This original method provides a virtual image about the ability of matrix to prevent "washing out" of solidified waste materials by underground flows.

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 (engineer trenches) can be expected. Expected period of time is 300 to 500 years.

This results may be expressed by incremental leakage rate R(cm/d) which has the dimensions of speed:

$$R(\text{cm/d}) = \frac{\Sigma a_n}{A_o} \cdot \frac{V}{s} \cdot \frac{1}{t_n} \qquad \text{(Eq. 2)}$$

whereas:

$a_n$ = radioactivity leaked during the leakage renewal period (Bq)
$A_o$ = radioactivity initially present above concrete or mortar disk (Bq)
V = volume of concrete or mortar disk (cm$^3$)
S = exposed surface area of concrete or mortar disk (cm$^2$)
$t_n$ = duration of leachant renewal period (d).

Table II gives incremental leaching rates, $R_{nm}$(cm/d), for six mortar compositions after 180 days of experiment.

**TABLE I**
Results Of Experimental Measurement of 40 Concrete Mixture Formulas

| Cement | Additive | No. | M(MPa)* | K($cm^2$) | Cement | No. | M(MPa)* | K($cm^2$) |
|---|---|---|---|---|---|---|---|---|
| $C_1$ | Fluidal VX-OC | 1 | 42.1 | $4.69x10^{-12}$ | $C_2$ | 1 | 44.2 | $2.80x10^{-13}$ |
| | | 2 | 47.6 | $6.50x10^{-12}$ | | 2 | 44.2 | $1.15x10^{-13}$ |
| | | 3 | 48.1 | $6.60x10^{-12}$ | | 3 | 49.8 | $1.17x10^{-13}$ |
| | | 4 | 43.4 | $1.18x10^{-12}$ | | 4 | 44.0 | $1.28x10^{-13}$ |
| | | 5 | 43.2 | $8.16x10^{-13}$ | | 5 | 51.2 | $2.66x10^{-13}$ |
| | | 6 | 40.6 | $1.50x10^{-11}$ | | 6 | 43.0 | $5.30x10^{-13}$ |
| | | 7 | 37.0 | $8.94x10^{-13}$ | | 7 | 38.0 | $8.12x10^{-13}$ |
| | | 8 | 40.0 | $8.16x10^{-13}$ | | 8 | 43.4 | $4.17x10^{-13}$ |
| | | 9 | 47.0 | $6.05x10^{-13}$ | | 9 | 47.5 | $6.62x10^{-13}$ |
| | | 10 | 42.0 | $9.38x10^{-13}$ | | 10 | 43.0 | $9.38x10^{-13}$ |
| | Super Fluidal | 1 | 44.4 | $2.68x10^{-13}$ | | 1 | 43.2 | $4.30x10^{-13}$ |
| | | 2 | 36.8 | $1.04x10^{-12}$ | | 2 | 47.2 | $1.07x10^{-13}$ |
| | | 3 | 46.3 | $7.61x10^{-13}$ | | 3 | 52.0 | $1.12x10^{-13}$ |
| | | 4 | 46.0 | $8.20x10^{-13}$ | | 4 | 46.2 | $1.56x10^{-13}$ |
| | | 5 | 44.6 | $9.71x10^{-13}$ | | 5 | 47.4 | $3.41x10^{-13}$ |
| | | 6 | 42.3 | $1.08x10^{-12}$ | | 6 | 43.0 | $5.20x10^{-13}$ |
| | | 7 | 37.0 | $1.94x10^{-12}$ | | 7 | 45.0 | $2.81x10^{-13}$ |
| | | 8 | 40.0 | $4.02x10^{-13}$ | | 8 | 44.0 | $2.62x10^{-13}$ |
| | | 9 | 47.0 | $2.34x10^{-13}$ | | 9 | 50.0 | $4.08x10^{-13}$ |
| | | 10 | 42.0 | $5.21x10^{-13}$ | | 10 | 44.1 | $2.88x10^{-13}$ |
| *Measured after 28 days. | | | | | | | | |

**TABLE II**
Incremental Leaching Rates, $R_{nm}$(cm/d) for Six Mortar Compositions

| Formulation | $M_1$ | $M_2$ | $M_{11}$ | $M_{12}$ | $M_{13}$ | $M_{15}$ |
|---|---|---|---|---|---|---|
| $R_{nm}{}^{60}Co$(cm/d) | $5.16x10^{-5}$ | $7.60x10^{-5}$ | $1.80x10^{-4}$ | $1.90x10^{-4}$ | $3.90x10^{-5}$ | $4.70x10^{-5}$ |
| $R_{nm}{}^{137}Cs$(cm/d) | $6.94x10^{-4}$ | $6.90x10^{-4}$ | $1.94x10^{-4}$ | $2.00x10^{-4}$ | $1.16x10^{-4}$ | $1.00x10^{-4}$ |
| $R_{nm}{}^{85}Sr$(cm/d) | $4.65x10^{-4}$ | $5.43x10^{-5}$ | $1.13x10^{-4}$ | $9.10x10^{-5}$ | $7.40x10^{-5}$ | $6.60x10^{-5}$ |
| $R_{nm}{}^{54}Mn$(cm/d) | $2.65x10^{-4}$ | $8.10x10^{-5}$ | $1.84x10^{-4}$ | $1.90x10^{-4}$ | $3.80x10^{-5}$ | $3.40x10^{-5}$ |

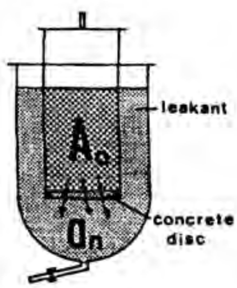



Fig. 3. Scheme of the experimental equipment for leakage rate measurement.

**Determining of Radionuclide Migration Through the Cement-Waste Forms in Static Semi-Real Static Condition**

**Radionuclide Release Scenario**

Final shallow land repository is under unsaturated condition, and the principal process of the radioactivity release to environment is the hydrological transport of radionuclides by the ground water.

Therefore we consider only this release pathway for simplicity. Radionuclide release scenarios are considered in both of normal and special cases (6).

a. Under normal condition, the rainfall percolates through the trench cover and concrete container and comes into contact with the waste matrix eventually. Then, the radionuclides are leached from the immobilized radioactive waste material (Fig. 4a), and

**TABLE III**
Incremental Leakage Rates, $R_n$(cm/d) for Experiment Innormal Condition,
After 485 Days, for $^{60}Co$ and $^{137}CS$

| τ = 7 d | | τ = 14 d | | τ = 100 d | |
|---|---|---|---|---|---|
| $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ |
| $3.90x10^{-8}$ | $4.08x10^{-7}$ | $2.70x10^{-8}$ | $3.22x10^{-7}$ | $7.20x10^{-8}$ | $6.28x10^{-7}$ |
| τ = 205 d | | τ = 320 d | | τ = 485 d | |
| $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ |
| $2.20x10^{-8}$ | $3.02x10^{-7}$ | $2.10x10^{-8}$ | $3.00x10^{-7}$ | $2.08x10^{-8}$ | $2.98x10^{-7}$ |

**TABLE IV**
Incremental Leakage Rates, $R_n$(cm/d) for Experiment Under Special Condition
("9.8 kPa"), after 485 Days for $^{60}Co$ and $^{137}Cs$

| τ = 7 d | | t = 14 d | | τ = 100 d | |
|---|---|---|---|---|---|
| $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ |
| $4.20x10^{-8}$ | $3.20x10^{-7}$ | $2.22x10^{-8}$ | $2.80x10^{-7}$ | $1.80x10^{-8}$ | $2.40x10^{-7}$ |
| τ = 205 d | | τ = 320 d | | τ = 485 d | |
| $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ | $^{60}Co$ | $^{137}Cs$ |
| $1.10x10^{-8}$ | $1.50x10^{-7}$ | $2.20x10^{-8}$ | $3.10x10^{-7}$ | $2.30x10^{-8}$ | $3.14x10^{-7}$ |

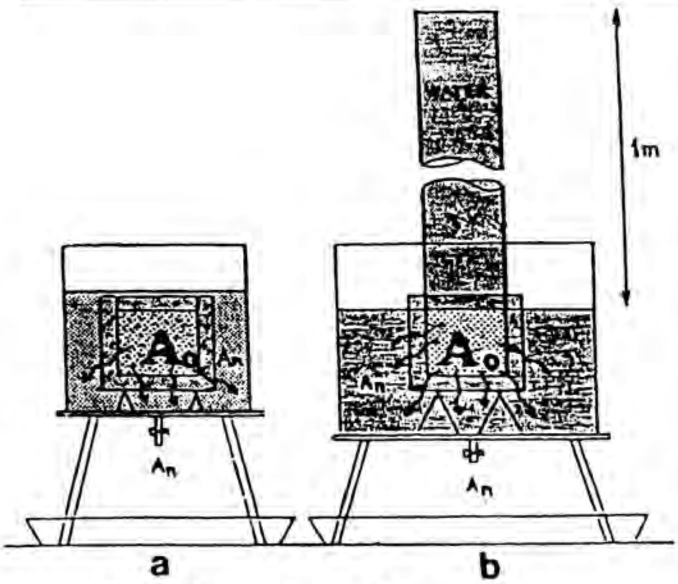



Fig. 4. Schematic layout of the apparatus.
a. under normal condition
b. under special condition (9.8 kPa)

b. Under special condition, where 1m column of water is above the concrete container, and water comes into contact with the radionuclides in immobilized cement matrix with additional pressure of 9,8 kPa! (Fig. 4b).

In both of normal and special condition, the leached radionuclides percolate downward through the concrete system and enter an aquifer. Based on these scenarios we have realized two environmental simulation testing apparatus in which we measured radionuclide migration velocities in semi real condition.

Table III gives incremental leakage rates, $R_n$(cm/d) for $^{60}Co$ and $^{137}Cs$, for experiment in normal condition, after 485 days.

Table IV gives incremental leakage rates, $R_n$(cm/d) for $^{60}Co$ and $^{137}Cs$, for experiment under special condition, additional pressure of "9.8 kPa", after 485 days.

**Determining of Radionuclide Migration Through Cement-Waste Forms in Dynamic Condition**

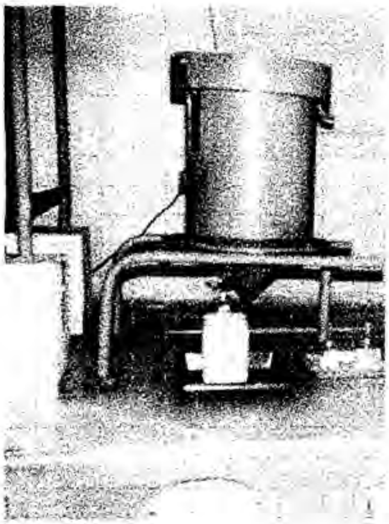
Based on described scenarios, we have constructed a simulation test apparatus which we used for measuring radionuclide migration velocities in real dynamic conditions.

Dynamic conditions leaching test apparatus has been constructed in Institute of Nuclear Sciences - Vina. This original method provides a virtual image about the ability of mortar and concrete barriers to prevent "washing out" of immobilized radionuclides by water, shown in Fig. 5. The concrete container, containing in mortar immobilized total activity of $7{,}72x10^{7}$Bq of $^{60}Co$, $^{137}Cs$ and $^{54}Mn$, was totally immersed in water and granulate, for a period of 365 days. A downward flow of water was carried out at a constant feed velocity of 500 ml/day.

The solution (leachant) were removed at selected intervals in order to determine the $^{60}Co$, $^{137}Cs$ and $^{54}Mn$ content in leachant, $A_n$ (Bq).

Measurements of the radionuclides presence in the leachant were performed by the "EG&G"-ORTEC spectrometry system and software", with the low-level background.

Leachant samples were introduced to a 500 ml Marinelli standard beaker, and measured up to 200000 s. Upon to the obtained spectra, there are no measurable nor significant



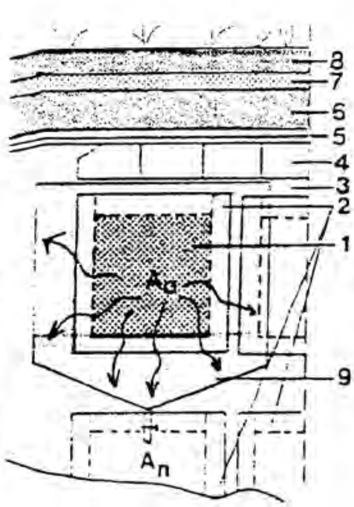



Fig. 5. Apparatus for leaching in real dynamic conditions.

amount of $^{60}$Co, $^{137}$Cs and $^{54}$Mn in the leachant after 365 days of the experiment duration! Nevertheless, experiment is still in progress, and the leachant renewal period, as well as a whole experiment, will be kept in the same conditions.

## CONCLUSION

Performed experiments experience has given the possibility to choose the best formulations for cement mixtures and obtained results gave us certain to claim that described methods and used matrix materials will serve as a barriers to preserve radionuclide migration to the surroundings, for at least 300 years.

Optimization of the processes and matrix-radwaste mixtures is in progress, and we hope that this work will influence the design of the future Yugoslav storage center, shallow land barrel type, for low and intermediate level radioactive wastes.

## ACKNOWLEDGMENT

Authors are grateful to the Serbian Ministry of Science and Technology for partial financial support.

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