OPTIMISATION BY MATHEMATICAL MODELING OF PHYSICOCHEMICAL CHARACTERISTICS OF CONCRETE CONTAINERS IN RADIOACTIVE WASTE MANAGEMENT

by

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A method for obtaining an optimal concrete container composition used for storing radioactive waste from nuclear power plants is developed. It is applied to the radionuclides ^{60}Co, ^{137}Cs, ^{85}Sr, and ^{54}Mn. A set of recipes for concrete composition leading to an optimal solution is given.

Key words: concrete, waste, permeability, mechanical characteristics, leakage test

INTRODUCTION

Radioactive waste has a high negative influence on the environment [1-3]. Due to this, it has to be properly conditioned in order to assure hazard-free transport and storage. This requires careful handling of the waste, *i. e.* the employment of adequate immobilization and packaging techniques. As a result of proper immobilization, radioactive waste is packed into a more desirable form defined by several important characteristics, the most notable being: leaching rate, permeability, and mechanical strength [1-6] which can be directly measured. The leaching rate, usually obtained by leaching tests, used to help assess the ability of a pollutant to partition from a waste into a surrounding liquid medium, is expressed by the incremental leaching rate R, while permeability pertains to the property of a porous material characterizing the case in which a fluid may be made to flow through the material by an applied pressure gradient, expressed by the coefficient of permeability K. The mechanical characteristics of immobilization are obtained by testing the concrete compressive strength, which is a classical method practiced in civil engineering. In our investigation the cube-shaped concrete samples of 10 cm 10 cm 10 cm were used.

The results of the measurements for a number of various concrete containers are presented in this paper. Using the obtained results as a basis, we propose a novel method for achieving a broad range of optimal concrete compositions and discuss the implications of our findings.

Experiment

A detailed description of the experimental set-up is given in [1, 2, 7], along with a thorough description of measured quantities: M – mechanical strength [MPa], K – permeability [cm^2], and R – leakage rate $[\mathrm{cm\,d^{-1}}]$. The analyzed concrete samples were made of following ingredients:

- portland cement, PC-20Z 45 MPa (further on, denoted as C_1) and portland cement, PC-55 45 MPa (denoted as C_2),
- sand and granulate,
- water attested according to Serbian standards, and
- additives: fluidal VX-OC and superfluidal.

The exact ingredients and initial radioactivity A_0 [Bq] of the analyzed concrete samples are shown in tab. 1.

Experimental results for the analyzed concrete compositions of concrete containers formulations are given in tab. 2.

In order to precisely parameterize each concrete composition, we have introduced variables p_1, p_2, p_3, and p_4 which correspond to a 0-2 mm percentage of sand and 2-4 mm, 4-8 mm, and 8-15 mm percentage of granulate fractions, respectively. The weight of the cement, water and additives was kept constant, while samples of the concrete were prepared for experimental measurements and cannot, therefore, be used for the parameterization of other concrete samples.

Comparison of various concrete groups and the choice of an optimal concrete composition

As noted in the previous Section, quantities p_1, p_2, p_3, and p_4, can be used to define a recipe for con-

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Table 1. Analyzed samples of concrete containers C, calculated as grams per 1000 cm^3 of concrete

	C_1	C_2	C_3	C_4	C_5	C_6	C_7	C_8	C_9	C_{10}
Cement Portland [g]	390	390	395	395	400	400	405	405	410	410
Sand 0-2 mm	838	600	655	505	670	842	672	525	750	580
Aggregate 2-4 mm	90	66	69	56	67	91	73	57	80	65
Aggregate 4-8 mm	550	596	417	495	450	575	300	430	410	466
Aggregate 8-15 mm	500	725	803	940	770	460	903	980	720	840
Water [ml]	165	165	170	170	185	185	190	190	195	195
Additive [ml]	4	4	4	4	4	4	4	4	4	4
^{60}Co [MBq]	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7
^{137}Cs [MBq]	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7
^{85}Sr [MBq]	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7
^{54}Mn [MBq]	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7	3.7

crete container composition. As they sum to unity and, in addition $p_2 = 0.11p_1$ (according to civil engineering standards), only two of these parameters are independent. Based on data from tab. 1, the choice was made to use the two parameters that span the parametric space most evenly, namely p_1 and p_3.

It is evident that the permeability and leakage rate should be minimized and mechanical strength maximized. These physical requirements influence the form of the objective function, $F(M, K, R) = K^q R^r/M^s$ which allows one to directly compare two different concrete compositions. Since for our practical purposes mechanical strength M is more important than permeability K and leakage rate R, the objective function is selected as follows: $F(M, K, R) = K R/M^3$. More details on the form of the objective function can be found in [1, 2, 7].

Based on experimental results shown in tab. 2 and the chosen objective function, it is possible to compare various concrete compositions. In our experiment, four groups of concrete were studied, each comprised of a different combination of portland cement (C_1 or C_2) and additives (fluidal VX-OC and superfluidal). In each group, a total of 10 mixtures were made, the recipes for each given in tab. 1. The goal is to enable one to determine optimal parameters p_1, p_2, p_3, and p_4 for which the objective function will be within a specified range. To make the process of fitting of experimental data given in tab. 2 easier, the following logarithm of the objective function was used

$$f = \log\frac{KR}{M^3} = \log(K) + \log(R) - 3\log(M) \quad (1)$$

The two mixtures (1 and 2) described by the set of values (M_1, K_1, R_1) and (M_2, K_2, R_2), respectively, (obtained from standardized measurements), are compared by calculating the objective function f. The mixture with a lower value of the corresponding function f is taken to be a better quality concrete composition. Next, function f was fitted inside the convex hull spanned by parameters p_1 and p_3 for each group of concretes. Based on these results, the optimal solution can be easily found. For completeness, the method of fitting is described next.

METHOD OF FITTING

We will use the method of fitting based on B-splines. In order to introduce the precise terminology, we give a basic spline definition first [8]:

Definition: Given $n > 0$, $k \geq 1$ and a non-decreasing sequence of real numbers, $U = (u_0, u_1, ..., u_{n+k})$, the knot vector hereafter, we define the functions $N_{i,k} : R \to R$, $0 \leq i \leq n$, recursively, as follows

$$N_{i,1}(u) = 1, \text{for } u_i \leq u < u_{i+1} \text{ and } = 0, \text{elsewhere} \quad (2)$$

If $k > 1$, then

$$N_{i,k}(u) = \frac{u - u_i}{u_{i+k-1} - u_i} N_{i,k-1}(u) + \frac{u_{i+k} - u}{u_{i+k} - u_{i+1}} N_{i+1,k-1}(u) \quad (3)$$

If the division by zero occurs in any of the terms of the algorithm, the term should be replaced by a zero. The function $N_{i,k}(u)$ is called i-th B-spline or the B-spline basis function, of order k and $k - 1$ degree with respect to knot vector U.

In this work we have used B-splines of order 2 in both parametric directions, with a knot vector [505.0, 505.0, 842.0, 842.0] for the p_1 parameter and knot vector [300.0, 300.0, 596.0, 596.0] for the p_3 parameter. Hence, function f is given by

$$f(p_1, p_3) = \sum_{i,j} C_{i,j} N_{i,2}(p_1) N_{j,2}(p_3) \quad (4)$$

where, coefficients $C_{i,j}$ are obtained using the least-squares method and data from tab. 2. The described method allows the use of both smaller and larger experimental datasets, while not increasing the spline order. In this manner, it is possible to set the degree of the fitting surface in advance, independently from the number of measurement points.

Table 2. Mechanical characteristics *M*, coefficient of permeability *K*, and leach rate measurements *R*, after 180 days, for ten concrete container compositions

Cement Portland and Additive	Sample	*M* [MPa]*	*K* [cm^2]**	*R* [cm/d]**
C_1, fluidal VX-OC	1	38.1	$4.20\ 10^{-12}$	$5.50\ 10^{-5}$
C_1, fluidal VX-OC	2	37.9	$6.80\ 10^{-12}$	$3.30\ 10^{-5}$
C_1, fluidal VX-OC	3	37.1	$6.50\ 10^{-12}$	$6.00\ 10^{-5}$
C_1, fluidal VX-OC	4	38.4	$2.60\ 10^{-12}$	$8.30\ 10^{-5}$
C_1, fluidal VX-OC	5	39.2	$7.10\ 10^{-13}$	$5.20\ 10^{-5}$
C_1, fluidal VX-OC	6	40.6	$1.80\ 10^{-11}$	$6.00\ 10^{-5}$
C_1, fluidal VX-OC	7	40.0	$6.54\ 10^{-13}$	$4.40\ 10^{-5}$
C_1, fluidal VX-OC	8	44.0	$7.10\ 10^{-13}$	$9.30\ 10^{-5}$
C_1, fluidal VX-OC	9	46.0	$6.00\ 10^{-13}$	$5.40\ 10^{-5}$
C_1, fluidal VX-OC	10	48.0	$6.30\ 10^{-13}$	$4.20\ 10^{-5}$
C_1, superfluidal	1	37.4	$3.68\ 10^{-13}$	$2.40\cdot 10^{-5}$
C_1, superfluidal	2	37.8	$4.05\ 10^{-12}$	$5.70\ 10^{-5}$
C_1, superfluidal	3	40.3	$3.64\ 10^{-13}$	$4.50\ 10^{-5}$
C_1, superfluidal	4	40.0	$6.24\ 10^{-13}$	$6.90\ 10^{-5}$
C_1, superfluidal	5	40.6	$5.76\ 10^{-13}$	$7.90\ \ 0^{-5}$
C_1, superfluidal	6	41.3	$4.05\ 10^{-12}$	$8.20\ 10^{-5}$
C_1, superfluidal	7	42.0	$4.95\ 10^{-12}$	$3.10\ 10^{-5}$
C_1, superfluidal	8	43.0	$6.03\ 10^{-13}$	$3.30\ 10^{-5}$
C_1, superfluidal	9	46.1	$3.36\ 10^{-13}$	$5.20\ 10^{-5}$
C_1, superfluidal	10	47.0	$4.25\ 10^{-13}$	$3.40\ 10^{-5}$
C_2, fluidal VX-OC	1	42.1	$5.80\ 10^{-13}$	$3.40\ 10^{-5}$
C_2, fluidal VX-OC	2	41.9	$3.15\ 10^{-13}$	$6.90\ 10^{-5}$
C_2, fluidal VX-OC	3	41.1	$1.88\ 10^{-13}$	$8.60\ 10^{-5}$
C_2, fluidal VX-OC	4	42.4	$3.86\ 10^{-13}$	$9.30\ 10^{-5}$
C_2, fluidal VX-OC	5	43.2	$3.66\ 10^{-13}$	$7.40\ 10^{-5}$
C_2, fluidal VX-OC	6	44.6	$5.50\ 10^{-13}$	$2.30\ 10^{-5}$
C_2, fluidal VX-OC	7	44.0	$7.43\ 10^{-13}$	$1.30\ 10^{-5}$
C_2, fluidal VX-OC	8	46.0	$5.17\ 10^{-13}$	$2.70\ 10^{-5}$
C_2, fluidal VX-OC	9	47.0	$6.90\ 10^{-13}$	$2.20\ 10^{-5}$
C_2, fluidal VX-OC	10	49.0	$8.98\ 10^{-13}$	$1.40\ 10^{-5}$
C_2, superfluidal	1	43.2	$5.30\ 10^{-13}$	$3.30\ 10^{-5}$
C_2, superfluidal	2	45.2	$3.07\ 10^{-13}$	$2.50\ 10^{-5}$
C_2, superfluidal	3	43.0	$3.12\ 10^{-13}$	$6.86\ 10^{-5}$
C_2, superfluidal	4	44.2	$5.01\ 10^{-13}$	$5.60\ 10^{-5}$
C_2, superfluidal	5	46.2	$4.41\ 10^{-13}$	$8.80\ 10^{-5}$
C_2, superfluidal	6	45.6	5.10 10–13	$3.60\ 10^{-5}$
C_2, superfluidal	7	45.0	3.81 10–13	$8.80\ 10^{-5}$
C_2, superfluidal	8	47.0	$3.62\cdot 10^{-13}$	$7.90\ 10^{-5}$
C_2, superfluidal	9	47.0	$5.10\ 10^{-13}$	$7.90\ 10^{-5}$
C_2, superfluidal	10	49.1	$2.88\ 10^{-13}$	$8.90\ 10^{-5}$

*Measured after 28 days; **Measured after 300 days

CHOICE OF OPTIMAL SOLUTION

Shaded contour plots for the 4 groups of concrete compositions are shown in fig. 1. Note that the fit (4) is only used inside the convex hull defined by parametric points (p_1, p_3) and not over the entire rectangular bounding box. This is done so as to prevent unphysical solutions (with negative *M*, *K*, and *R*) in areas with low

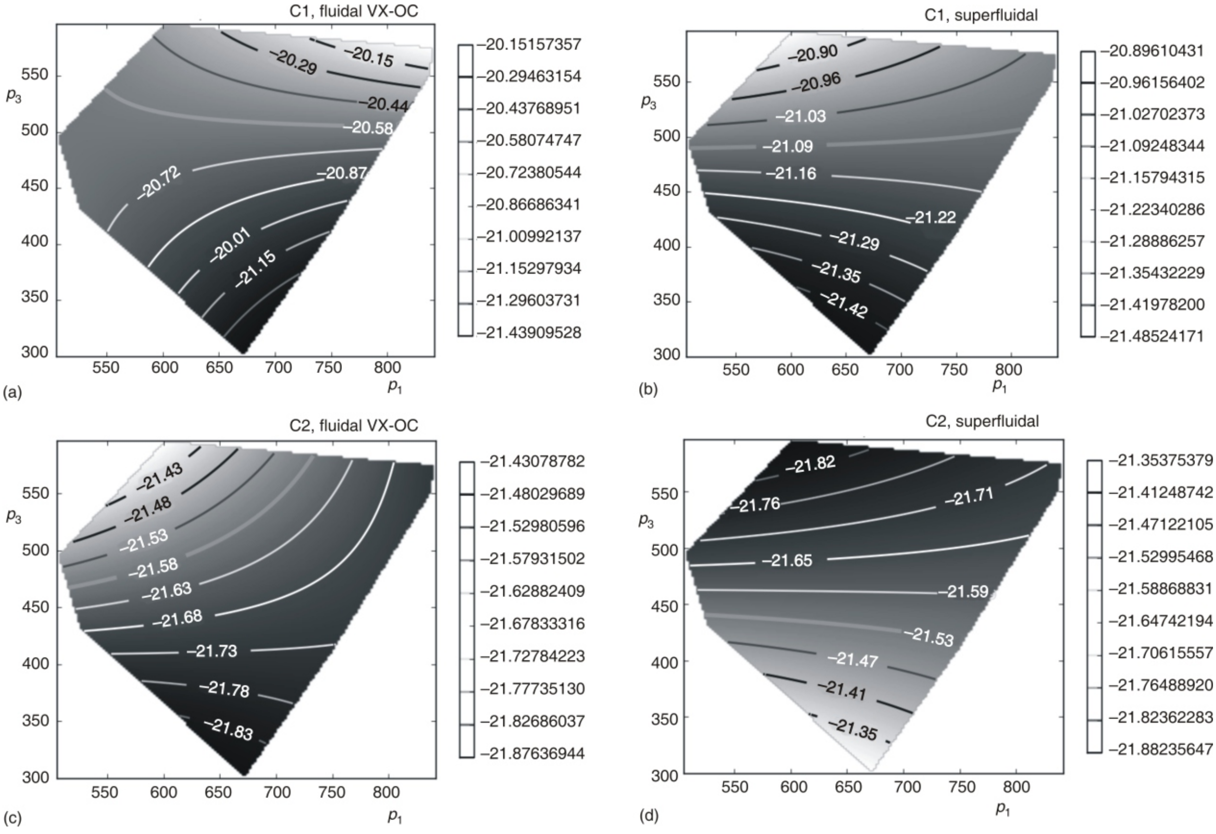

Figure 1. Plot of $f(p_1, p_3)$ for the cement composition using
(a) – cement C_1 and additive fluidal, (b) – cement C_1 and aditive superfluidal, (c) – cement C_2 and additive fluidal, (d) – cement C_2 and additive superfluidal

values of the objective function which would be a result of an extrapolation outside of the (p_1, p_3) convex hull.

Parameter areas with low (desired) values of function *f* are shown in a darker colour, while the areas with higher (undesired) values of function *f* are shown in a brighter colour. All 4 plots corresponding to the 4 different combinations of cement/additive have the same number of isolines (9 isolines, and a single line degenerating to the minimal point of the convex hull, where the level of each isoline is shown on the line itself and, more precisely, in the figure legend). Isolines divide the parametric space into 10 regions, where each region can be considered as being of an approximately equal quality concrete composition. Here, the terms better or equal quality mean a smaller or equal value of the objective function.

In addition to finding an optimal solution within a single group of concrete compositions, it is also possible to determine the best solution for all 4 groups. For example, looking at fig. 1(a), we observe that the optimal mixture has an objective function in ranges [–21.439, –21.296], and that the region of the optimal values of parameters p_1 and p_3 is bounded by the convex hull and the first 2 isolines (note again that the first line has degenerated because the extrapolation wasn't used). Comparing the regions with the lowest values of the objective function in all four groups of concrete compositions, it can be seen that groups 3 and 4, where Portland cement C_2 was used, have an advantage over groups 1 and 2 in which Portland cement C_1 was the one applied. With groups 3 and 4 isolated as the best candidates for an optimal solution, it is interesting to note that the parametric range of the highest quality region differs entirely in these two cases, leading to the conclusion that additives can play a crucial role in the quality of cement composition. This fact can be used to determine which cement and additive to use when the percentage of 0-2 mm sand (p_1) and a percentage of granulate (p_2, p_3, and p_4) is given and *vice versa*.

CONCLUSIONS AND DISCUSSION

In the present paper, a thorough study of concrete compositions used in low and intermediate level waste conditioning is presented. The comparison of concrete compositions, where each composition is described via experimentally obtained mechanical strength, permeability and leakage rate, was made possible by a suitably defined objective function. The objective function was fitted by means of the B-spline surface across the convex parametric region which spans the space in which the optimal solution is to be sought. The continuous objective function obtained for the 4 combinations of cement and additives enables a direct comparison of compositions belonging either to the same or a different combination. Compositions involving portland cement PC-55 45 MPa (C_2) turned out to be a better choice than the ones involving portland cement PC-20Z 45 MPa (C_1), as far as the defined objective function is concerned. Among the compositions using the specified cement and additives, the precise range was given for a percentage of 0-2 mm sand (p_1) and a percentage of granulate (p_2, p_3, and p_4) that gives an optimal mixture.

It should also be noted that a minimum of the objective function in the strict mathematical sense was not observed, because it is not present in the considered convex hull. The objective function assumes smaller values near the boundaries of the parameter space, but not the local minimum. This suggests that further measurements in a broader range of parameter space would be desirable. Such results would allow a more complete optimisation.

In papers [1, 7], it was claimed that a true minimum of the objective function is analytically obtained. However, this conclusion is somewhat deceiving because it supposes linear dependences of M and K as functions of L [7]. Since this linear fitting is unreliable, even a slight change of coefficients in corresponding linear functions may lead to a drastic change of the position of the minimum obtained in this manner and, sometimes, even render unphysical results. In the method used in the present paper, the dependency of M and K as functions of L is not supposed, because there are no physical grounds for such a dependency, especially not a linear one. In this sense, the proposed novel approach where the dependence of the objective function of independent parameters is explicitly used should be considered as a new and significant step towards a more reliable optimisation procedure. To avoid the drawbacks of the earlier unreliable analytic approaches, by using derivative functions, we have introduced a corresponding graphical presentation, rather than an analytical treatment of the problem.

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AUTHOR CONTRIBUTIONS

Experiments were carried out by I. Plećaš, while theoretical analysis, discussion of results and composition of the paper were done by all the authors.

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МАТЕМАТИЧКО МОДЕЛОВАЊЕ ФИЗИЧКО-ХЕМИЈСКИХ КАРАКТЕРИСТИКА БЕТОНСКИХ КОНТЕЈНЕРА ЗА ПОТРЕБЕ РУКОВАЊА РАДИОАКТИВНИМ ОТПАДОМ

Отпад ниског и средњег нивоа чини 90% укупног радиоактивног отпада и одлаже се у специјалне бетонске контејнере. Пошто ови контејнери треба да безбедно чувају радиоактивни отпад у периоду око 300 година, одабир и прецизна контрола физичких и механичких карактеристика материјала од велике су важности. У овом раду, приказан је математички модел састава бетонског контејнера који се користи за складиштење радиоактивног отпада из нуклеарних електрана. Представљена је оптимизација матрице од малтера појачаног бентонитском глином, која се користи за имобилизацију радионуклида ^{60}Co, ^{137}Cs, ^{85}Sr, и ^{54}Mn. Дат је скуп могућих састава бетона који доводе до оптималног решења, а који су базирани на резултатима мерења.

Кључне речи: бетон, отпад, пропустљивост, механичка карактеристика, тест цурења