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Optimization of Strontium Removal Process from Contaminated Water using Zeolite Nanocomposites

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ABSTRACT

The effective removal of strontium from polluted water is an emerging issue worldwide, especially in Japan, after the destruction of Fukushima's Daiichi Nuclear Power Plant. In the strontium removal process, statistical optimization of associated factors is needed to reduce the quantity of chemicals and the number of experimental trials. In this study, response surface methodology based on the central composite design was employed for assessing the influence of different factors and their interaction effects on the efficiency of strontium removal. We have considered nanoscale zero-valent iron-zeolite (nZVI-Z) and nano-Fe/Cu zeolite (nFe/Cu-Z) as adsorbents for the effective removal of strontium. The present study showed that the most statistically significant potential contributor was initial concentration, followed by contact time in the removal process. The study indicated that the interaction effect between contact time and initial concentration was statistically important, suggesting the need for a multi-mechanism technique in the removal phase of strontium. Tóth, Langmuir, Dubinin-Astakhov (D-A), Freundlich, and Hill isotherm models were also fitted with the experimental strontium adsorption data, in which the Tóth model fitted best compared to the other models based on the RMSD.

Keywords: adsorption; central composite design; optimization; removal; strontium.

Abbreviations

df	Degrees of freedom
F-value	Value of F-statistic
MS	Mean sum of squares
p-value	Probability value
RMSD	Root-mean-square deviation
R ² (adj)	Adjusted R ²
std. error	Standard error of estimates
SS	Sum of squares
t-value	Value of t-statistic

Greek symbols

α	Level of significance (-)
τ	Mean removal of strontium (mg g ⁻¹)
λ_0	Overall mean (mg/g)
λ_i	Coefficients of predictor (-)

1. Introduction

The magnitude-9 earthquake in northeastern Japan and hence, tsunami on March 11, 2011, truly smashed the Daiichi Nuclear Power Plant of Fukushima (Shibata et al. 2016). As a result, a significant number of radioactive materials (⁹⁰Sr and ¹³⁷Cs) have been added to groundwater, surrounding soil, seawater, and the surface of the earth (Shibata et al. 2016; Aoyama et al. 2020). Moreover, due to the rapidly growing reliance on nuclear energy, the handling of radioactive waste has become one of the utmost substantial issues in the worldwide management of nuclear waste and the nuclear industry (Hong et al. 2016a). One of the most prevalent dangerous pollutants that badly affect our atmosphere is radioactive strontium(⁹⁰Sr). ⁹⁰Sr works with a respective half-life of 28.8 years as a beta-gamma emitter (Liu et al. 2014; Hong et al. 2016b). Strontium is commonly used in various manufacturing industries, such as fireworks, ceramics, ferrite magnets, and optical materials (Ryu et al. 2016). ⁹⁰Sr discharges a massive amount of gamma radiation which can act

as the basis for human tissue and bone cancer, and leukemia. Further, ^{90}Sr is the ultimate cause of damage to humans by genetically affecting seawater organisms (Liu et al. 2014; Hong et al. 2016b).

The removal of strontium from polluted water is therefore of great concern. The most widely available technique of decontamination is ion exchange, adsorption, evaporation, and magnetic assistance (Ambashta and Sillanpää 2010; Faghihian et al. 2013; Lee et al. 2016; Vipin et al. 2016; Ramanayaka et al. 2020). Besides, chemical precipitation techniques are used for the elimination of radioisotopes from wastewaters (Ambashta and Sillanpää 2010). Currently, ion exchange is one of the sophisticated decontamination techniques. Shubair et al. (2019) investigated the removal of strontium by ion exchange technique using two different adsorbents named nZVI-Z and nFe/Cu-Z. Authors synthesized and characterized considered nanocomposites, allowing contact time, initial concentration, doses, pH, and temperatures as prominent factors. It has been observed that both composites have been effective in removing strontium from the aqueous solution based on adsorption and kinetics properties. El-Kamash (2008) performed a related study to extract strontium (Sr) and cesium (Cs) from polluted water using zeolite A as inorganic ion exchange material. The author found that Sr^{2+} ions had higher column performance and adsorption capacity compared to Cs^{+} ions. A study directed by using nano-Fe/Cu particles as an adsorbent for cesium removal from contaminated water and found that temperature and pH were the dynamic factors (Shubair et al. 2018). Nanosized metal oxides (NMOs) provides a high surface area for removing the heavy metal from contaminated water (Hua et al. 2012). Nanocomposites can eliminate 99% of Hg^{2+} , up to 10% and 90% greater than the removal attained by the zeolite and the parent fly ash, respectively (Tauanov et al. 2018).

However, in the previous studies, one variable at a time technique was applied, including Shubair et al. (2019). The variables assumed independent to examine the single effect for various aspects of the removal method. The independence assumption is not true, and in these circumstances, it is required to take into account numerous effects concomitantly (Echeverría et al. 2005).

For this reason, factorial experimental design is a very convenient technique, as it produces statistical and mathematical models that describe the interactions between the optimized variables (Can and Yildiz 2006; Ponnusami et al. 2007; Saadat and Karimi-Jashni 2011; Geyikçi and Büyükgüngör 2013). In the factorial design, response surface methodology (RSM) is an

extensively used mathematical and statistical model for conducting experiments and optimize process conditions (Ranic et al. 2014; Wang et al. 2016, 2019; Ghafarzadeh et al. 2017; Mohammed et al. 2017; Yildirim et al. 2018; Bahrami et al. 2019; Shahnaz et al. 2020). Furthermore, RSM gives a vast amount of information for reducing the number of the experimental trial, the quantity of chemicals, the time, and overall research cost. For instance, magnesium-rich tricalcium aluminate was employed to remove ammonium and phosphorus from wastewater, and the optimal removal process is identified using the response surface methodology (Mao et al. 2021). This study suggested that the removal significantly depends on temperature, dosage, initial concentration, and pH. Moreover, the statistical factorial experimental design was an effective method applied by various researchers in several scientific studies to optimize the heavy metal removal from polluted water using different types of adsorbents (Seki et al. 2006; Geyikçi and Büyükgüngör 2013; Rêgo et al. 2013; Dong and Sartaj 2016; Ghaedi et al. 2018; Jaria et al. 2019; Perez Mora et al. 2019; Maamoun et al. 2020; Rahman et al. 2020a, b). Moreover, response surface methodology was applied to optimize the removal condition and mechanism of Cr(VI) by Na₂SO₃/CaO (Zhao et al. 2017). Besides, it has been shown that the response surface method is used to examine the potential of Fe₃O₄/mordenite zeolite for the purification of oily wastewater and observed that pH is the most important factor influencing Chemical Oxygen Demand (COD) and Biochemical Oxygen Demand (BOD) removal (Hesas et al. 2019).

To attain the optimal level of strontium removal from polluted water, the literature and context of this study suggest that the statistical factorial experimental technique be an underlying instrument for identifying the essential factors and their interactions. That is why the response surface methodology is necessary for optimizing strontium removal. RSM was first used in the current study to explore the interaction / combined impact of the various key factors on the efficacy of strontium removals, such as initial concentration, contact time, and pH, and to identify optimal conditions using iron-based nanoparticles-zeolite. Normal probability plot, contour graph, surface plot, and main and interaction effect graph have been constructed, and nonlinear regression analysis and analysis of variances have been conducted for finding significant factors of strontium removal. Assumptions validation of the response surface methodology has been performed by making the residual versus fit and normal probability plots. In the existing literature, only Langmuir and Freundlich models are available. In the present study, we have considered five

different most popular isotherm models to relate to the experimental data that could diagnose the best isotherm model for strontium removal.

2. Materials and methods

2.1 Chemicals and preparation of the nanocomposites

Available commercial zeolite in powdered form with particle size less than $45\ \mu\text{m}$, a pH_{pzc} value of 7.1, and surface area $900\ \text{m}^2\text{g}^{-1}$ were purchased from Sigma Co., Germany. Strontium chloride ($\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, 99.9%, Wako Co., Japan) was used as a basis of Sr^{2+} solution for assessing the Sr^{2+} sorption dimensions of the composites (Shubair et al. 2019). For preparing the magnetic particles, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (99%, Junsei Chemical Co., Japan), NaBH_4 (98%, Sigma-Aldrich Inc., USA), and CuCl_2 (99.9%, Aldrich Inc., USA) were used. For this research, all chemicals were procured, confirming analytical purity. Suspensions and solutions were made by using deoxygenated deionized water. The nZVI-Z composite was made by following the technique that was first introduced by Wang et al. (2010). In short, 0.5 g of zeolite was mixed with 2.5 g of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ in 100 mL of deoxygenated deionized water for making the first solution mentioned in equation 1. Using ultrasound, the liquid-solid mixture was processed for 10 min and then blended for 30 min at ambient temperature. The mixture (1st solution) was then put in a four-necked container, and 50 mL of 0.58 M NaBH_4 solution (2nd solution) was thoroughly pumped at 1.2 L/h with intense stirring (~ 400 rpm) and a stable temperature of $30\ ^\circ\text{C}$. The resultant mixture was agitated for an extra 5 minutes as an aging period after adding NaBH_4 . The generation of nZVI particles (Fe^0) was the result of a redox reaction in which NaBH_4 (reducing agent) electrons were supplied to Fe^{3+} as per equation 2.

The solid products were separated from the aqueous process by centrifugation at 3500 rpm for 60 minutes and then consecutively cleaned with deoxygenated deionized to eliminate remaining salts. Similarly, the $n\text{Fe}/\text{Cu}-\text{Z}$ composite was made as mentioned above and following the equation 3.

2.2 Characterization of adsorbents

The crystalline structure of the nanocomposites was investigated by X-ray diffraction (XRD) applying Cu $K\alpha$ radiation at 0.15418 nm with a tube voltage of 40 kV and a current of 40 mA. All data were taken within the range of scanning speed of min^{-1} .

2.3 Strontium (Sr^{2+}) adsorption experiments

Adsorption experimentations were performed at 298 K through batch technique. The adsorption capabilities of both nanocomposites for Sr^{2+} were investigated through aqueous solutions of this metal. The standard solutions of Sr^{2+} were formed in deoxygenated deionized water by dissolving $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$. 0.2 g of nanocomposite was added into 100 mL of 100 mgL^{-1} initial Sr^{2+} concentration into a conical flask, and several identical conical flasks were prepared for the experimental measurements. The mixture was then wrapped and kept on a stirrer with magnets (RSH-6DN, As One Corporation, Japan) with a rotation speed of 1000 rpm. In these experiments, the primary pH level of the solution was kept at close to 6. The supernatants were filtered by a $0.2 \mu\text{m}$ PVDF syringe filter after the solutions were stimulated for 1 h to reach equilibrium. The concentration of Sr^{2+} was determined by inductively combined plasma mass analysis in the filtrate (ICPM-8500, Shimadzu Corporation, Japan). The impact of pH on the Sr^{2+} adsorption ability was investigated at different initial pH within the given range of 3 and 12. The adjustment of pH was managed by using volumes of 0.01-1.0 M HCl or NaOH. To identify the influence of dosage, experiments were conducted with different dosages of prepared nanocomposites ranging from 1 to 10 gL^{-1} . The experiments were also performed at various temperatures such as 298, 313, 328, and 343K for thermodynamic aspects. The kinetic of Sr^{2+} adsorption on both the nanocomposites was investigated at various initial Sr^{2+} concentrations such as 1, 5, 50, 100, 150, and 200 mg/L . The concentration of Sr^{2+} was determined for the different time periods ranging from 5 min to 60 min. To measure the equilibrium adsorption isotherm, experiments were conducted with an initial concentration of Sr^{2+} in the given range of $1\text{-}1000 \text{ mgL}^{-1}$. All measurements were replicated three times, and the average value was taken into account for analysis.

The Sr^{2+} adsorption capability was measured by applying the following equation:

$$q_e = (C_{\text{initial}} - C_{\text{equilibrium}}) \times (V / m) \quad (4)$$

The coefficient of distribution was measured by considering the following equation:

$$K_d = (C_{\text{initial}} - C_{\text{equilibrium}}) / C_{\text{equilibrium}} \times (V / m) \quad (5)$$

Where q_e (mgg^{-1}) represents the quantity of Sr^{2+} ions adsorbed, C_{initial} (mgL^{-1}) and $C_{\text{equilibrium}}$ (mgL^{-1}) denote the initial and equilibrium concentration of Sr^{2+} , respectively; V (L) represents the volume of the aqueous solution, and m (g) denotes the amount of composites.

2.4 Response surface methodology

Response surface methodology (Bingöl 2011; Kumari and Gupta 2019; Abdulgader et al. 2020) is used to recognize the optimum combination of the factors and their interactions with minimum effort, which is not possible in the traditional statistical analysis. Levels for different factors are assigned in Table 1. The value of high and low levels is considered according to some primary experiments.

Table 1. Factors with notations and levels in response surface methodology.

Factors (units)	Notation of factors	Levels		
		-1	0	1
pH (-)	A	6	9	12
Contact time (min)	B	5	17.5	30
Initial concentration (mgL^{-1})	C	100	150	200

Besides, it generates mathematical models and some critical plots. A few numbers of experiments are used to discover a lot of information. If nonlinear relation exists, then a polynomial function containing quadratic terms is required, which is executed in this analysis. In this analysis, response surface methodology based on the central composite design is used. Second-order polynomial model using response surface methodology is given in the following equations-

$$y = \gamma_0 + \sum_{i=1}^k \gamma_i x_i + \sum_{i=1}^k \gamma_{ii} x_i^2 + \sum_{1 \leq i < j}^k \gamma_{ij} x_i x_j + \varepsilon \quad (6)$$

Where y represents the outcome variable, γ_0 denotes constant, γ_i represents the linear coefficient, γ_{ii} denotes the quadratic coefficient, γ_{ij} represents the interaction coefficient, x_i and x_j is the coded coefficient, and ε is the random error term.

2.5 Adsorption isotherm model and error evaluation function

It is imperative to fit the isotherm models to know the maximum adsorption capacity, adsorption behavior, and its mechanism. In this study, five popular isotherm models, such as Tóth, Langmuir, Freundlich, Dubinin-Astakhov (D-A), and Hill, are fitted with experimental results.

2.5.1 Tóth model

In both the low and high concentration areas, the Tóth model describes the adsorption mechanism properly (Pal et al. 2016, 2017). It explains well in many adsorption systems with the adsorbent's sub-monolayer coverage. Tóth model is obtained by adding a heterogeneity parameter t in the Langmuir model. The following equations can be used to express it-

$$\frac{W}{W_0} = \frac{b_0 e^{\frac{Q}{RT}} \cdot C_e}{\left(1 + \left(b_0 e^{\frac{Q}{RT}} \cdot C_e\right)^t\right)^{1/t}} \quad (7)$$

where, W is instantaneous removal (mgg^{-1}), W_0 is maximum removal (mgg^{-1}), C_e denotes equilibrium concentration (mgL^{-1}), b_0 represents equilibrium constant (Lmg^{-1}), Q represents isosteric heat (Jmol^{-1}), R represents universal constant ($\text{Jmol}^{-1}\text{K}^{-1}$). Parameter t is meant to explain the system heterogeneity.

2.5.2 Langmuir model

The monolayer adsorption process is characterized by the Langmuir isotherm model. This model assumes that all energy sites on the adsorbent surface are homogeneous and there is no adsorbent-adsorbate interactions (Pal et al. 2017) and presented as,

$$\frac{W}{W_0} = \frac{K_L C_e}{1 + K_L C_e} \quad (8)$$

where W is instant removal (mgg^{-1}), W_0 is maximum removal (mgg^{-1}), K_L indicates Langmuir constant (Lmg^{-1}), and C_e indicates equilibrium concentration (mgL^{-1}).

2.5.3 Freundlich model

Freundlich isotherm is the most basic model that describes reversible and non-ideal adsorption. For understanding the adsorption mechanism, this model is described as follows (Pal et al. 2017).

$$W = K_f C_e^{1/n} \quad (9)$$

where, W is strontium removal (mgg^{-1}), K_f indicates Freundlich constant ($\text{mgg}^{-1})(\text{mgL}^{-1})^{-(1/n)}$, and n represent the heterogeneity constant. Henry's law is a special case of the Freundlich isotherm model when $n = 1$.

2.5.4 Dubinin-Astakhov (D-A) model

The (D-A) isotherm model (Rocky et al. 2019) is used by the researchers because it can accurately estimate the adsorption quantity for different adsorption pairs. Generally, this model is used for a high degree of the heterogeneous adsorbent. The D-A model is slightly modified for removal application by replacing pressure (P) with equilibrium concentration (C_e) and saturation pressure with strontium density (ρ_c). It can be stated as,

$$\frac{W}{W_0} = \exp \left[- \left(\frac{RT}{E} \ln \left(\frac{\rho_c}{C_e} \right) \right)^n \right] \quad (10)$$

Where the parameters n and E indicate heterogeneity factor (-) and characteristic energy (Jmole^{-1}), respectively. R and T represent the universal constant ($\text{Jmole}^{-1}\text{K}^{-1}$) and K , correspondingly.

2.5.5 Hill model

In order to explain the attaching of different species to homogeneous substrates, the Hill equation (Rahman et al. 2019) is used. It can be presented as follows-

$$\frac{W}{W_o} = \frac{\left(\frac{\rho_c}{C_e}\right)^{n_H}}{K_D + \left(\frac{\rho_c}{C_e}\right)^{n_H}} \quad (11)$$

Here, K_D is Hill constant, n_H indicates binding interaction coefficient, C_e and ρ_c denote equilibrium concentration (mgL^{-1}), and strontium density (mgL^{-1}).

2.5.6 Root mean square deviation (RMSD)

Researchers used RMSD frequently to estimate errors between experimental data and a model's expected value (Rahman et al. 2018), which can be stated as,

$$RMSD = \sqrt{\frac{\sum_{i=1}^n (W_{\text{experimental}} - W_{\text{calculated}})_i^2}{N}} \quad (12)$$

In this analysis, a generalized reduced gradient (GRG) nonlinear optimization technique was used to optimize the parameters using the RMSD error function. The RMSD follows the normal distribution, which is the required assumption of the ordinal least square method.

3. Results and discussion

Response surface methodology was considered to evaluate the importance of the main factors pH, contact time (min), and initial concentration (mgL^{-1}), and their interaction effects on strontium removal. The removal of strontium from an aqueous solution is regarded as the outcome variable in this study.

3.1 Characterization of nanocomposites

Fig. 1(a) shows the XRD pattern of zeolite, which discovered key characteristic 2θ peaks at 28.74° , 29.35° , 29.48° , 31.63° , 36.60° , 36.66° , 43.53° and 47.57° belonging to SiO_2 and 2θ peaks at 12.50° , 26.18° , 29.33° , 31.89° , 34.53° , 34.81° , and 36.62° correspond to the presence of Al_2O_3 (Amen et al. 2017). Fig. 1 (b) and Fig. 1(c) show the XRD pattern diffraction peaks of nZVI and nFe/Cu at $2\theta = 44.60^\circ$, 64.68° and 82.35° , indicating the presence of pure iron metal (Amen et al. 2018). Fig. 1(d) and Fig. 1(e) show the XRD pattern of nZVI-Z, and nFe/Cu-Z composites, the crystal structure of iron and zeolite changes were detected in their XRD analysis. Following the

iron loading on zeolite, the diffraction peaks displayed a lower intensity compared to pure iron and zeolite. This meant that the crystal structure of zeolite and iron was affected in the period of the synthetic process of the nanocomposites.

Fig. 1. XRD patterns of (a) zeolite, (b) nZVI, (c) nFe/Cu composite (d) nZVI-Z composite (e) nFe/Cu-Z composite.

3.2 Normal plot of standardized effect

The normal plot presented below for the strontium removal onto two different adsorbents. It determines the importance, direction, and magnitude of the effects. The plot depicted in Fig. 2

displays the standardized effects comparative to a distribution fit line for the case when all the effects on strontium removal are zero. The standardized effects are measured based on t-statistic, considering the impact of all factors is zero as a null hypothesis.

Fig. 2. Probability graph for (a) nZVI-Z and (b) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

Each point on the plot denotes the effect of factor, and effects which are distant from zero are more potential compared to the adjacent one. Statistically significant and insignificant factors are presented by different colors and shapes of the points. The points placed on the right hand side and left hand side of the zero lines indicate the positive and negative effects on the strontium removal, respectively. It is evident from Fig 2(a) for adsorbent nZVI-Z, that the point indicating initial concentration (C) has the highest chance of statistically significant positive effect on strontium removal followed by contact time (B), pH (A), and the interaction between contact time and concentration (BC) at 5% level of significance. This meant that the probability of removal of strontium increases when the process changes from low level to high level of the associated factors.

The quadratic effect of pH, time, and initial concentration, on the other hand, has a statistically significant negative impact on the removal of strontium. Furthermore, only the BC has a significant positive effect, whereas the other two interactions, AB and AC, are insignificant. Except for the interaction effect BC (contact time and concentration) in Fig. 2(b), similar outcomes are found for the nFe/Cu-Z. The effect of pH is high than the interaction effect BC for the adsorbent nZVI-Z, whereas the reverse is true for the nFe/Cu-Z.

3.3 Main and interaction effect

The main and interaction effect plots for every factor in the optimization process were used to understand the mean differences of the output variable of each factor. The main effect plot expressed in Fig. 3(a) illustrates a positive relationship between adsorption of strontium removal and each of the three factors, meaning that removal increases when the factors (pH, contact time, and concentration) level increases from the lower level to the upper level. By comparing the three graphs expressed in Fig. 3(a), it is found that the concentration curve is sharper than other factors, meaning that concentration is a more influential factor than others. Fig. 3(b) represented the interaction effect, as the lines are not parallel for the interaction between contact time and initial concentration (BC) on the removal of strontium. Interaction occurs when one factor produces a different effect on the response (strontium removal) at different levels of another factor. The existence of the interaction effect designates the necessity of a multi mechanism technique for the removal process. The lines are parallel for other interactions (pH and contact time) and (pH and concentration), suggesting no interaction effect on the removal process.

Fig. 3(a). Main and (b) interaction effect graph for the nZVI-Z ($\alpha = 5\%$).

Similar findings are found for nFe/Cu-Z composite portrayed in Fig. 4(a) and Fig. 4(b).

Fig. 4(a). Main and (b) interaction effect graph for the nFe/Cu-Z ($\alpha = 5\%$).

3.4 Contour plot for the optimum area of strontium removal

The contour plot is used to observe the association between predictor variables and regressed variables. It gives an idea of a two-dimensional view so that all points that have a similar response are related to producing contour lines of constant responses. It comprises three elements, i) predictors on the x-axis and y-axis, ii) contour line that connects points having the same response and, iii) contour bands of the same color indicate ranges of the response variable. The contour plot presented in Fig. 5 illustrates the relationship between factors (contact time, initial concentration, and pH) used to remove strontium from wastewater and the amount of strontium removal for adsorbent nZVI-Z. The darker region represents higher removal. This higher removal seems to form the upper right corner of the graph, which is the expected region. On the other hand, the region of the lower left corner of the graph represents the lower removal process, which is not

desirable. Curvature within the contour plot also assures that contact time and initial concentration has interacted. A similar pattern is observed in Fig. 6 for nFe/Cu-Z.

Fig. 5. Contour graph of strontium removal for the nZVI-Z ($\alpha = 0.05$) [Hold values: Concentration 150 (mg/L), time 17.5 (min) and pH 9, respectively].

Fig. 6. Contour graph of strontium removal for the nFe/Cu-Z ($\alpha = 5\%$) [Hold values: Concentration 150 (mg/L), time 17.5 (min) and pH 9, respectively]

3.5 Factors explain the most variability of the strontium removal

Fig. 7. Pareto chart of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

The Pareto chart clarifies whether the variables are statistically significant or not. It also helps to identify the most potential factor of the removal process. The perpendicular red dashed line in Fig. 7 indicates the reference line, and the effects are statistically significant if it falls on the right side of the reference line. According to Fig. 7, all effects except interaction AC and AB are statistically significant for both composites, and initial concentration has the most potential impact on strontium removal followed by contact time, pH, and interaction BC.

3.6 Second-order model for the strontium removal

For the rigorous analysis of the strontium removal, a nonlinear regression technique and the analysis of variance (ANOVA) have been conducted using the central composite design (CCD). Table 2 displayed the estimated coefficient and their standard error, t-value, and p-value for each main effect, quadratic effect, and interaction effect for both adsorbents.

From Table 2, it can be shown that the main effects pH, contact time, and initial concentration are statistically significant ($p\text{-value} < 0.05$) at 5% level, which means the mean strontium removal at two different levels are not equal. Moreover, the square terms of the main effects are statistically significant, meaning that the association between the factor and the strontium removal follows a nonlinear relationship. Besides, the statistically significant interaction effect BC (contact time and initial concentration) indicates the influence of initial concentration on strontium removal is different at different levels of contact time, which coincides with the results obtained in the normal

plot and main and interaction effects plot for both the adsorbent. On the contrary, the interaction effects (AB and AC) are not statistically significant.

The codified mathematical model of response surface method considering three factors are given below:

$$\tau = \lambda_0 + \lambda_1 A + \lambda_2 B + \lambda_3 C + \lambda_4 A^2 + \lambda_5 B^2 + \lambda_6 C^2 + \lambda_7 AB + \lambda_8 AC + \lambda_9 BC + \varepsilon \quad (13)$$

where, τ represents average removal of strontium, λ_0 is the overall mean, λ_i indicates the regression coefficients, and A, B and C represents pH, contact time (min) and initial concentration (mg/L), respectively. Based on the calculated regression coefficient for nZVI-Z adsorbent in presented Table 2, the subsequent model is given as,

$$\tau = 24.656 + 2.2003A + 2.7777B + 5.9405C - 0.0365A^2 - 0.0341B^2 - 0.0353C^2 + 0.0016AB - 0.0024 AC + 1.5550BC + \varepsilon \quad (14)$$

The final model presented in equation 15 ignoring the insignificant interaction term AB and AC is given as,

$$\tau = 24.656 + 2.2003A + 2.7777B + 5.9405C - 0.0365A^2 - 0.0341B^2 - 0.0353C^2 + 1.5550BC + \varepsilon \quad (15)$$

Similarly, for nFe/Cu-Z, the model can be given as,

$$\tau = 23.5142 + 0.8961A + 3.4769B + 5.9246C - 0.04934 A^2 - 0.04639 B^2 - 0.04757 C^2 + 2.07333 BC + \varepsilon \quad (16)$$

387 Table 2. Response surface regression: strontium removal versus pH, Time, Concentration.

Factor	nZVI-Z			nFe/Cu-Z		
	Estimate (std. error.)	t-value	p-value	Estimate (std. error)	t-value	p-value
Intercept	24.656 (0.0054)	4535.46	1.8×10^{-13}	23.5142 (0.0055)	4260.20	1.9×10^{-12}
A	2.2003 (0.0036)	555.44	1.7×10^{-8}	0.89618 (0.00366)	244.72	1.6×10^{-9}
B	2.7777 (0.0036)	770.12	1.9×10^{-13}	3.47699 (0.00366)	949.46	1.8×10^{-12}
C	5.9405 (0.0036)	1646.97	1.4×10^{-10}	5.92460 (0.00366)	1617.83	1.3×10^{-10}
A²	-0.0365 (0.0035)	-10.41	1.1×10^{-2}	-0.04934 (0.00356)	-13.84	1.2×10^{-2}
B²	-0.0341 (0.0035)	-9.74	1.1×10^{-2}	-0.04639 (0.00356)	-13.01	1.6×10^{-3}
C²	-0.0353 (0.0035)	-10.07	9.3×10^{-2}	-0.04757 (0.00256)	-13.34	9.4×10^{-3}
AB	0.0016 (0.0047)	0.35	0.725	0.00083 (0.00478)	0.17	0.8620
AC	-0.0024 (0.0047)	-0.51	0.610	-0.00000 (0.00478)	-0.00	0.999
BC	1.5550 (0.0047)	329.96	2.0×10^{-3}	2.07333 (0.00478)	433.32	1.9×10^{-3}

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3.7 Analysis of variance (ANOVA)

Analysis of variance is the process of distributing total variation contained in a data set into two components, which are assignable causes of variation and unassignable causes (error) of variation. ANOVA is used in this study to assess whether or not the average effects of different variables at different levels are equal. A comparison of the strontium removal at different levels of factors identifies the most influential factors and their effects. Table 3 displayed an adjusted sum of squares, the adjusted mean sum of squares, F-value, and P-value, and P-value less than 0.01 indicates the effect is statistically significant at 1% level. Both main and interaction effects, excluding the AB and AC interaction effects, have been shown to be statistically significant. Results indicated that the sum of square (SS) of factor C is the highest (1445.86 and 1438.11 for nZVI-Z and nFe/Cu-Z, respectively), which indicates initial concentration is the most explainable variable followed by contact time and pH for strontium removal for both adsorbents. Lack of fit for both adsorbents is insignificant, which means the model does not suffer from a lack of fit and no need to introduce higher order terms in the analyzed model. The high R^2 value for both adsorbents indicates the model fits well.

Table 3. Optimization of factors by using ANOVA technique.

nZVI-Z					nFe/Cu-Z		
Factor	df	SS (MS)	F-value	p-value	SS (MS)	F-value	p-value
Linear effect							
A	1	164.45 (164.45)	308510.62	2.1× 10 ⁻⁸	32.91 (32.91)	59888.01	2× 10 ⁻¹¹
B	1	316.13 (316.13)	593082.62	2.0× 10 ⁻¹⁶	495.31 (495.31)	901476.55	2× 10 ⁻¹²
C	1	1445.86 (1445.86)	2712501.57	2.3× 10 ⁻¹⁰	1438.11 (1438.11)	2617381.9	2× 10 ⁻¹²
Quadratic effect							
A ²	1	0.06 (0.06)	108.31	2.1× 10 ⁻¹⁰	0.11 (0.11)	191.52	2× 10 ⁻³
B ²	1	0.05 (0.05)	94.78	2.0× 10 ⁻⁹	0.09 (0.09)	169.33	2× 10 ⁻⁴
C ²	1	0.05 (0.05)	101.43	2.2× 10 ⁻¹¹	0.10 (0.10)	178.04	2× 10 ⁻³
2-Way interaction							
AB	1	0.00 (0.00)	0.13	0.725	0.000 (0.000)	0.03	0.862
AC	1	0.00 (0.00)	0.26	0.610	0.000 (0.000)	0.00	1.00
BC	1	58.03 (58.03)	108871.72	2.2× 10 ⁻¹¹	103.17 (103.17)	187769.87	2× 10 ⁻¹²
Error	50	0.003 (0.00)			0.000 (0.000)		
Lack-of-Fit	5	0.00 (0.00)	0.53	0.751	0.00 (0.00)	0.30	0.909
Pure Error	45	0.03 (0.00)			0.03 (0.00)		
R ² = 100.00% and R ² (adj) = 98.97%					R ² = 100.00% and R ² (adj) = 99.68%		

3.8 Validation of assumption

3.8.1 Residuals versus fits plot

The required assumption of the distribution, such as randomness and constant variance of residuals, can be verified by using the residual versus fit graph presented in Fig. 8. As the residuals are randomly scattered on both sides of the zero line, and there is no recognizable pattern of the residuals, we can conclude that the model meets the given assumptions.

Fig. 8. Residual vs. Fits plots of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent.

3.8.2 Residuals versus order plot

The assumption of independent residuals from one another can be checked by the residual versus order plot presented in Fig. 9 for the strontium removal process. Residuals in time order did not show any pattern, and it falls randomly around the centerline for both adsorbents that indicates the residuals are independent of one another.

Fig. 9. Residual vs. order plots of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent.

3.8.3 Normal probability plot

The assumption of normally distributed residuals can be verified by using the normal probability plot. The normal plot of the residuals for strontium removal using two different nanocomposites is portrayed in Fig. 10. The residuals showed a more or less straight line meaning that residuals meet the normality assumption.

Fig. 10. Normal plots of residuals for a) nZVI-Z and ii) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

3.9 Response optimization

The optimization plot displays the effect of factors on the predicted responses (strontium removal). It identifies i) optimum settings of the predictors and ii) point estimate and prediction interval of the responses. Response optimization plots of strontium removal for two different nanocomposites are displayed in Fig. 11. The current factor settings are pH = 14.0454, contact time = 38.5224 minutes, and initial concentration = 234.0896 (mg/L). The goal of this study was to maximize the removal of strontium. Its predicted value is 46.7847, and the 95% prediction interval (PI) is (46.7099, 46.8596) for the nZVI-Z adsorbent. Narrower prediction interval (PI) indicates a more precise prediction of the strontium removal.

(a)

(b)

Fig. 11. Response optimization plots of strontium removal for a) nZVI-Z and b) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

3.10 Isotherm model fitting

In various studies, few isothermal models such as Freundlich and Langmuir have been applied to remove the radioactive materials from wastewater (Piccin et al. 2017; Eljamal et al. 2019; Shubair et al. 2019; Xanthopoulou et al. 2021). In this study, five isotherm models are fitted with experimental results, and the best isothermal model is diagnosed. The relation between the removal of strontium and concentrations is presented in Fig. 12. It can be stated that the concentration of strontium is positively associated with strontium removal.

Fig. 12. Sorption isotherm of models onto (a) nZVI-Z and (b) nFe/Cu-Z using RMSD. Fixed parameter: dosage = 5 gL⁻¹, pH = 6 and temperature = 298 K).

Table 4 displays the estimated parameters and RMSD values of the fitted models. According to the values of R^2 , it can be stated that strontium removal is adequately described by these models. Results suggested that the Tóth model is lower than that of other models based on RMSD value, suggesting that the Tóth model performs better than other models. The value of n used in the Freundlich model is greater than unity, meaning adsorption increases with concentration increases. Based on W_0 and K_f , it is mentioned that the sorption capability of the nFe/Cu-Z adsorbent is greater than that of nZVI-Z.

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Table 4. Parameter estimation of different isotherms models.

Model	Adsorbent	Parameter					
Tóth		W_0 (mgg ⁻¹)	b_0 (Lmg ⁻¹)	Q (Jmol ⁻¹)	t (-)	R^2	RMSD
	nZVI-Z	85	7.55E-05	15909.22	0.726	0.9957	3.947
	nFe/Cu-Z	89	8.69E-05	13821.74	0.812	0.9978	6.984
Langmuir		W_0 (mgg ⁻¹)	K_L (Lmg ⁻¹)				
	nZVI-Z	84.12	5.21E-05				
	nFe/Cu-Z	88.74	5.23E-05				
Freundlich		K_f (mgg ⁻¹)(mgL ⁻¹) ^{-(1/n)}	n (-)				
	nZVI-Z	2.03	1.52				
	nFe/Cu-Z	2.05	1.46				
D-A		W_0 (mgg ⁻¹)	E (Jg ⁻¹)	n (-)			
	nZVI-Z	82	338.64	7	0.972	7.109	
	nFe/Cu-Z	83	336.30	7.21	0.982	7.186	
Hill		W_0 (mgg ⁻¹)	n_h	K_D			
	nZVI-Z	85	0.75	0.000356	0.976	4.4911	
	nFe/Cu-Z	88	0.76	0.00046	0.982	8.0770	

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494 **4. Conclusions**

495 To explore the impact of different factors and their interaction effects on the efficacy of
 496 strontium removal, response surface methodology based on the central composite design was used
 497 to identify the optimal conditions for removing strontium from contaminated water using iron-
 498 based nanoparticles-zeolite. The main findings can be summarized as follows:

- 499 1) The nanocomposite nZVI-Z and nFe/Cu-Z are efficient adsorbents to extract strontium
 500 from contaminated water.
- 501 2) Regression results showed that initial concentration, contact time, and pH have a
 502 statistically significant impact on the process of removing strontium, and concentration
 503 was found to be the most potential factor followed by contact time for this removal
 504 process.

3) The statistically significant effect of the interaction between the initial concentration and contact time was observed and suggested that the impact of the initial concentration on the removal of strontium differs at different contact time levels. Subsequently, the chemicals used in experimental work and research costs may be reduced.

4) Tóth, Langmuir, Dubinin-Astakhov (D-A), Freundlich, and Hill models were fitted with the removal data and observed that the Tóth model performed very well, followed by Hill and D-A model based on RMSD. The fitting of Tóth, Dubinin-Astakhov (D-A), and Hill models certainly contributes to existing literature, as only Langmuir and Freundlich models are available in previous studies.

The findings may contribute to developing an efficient strontium removal system from wastewater. It is an emerging issue worldwide, particularly in Japan, after the destruction of the Fukushima Daiichi Nuclear Power Plant.

Declarations

Ethics approval and consent to participate

Not application

Consent for publication

Not applicable

Availability of data and materials

All data used and analyzed in this study are available from the corresponding author on reasonable request.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

Shamal Chandra Karmaker: Conceptualization, Formal analysis, Methodology, Writing-original draft. Osama Eljamal: Investigation, Experiments, Writing-review & editing. Bidyut Baran Saha: Conceptualization, Methodology, Writing-review & editing, Supervision.

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Figures

Figure 1

XRD patterns of (a) zeolite, (b) nZVI, (c) nFe/Cu composite (d) nZVI-Z composite (e) nFe/Cu-Z composite.

Figure 2

Probability graph for (a) nZVI-Z and (b) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

Figure 3

(a). Main and (b) interaction effect graph for the nZVI-Z ($\alpha = 5\%$).

Figure 4

(a). Main and (b) interaction effect graph for the nFe/Cu-Z ($\alpha = 5\%$).

Figure 5

Contour graph of strontium removal for the nZVI-Z ($\alpha = 0.05$) [Hold values: Concentration 150 (mg/L), time 17.5 (min) and pH 9, respectively].

Figure 6

Contour graph of strontium removal for the nFe/Cu-Z ($\alpha = 5\%$) [Hold values: Concentration 150 (mg/L), time 17.5 (min) and pH 9, respectively]

Figure 7

Pareto chart of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

Figure 8

Residual vs. Fits plots of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent.

Figure 9

Residual vs. order plots of strontium removal for a) nZVI-Z and ii) nFe/Cu-Z adsorbent.

Figure 10

Normal plots of residuals for a) nZVI-Z and ii) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

(a)

(b)

Figure 11

Response optimization plots of strontium removal for a) nZVI-Z and b) nFe/Cu-Z adsorbent ($\alpha = 5\%$).

Figure 12

Sorption isotherm of models onto (a) nZVI-Z and (b) nFe/Cu-Z using RMSD. Fixed parameter: dosage = 5 gL⁻¹, pH = 6 and temperature = 298 K).