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Factorial Experimental Design for Optimization of Cesium Removal from Aqueous Solutions

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Abstract:_The effective removal of cesium from aqueous solution is an emerging issue all over the world particularly in Japan after destroyed Daiichi nuclear power plant of Fukushima. To reduce the expended chemicals and reagents in experimental work and number of the experiment, it is required to implement statistical optimization of the factors forthe cesium removal process. In this study, factorial experimental design and multivariate regression technique were employed to evaluate the main effects and interactions among the pH, initial concentration and contact time in the cesium removal process using nZVI-Z as an adsorbent. The study revealed that maximum cesium removaloccurred at pH level 6, initial concentration of 200 mg/L and contact time of 30 minutes.Initial concentration was the statistically significant factor followed by contact time. Moreover, the significant interaction effect was observed between contact time and initial concentration._

Keywords: Analysis of variance; Cesium removal; Factorial experiment; Interaction effect; Main effect.

1. INTRODUCTION

The earthquake and the resultant Tsunami on 11th March 2011, completely destroyedthe Daiichi Nuclear Power Plant of Fukushima[1]. As a consequence, a vast amount of radioactive materials discharged into groundwater, seawater and surrounding soil [2]. The most common radioactive form of cesium is Cs-137 which is made by nuclear fission. Cesium is regarded as a deadly hazard contaminant material that severely affects our environment and has half-life is 30.17 years[2]. Hence, cesium removal from liquid waste is a major concern. Ion exchange method has become one of the utmost cesium removal techniques due to its simplicity, efficiency, and selectivity. Eljamal et al. [3,4] developed novel nanoscale composites (nZVI-Z and nFe/Cu-Z) for cesium elimination through ion exchange technique followed by liquid-phase reduction. Takami et al.[5]examined the removal of phosphorus from aqueous solutions using nano-scale zero valent iron. Earlier researchers employed the traditional 'one variable at a time' approach. The variables were autonomous to investigate the single effect of numerousaspects on the removalmethod. In fact, this is not true, and in these circumstances, it is required to take into account numerous influences concurrently[6]. The factorial experimentaldesign is a very expedient technique for this purpose, as it gives statistical models which explain the interactions among the factors that have been optimized[7,8]. Moreover, statistical models give an enormous quantity of information, decrease the total number of experiments, time, amount of chemicals, and total research expenses.

In this study, a factorial design has been employed to investigate the influence of several factors and their relations on the efficiencyof cesium removal and then evaluate the optimum conditions for the removal of cesiumusing iron based nanoparticles-zeolite.

2. MATERIALS AND METHODS

The preparation and characterization of nanoscale zero valent iron-zeolite(nZVI-Z) composite was described by Osama Eljamal et. al [3]. Data have been extracted from this article[3]. In this study, 2^3 factorial design is considered and the high and low levels defined for this design are listed in Table 1.

Table 1. Parameters considered for full factorial designs.

Factors (units)	Levels	
	Low	High
pH	6	10
Initial concentration(mg/L)	100	200
Contact time (min)	5	30

3. RESULTS AND DISCUSSION

3.1 Normal probability plot for 2^3 factorial experiment

For the main and interaction effects, a normal plot is an appropriate way for relating the statistically significant and degree of significance of factorial experiments. Every point on the normal plot indicates a factor and the points close to the slant line are less potentials compared to the distant one (Fig. 1). Factors presented by red square symbols describe the significant effect and away from the slant line are regarded as more significant which affects the removal efficiency of cesium. The plot illustrates that the points representing initial concentration (B), contact time (C), the interaction of concentration and time (BC), pH and concentration(AB), and interaction of pH, concentration and time (ABC) lie on the right portion of the slant line. It implies that studied factors have a positive significant effect on the cesium removal. On the contrary, the points that lie on the left portion of the slant line has negative effect. An increase pH might reduce removal of cesium. The initial concentration (B) has a

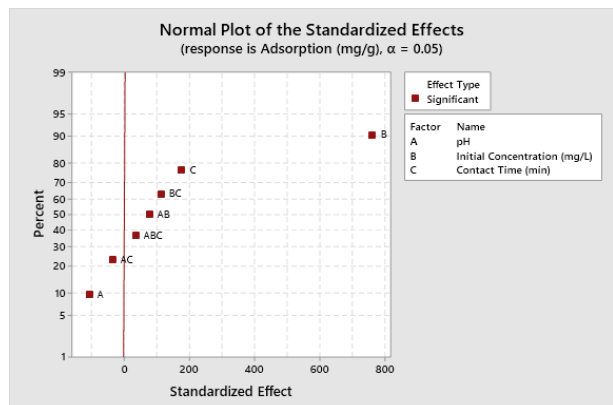leading effect due to its point lies farthest from the slant line.

Fig. 1. Normal probability plot of standardized effects.

3.2 Main effects plotof cesium removal

At two levels in the factorial experiment, the main effects plot is adopted to understand the mean differences of each factor. Fig. 2 illustrates a positive relationship between the cesium removal and contact time and each of initial concentration. It means that cesium removal increases if the initial concentration and contact time increase. Instead, there is a negative correlation between pH and cesium removal. It is observed that if pH increases from 6 to 10, the removal of cesium decreases.
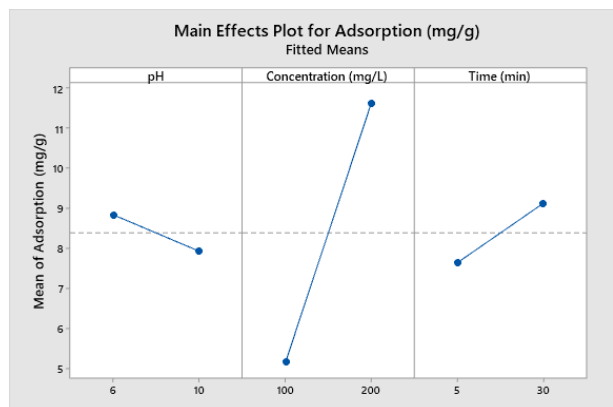

Fig. 2. Main effects plot for cesium adsorption.

3.4 Multivariate regression for the removal of cesium

For more statistical evidence of the adsorption of cesium, statistical analysis containing coefficient of estimated effect, standard error of coefficient, t-value and p-value has been demonstrated in Table 2.

Using the regression coefficient in Table 2 the resulting equation is expressed as equation (1).

$$\eta = 8.382 - 0.456A + 3.232B + 0.744C + 0.329AB - 0.156AC + 0.475BC + 0.143ABC \qquad (1)$$

The coefficient expressed in equation (1) represents the effect of the factors as well as interactions. Here, η represents the amount of cesium removal (mg/g). A positive coefficient means that removal of cesium increases with change in the low to high levels. Oppositely, the negative effect reveals a negative correlation with the cesium removal.

Table 2. Estimated effects and coefficients of 2^3 factorial experiment for cesium removal.

Factor	Effect	Coef.	SE	t-value	P-value
Const.		8.382	0.004	1980.04	2E-16
A	-0.912	-0.456	0.004	-107.81	2E-15
B	6.464	3.232	0.004	763.41	2E-16
C	1.488	0.744	0.004	175.78	2E-15
AB	0.659	0.329	0.004	77.87	2E-14
AC	-0.312	-0.156	0.004	-36.85	2E-14
BC	0.950	0.475	0.004	112.30	9E-15
ABC	0.286	0.143	0.004	33.88	2E-14

4. CONCLUSIONS

A 2^3 full factorial experimental design is employed to identify the effect of initial concentration, contact time and pH and their interaction on cesium removal. Moreover, multivariate regression technique is applied to determine the optimum cesium removal process. Statistical analysis results reveals that the initial concentration, contact time and pH have a significant effect to enhance the removal method. Initial concentration is the most potential factor for this removal process followed by contact time, and interaction between initial concentration and contact time.

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