EXPERIMENTAL DESIGN APPROACH IN RADIONUCLIDE SORPTION

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Abstract

This paper presents the utilization of experimental design for investigation of sorption from multi-component systems. i.e. suspensions similar to real liquid radioactive waste (LRW). Experimental design methodology is a powerful tool in the examination of selected ions/radionuclides competition during the sorption. For better understanding, an example of competitive sorption of a multi-component solution of strontium, cobalt, and nickel divalent ions onto waste concrete is shown.

Keywords: *liquid radioactive waste, immobilization, solidification*

INTRODUCTION

Experimental design methodology is a powerful tool in the examination of selected ions (radionuclides) competition during the sorption from multi-component suspensions (systems) which are similar to real liquid radioactive waste (LRW). The experimental design represents a practical method, applicable in all research disciplines, able to investigate relations between the cause and the effect. The main characteristic of this approach is the simultaneous variation of independent variables input. This is an excellent mechanism to minimize the total number of experimental trials, in regard to classic approach (one factor at a time). Depending on problem type and purpose of the investigation, different full or fractional factorial designs, surface methodology, block designs, and others could be utilized. These methodologies are especially useful for the investigation of the experimental conditions modification effects. Otherwise, the influence of mixture composition variation on system response can be investigated using a mixture design as a type of surface response methodology.

EXPERIMENTAL DESIGN

Experimental design is a statistical method that allows simultaneous variation of all factors of an examined process, whereby a large amount of information about a given process is obtained with a relatively small number of experiments. This design allows the definition of empirical mathematical models that describe a given process. Proper selection of independent variables, their variation, and monitoring of system responses can define the influence of factors and the influence of their mutual interactions on the system response, i.e. dependent variable. Depending on problem type and test purpose, different types of experimental designs are developed. If the goal is to define significant process variables (screening), factorial design (complete or fractional) and Plackett-Burman design are most often used. In order to optimize a process, different types of response surface designs are used (e.g. Box-Behnken design, central composite design, Taguchi design, etc.).

Mixture design is a special design type in which the quantities or shares of different components in the mixture (whose influence is examined on some specific property of the mixture: dependent variable, system response, etc.), which are assumed to be only a function of the mixture composition, are varied as independent factors. Three mixture designs are available: Simplex Lattice, Simplex Centroid, and Extreme Vertices design. The experimental domains of Simplex designs cover all components in the mixture, i.e. covers the entire surface of Simplex, while the design of extreme peaks covers only part of Simplex and is used when there are restrictions on the amount/share of some or all components in the mixture.

SIMPLEX CENTROID DESIGN EXAMPLE

The Simplex Centroid experimental design is customarily used to examine ion sorption competitiveness in multi-component solutions during the sorption process. If a three-component mixture is considered (q = 3, where q is a number of components in the mixture), the number of experimental points corresponding to the number of one-, two- or three-component equimolar mixtures in Simplex Centroid design is 7 (2q – 1 = 7). An example of competitive sorption tests of a multi-component solution of strontium, cobalt, and nickel divalent ions onto waste concrete is given below (Table 1). Sorbed amounts of the investigated cations are presented in Figure 1.

A special cubic model is used for the description of data obtained by the Simplex Centroid design. For a three-component solution, this general model can be presented by following equitations (the equations are derived, including only statistically significant terms):

$$\text{Concrete}-\text{Sr} = 0.082151\text{Sr} + 0.006431\text{Co} - 0.008789\text{Ni} \quad (1)$$

$$\text{Concrete}-\text{Co} = -0.001552\text{Sr} + 0.221311\text{Co} + 0.011582\text{Ni} + 0.576376\text{Sr}-\text{Co} \quad (2)$$

$$\text{Concrete}-\text{Ni} = 0.04441\text{Sr} - 0.01226\text{Co} + 0.31937\text{Ni} \quad (3)$$

These equations imply that the sorption of investigated cations is directly proportional and principally affected by their own concentration in the multi-component solution. A significant interaction was found only in the case of Co^{2+} sorption from the mixture, i.e., the interaction between Sr^{2+} and Co^{2+} ions enhanced the efficiency of Co^{2+} removal. According to coefficients in equations 1 – 3, both positive and negative effects of coexisting cations were at least one order of magnitude lower in respect to the most dominant effect. These equations can be graphically presented by the construction of ternary contour plots (Figure 2).

Table 1 - Simplex Centroid Design Matrix

Experimental run	Molar proportions of cations in the mixtures		
	Sr	Co	Ni
1	2/3	1/6	1/6
2	0	1/2	1/2
3	1/3	1/3	1/3
4	0	1	0
5	1/2	0	1/2
6	1/2	1/2	0
7	1/6	1/6	2/3
8	0	0	1
9	1	0	0
10	1/6	2/3	1/6

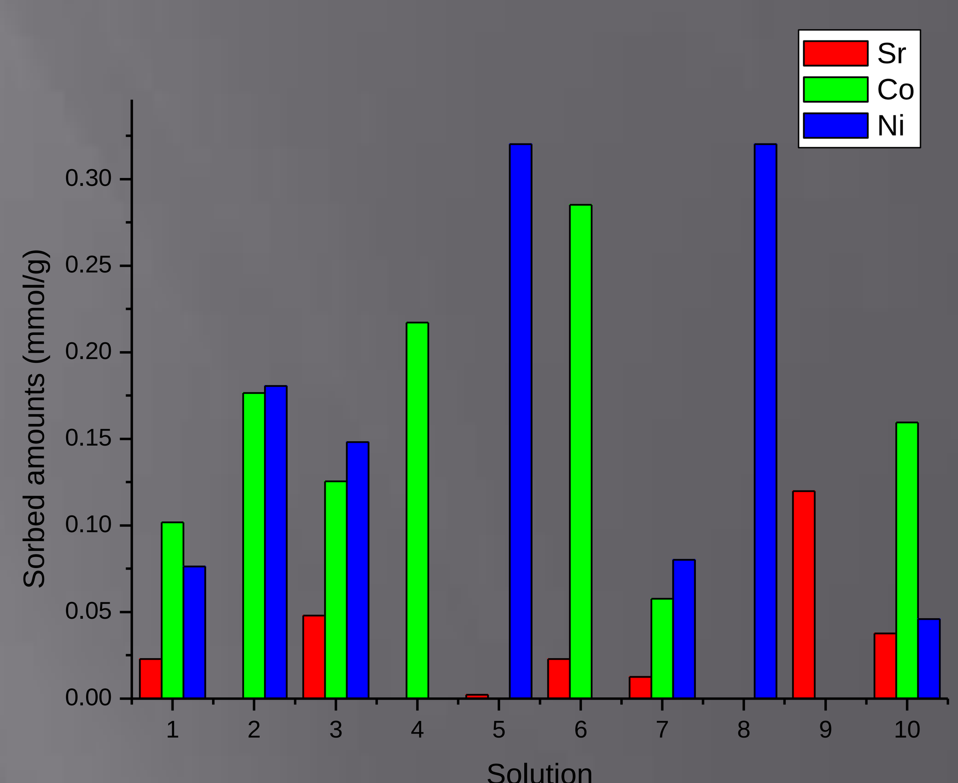

Figure 1. Sorbed amounts of Sr^{2+}, Co^{2+}, and Ni^{2+} from multi-component solutions by waste concrete

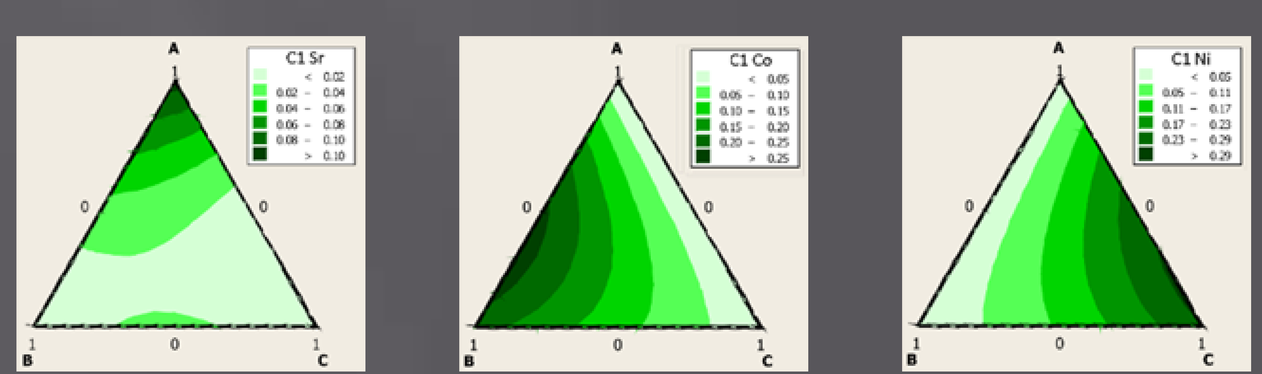

Figure 2. Ternary contour plots of (a) Sr^{2+}, (b) Co^{2+}, and (c) Ni^{2+} sorbed amounts onto waste concrete

CONCLUSION

The experimental design represents a practical method, applicable in all research disciplines, able to investigate relations between the cause and the effect. This design allows the definition of empirical mathematical models that describe a given process. It is a powerful tool in the examination of selected ions (radionuclides) competition during the sorption from multi-component suspensions (systems) which are similar to real liquid radioactive waste (LRW). The Simplex Centroid experimental design is customarily used to examine ion/radionuclide sorption competitiveness in multi-component solutions during the sorption process. For clarity, the example was shown, with given equations and their graphical presentation.