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Modeling of physico-chemical characteristics of mortar-waste mixture in radioactive waste management

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An optimization of mortar (as matrix), improved with vermiculite clay, used for immobilization of radionuclide caesium-137 is presented. A relatively simple mathematical model is given, which permits minimization of leach rate and permeability and maximization of compressive strength. An optimal solution, based on experimental data, is given. These results will be used for a future radioactive waste disposal center in Serbia.

1. Introduction

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.

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

(1) leach resistant so that the release of radionuclides will be slow even though flowing water may contact them.
(2) mechanically, physically and chemically stable for handling, transport and disposal (The Management System, 2008; Jacob, 1972)

According to IAEA Safety Guide (The Management System, 2008), intermediate level wastes must be put in concrete arrangements. To meet this specification, trenches are dug below ground level but above the water table. Then the waste containers are placed in the trench. After each level is filled 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 can be built and engineering trench systems should provide three biological protection barriers as follows:

- Mortar for immobilizing the waste and filling the concrete containers (**M**).
- Concrete container (**C**).
- Concrete for filling trenches (**T**).

These three kinds of mortar and concrete, which have totally different composition and function, make up the whole technological unit. For mortar for container filling, being the most important element of the system, material choice is very important. Granulometric composition calculation and rigorous control of its main physico-mechanical characteristics are very important. In this paper we optimize mortar for filling container, (**S**).

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2. Experimental

2.1. Mortar composition

About hundred different formulations of mortar were examined to optimize their mechanical and physicochemical properties. In this paper we discuss the six mortar samples, **S**. The composition and initial radioactivity A_0(Bq) per sample are shown in Table 1.

2.2. Mechanical characteristics

Testing of concrete compressive strength is a classical method which is practiced in civil engineering. Cube shaped concrete samples 10 × 10 × 10 cm were used. Compressive strength (M) is expressed in MPa (Jacob, 1972). Data for mechanical characteristics M (MPa).

2.3. Permeability

Permeability is the property of a porous material, such as mortar and concrete, that is an indication of the ability for fluids (gas or liquid) to flow through this material. The SI unit for permeability is m^2. A practical unit for permeability is the darcy (d). The equation which defines permeability in terms of measurable quantities is called DARCY's law (Jacob, 1972).

Each mortar sample was subjected to a nitrogen permeability measurement with an absolute pressure of 1 MPa and a temperature of 20 °C.

The permeability coefficient (for cylinder shaped samples) K (cm^2) is calculated:

$$K = Q \cdot H \cdot \eta / S(P - P_o) \tag{1}$$

$$Q = V \cdot d \cdot P_i / P_m \cdot t$$

where

K – permeability coefficient (cm^2),
Q – gas flow (cm^3/s),
H – sample height (cm),
η – gas viscosity (Pa·s),
S – sample cross section (crn^2),
P – starting pressure (Pa),
Po – absolute pressure (Pa),
V- volume (cm^3),
dPi – actual pressure (Pa),
Pm – mean pressure (Pa),
t – time (s).

Mortar samples are cylindrical in shape (diameter 10 cm, height 10 cm).

Data for coefficient of permeability K(cm^2) are listed in Table 2.

2.4. Leaching test

Several standard methods and empirical formulas to calculate the rate of release of radionuclides to the environment have been proposed in the literature. Among these Hespe's leach test proposed by the International Atomic Energy Agency was chosen and followed in the present study (Hespe, 1971). The rate of leaching of radioactive isotopes from solidified wastes is generally not constant but is dependent upon the duration of contact with water. The leaching rate is maximum during the initial contact with water. Then the leaching rate usually decreases and finally becomes almost constant. The magnitude of the decrease in leach rate and the time required for the leach rate to reach an essentially steady-state value differ for different solid forms (Atkinson and Nickerson, 1988; Glasser, 2013; Atkinson et al., 1986; Plecas et al., 1991b,c; Plecas et al., 1992f).

Leach curves were obtained by plotting the cumulative leach fraction (Σa_N/Ao)/(S/V) **versus** square root of total leachant renewal period $t^{1/2}$, where:

Ao – radioactivity initially in the specimen (Bq),
a_N – radioactivity leached during the leachant renewal period N (Bq),
t – duration in days of leachant renewal period (d),
S – exposed surface area of. The specimen (cm^3).
V – volume of the specimen (cm^3).

Data for leach factors (L) are given in Table 2, where:

$$L = \pi m^2 / 4 \left(cm^2 / d \right) \tag{2}$$

where **m** is the slope of straight line from a plot of (Σa_N/Ao)/(S/V) versus $t^{1/2}$.

Table 1
Mortar composition (calculated as grams for 1000 cm^3 of mortar).

	S_1	S_2	S_3	S_4	S_5	S_6
Portland cement, g	750	720	700	680	660	640
Sand (0–2), mm	1250	1270	1290	1310	1330	1350
Water, ml	270	275	280	300	310	330
Additives, ml	15	15	14	14	13	13
Vermiculite, g	–	–	50	40	30	25
^{137}Cs (MBq)	3.7	3.7	3.7	3.7	3.7	3.7

3. Results

Experimental results for 6 mortar formulations are shown in Table 2.

4. Mathematical modeling and optimization

The most important mechanical and physical characteristics of a mortar waste composition are:

- (**M**) – Mechanical strength (MPa) (compressive strength)
- (**K**) – Permeability (cm^2)
- (**L**) – Leaching rate (cm^2/d)

It is well known that permeability and leaching rate should be minimized and mechanical strength maximized. These physical requirements suggest the following selection of the objective function, **F**:

$$\mathbf{F(M, L, K) = K^x L^y / M^z} \tag{3}$$

Considering the physicochemical nature of these parameters, the following functional relations may be formulated:

$$\mathbf{F_1(M, L) = 0} \tag{4}$$

$$\mathbf{F_2(K, L) = 0} \tag{5}$$

Values for **M**, **K** and **L** (^{137}Cs) shown in Table 2 have been measured experimentally. Functional Relations (4) and (5) have been analyzed by polynomial least –squares approximation of the experimental data. It has been found that a polynomial of first and second order approximates the measured data with satisfactory. Linear relationships represent the physical nature of the variables better and simplify the optimization (Atkinson and Nickerson, 1988; Glasser, 2013; Atkinson et al., 1986; Plecas et al., 1991b,c; Plecas et al., 1992f).