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Sustainable Method to Treat Fluoride using Rice Husk: RSM Optimization Via JMP Analysis

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Abstract

Since only one percent of the world's water supplies are immediately usable, freshwater availability poses a serious threat to human well-being and global development. Unfortunately, forty percent of the world's population currently lives in arid or semiarid areas, and by 2025, that percentage is expected to rise to more than two-thirds. At the same time, some of the most pressing issues of our time are the rapid growth of industrialization, increasing pollution, and environmental degradation. The ecosystem benefits from the abundance of resources on Earth, which are also necessary to preserve public health. This work aims to use response surface methodology (RSM), a potent statistical and mathematical technique, to model and enhance the factors controlling fluoride elimination. The study investigated the fluoride removal capacity of natural adsorbents, such as rice husks, due to their wide surface area and porous nature. Doses of 1 to 12 g/L of rice husk were used to remove 67% of the fluoride. pH 2 and dosages of 1 g/L and 6.5 g/L produced the best outcomes. The stirring rate was 10 minutes of rapid mixing at 100 rpm after 30 minutes of gentle mixing at 40 rpm throughout a contact period of 20 to 180 minutes. These findings suggest that rice husks can be employed as cost-effective, environmentally benign, and efficient adsorbents to extract fluoride from groundwater. A workable solution to guarantee the production of safe drinking water while reducing the health risks associated with excessive fluoride consumption is to use these natural resources.

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Introduction

Water is an abundant Natural assets that is necessary for maintaining life and the ecosystem, as we have long understood.[1] it has been recognized as an essential natural resource and a basic human need, contributing significantly to increased life

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expectancy and the advancement of humanity One of the most frequent ions in drinking water is fluoride, that depending on its concentration and consumption levels, can either be advantageous or harmful to human health.[2] Sellaite (MgF_2), fluorspar (CaF_2), cryolite (Na_3AlF_6), and fluorapatite [$_3Ca_3(PO_4)_2$ Ca (F, Cl_2)] are the major minerals that contain fluoride. The proper level of fluoride in drinking water reduces the incidence of cavities and protects teeth from deterioration. Consequently, fluoride compounds will only be present in groundwater when optimal conditions for their decomposition occur or when huge quantities of fluoride-containing industrial effluent are released into lakes and rivers.[3,4] The article discusses diseases associated with excessive fluoride intake and explores various methods used for defluorination.[5] These companies' waste water range in fluoride concentration from ten to thousands of mg/L, which is higher than that found in pure waters. Over 200 million people globally are thought to depend on drinking water with fluoride materials above the WHO guideline of 1.5 mg/L.[6] For nations with hot, dry climates, the maximum fluoride concentration in drinking water may not be appropriate.[7] A nation that has largely affected by fluoride is India. In India, fluoride ions occur in highly populated regions. In accordance with EPA and WHO guidelines, fluoride levels should not exceed 1.5 mg/L.[8] Next, each fluoride agent's strength of recommendation was classified as strong, weak, or conditional using the grade system. a variety of problems might arise if the fluoride ion level exceeded 1.5 mg/l: bone shortage, enamel in tooth cavities, paralysis, gene mutation, stimulation of DNA/RNA synthesis, etc. In accordance with the World Health Organisations that want fluoride is accountable for roughly eighty percent of conditions carried on by polluted drinking water and about 65% of regional fluorosis[8]

Skeletal manifestation begins when the fluoride level rises to 6 mg/L daily. Because of the superior blood supply of cancellous bone as compared to the cortex, fluoride has a selected tendency to collect in cancellous (spongy) bones. The severe condition known as skeletal fluorosis poses a serious risk to people over 65. The causes of skeletal fluorosis include osteosclerosis, bone loss or osteoporosis, peri-osseous soft tissue swelling, and degenerative changes to cartilage and joints. The clinical basis of a variety of bone-forming diseases and essential aspects of the course of skeletal fluorosis include higher bone turnover and active bone growth. The biological cause of fluorosis at the molecular and genetic levels was the main topic of discussion at the The International Association for Fluoride Research held its 33rd Conference in India in 2016.9 Different methods for fluoridation, such as coagulation-precipitation, membrane separation procedures, ion exchange, adsorption techniques, and others (electro-dialysis and electrochemical), can be utilized for addressing the adverse impacts of fluorosis on human health.[10] All method experienced advantages and disadvantages, but under the right conditions, they all eliminated fluoride to a greater amount. Each of the above methods will be explained briefly in addition to its advantages and disadvantages.[11] Utilizing neglected plant material, such as maize leaves, Carica papaya timber, Lalang crushed teak leaves extracts, Coriandrum sativum, sago waste, peanut hull pellets, teak leaf powder, grapevine debris, husk ash, and neem bark, among others, was the primary objective of the adsorption studies being conducted by various.[12-14]

However, In order to improve the learning of the experimental methods, this study uses a statistical experimental design called Response Surface Methodology (RSM). Furthermore, RSM has proven to be a useful tool for creating and improving a range of experimental designs for food preservation, wastewater treatment, separation, and other important engineering fields.[15-18]

The novelty of the present work is to use RSM to assess how quantitative factors affect a response variable. RSM is known to be a potent tool for designing experiments to develop new processes, modify existing designs, and improve their efficiency. It is considered to be the most reliable statistical and empirical approach and a viable method for systematically adjusting each variable to evaluate fluoride removal effectiveness and to the development of a model to predict responses and to assess the components' quadratic, linear, and effects of interaction.

The purpose of this study is to create a forecasting model for eliminating fluoride using RSM. This will consider various independent factors like catalyst amount, contact time, pH, and fluoride concentration. JMP Pro 17 is used to customize the number of experiments.[13] set of the experiment will be conducted, each with different catalyst amounts, settling times, and pH levels.

Materials and Methodology

Collection of water Sample

The water sample used in this investigation was taken from a Pulladigunta, Guntur District, Andra Pradesh, India.

Preparation of Rice Husk Powder

cellulose, protein-rich foods, and respiration functions (carboxyl, hydroxyl, etc.) are enhanced in rice husk. The technique of desorption has become used in the past few years owing to the tremendous surface space and biochemical characteristics of materials that adsorb. The technique involves chemical as well as physical interactions (small molecules clinging to the top due to effects of gravitation and the power that oxygen has to interact with dangerous ions) and has an impact on the pH level, the temperature, and duration of contact. The goal of this research is to determine how well-available, affordable materials such as rice husk could remove fluorine from water. The recently harvested rice husk was certainly cleaned using distilled water and let to dry in the sun for a few days. After that, the sample was finely crushed and kept until further analysis in a pristine recyclable container.[19]

Experimental Design

Lower limits and upper limits range given in the JMP Pro 15 software was pH (2 to 12), dosage of rice husk powder (1 to 12 g/L), 20–180-minute time range. The glass jar test apparatus was utilized for carrying out the experiment. A second variable-speed motor enables each one of the six-bladed stirrers in the container of test apparatus to spin at various rates, between 0 to 150 rpm (Machine ability). After providing the water sample a thorough shake, the contents of it had been split and transferred partly to the jar test beakers. Three stages make up the experiment: ten minutes of fast mixing at 100 rpm, thirty minutes of gradual mixing at 40 rpm, and finally settling for a 20–180-minute time range at 30±2ºC. After the settling time, the settled part was disposed of, and the remaining material was removed and put into the flask for further examination. The average value of the quadriporate precipitate description was used to calculate the response.[21]

SEM analysis was applied in the present investigation for analyzing the morphology structure of rice husk. To evaluate its morphology structure, crushed rice husk was collected and sieved. The coagulant dose, medium pH, and initial fluoride value (independent variables) were modified using RSM (Response surface methodology is an empirical modeling technique that estimates the correlation between a collection of experimental variables that can be controlled and the results that are observed.) via Costume Design of Experiment in the Design Expert program in order to remove fluoride (dependent variable) using the bio-absorbent rice husk powder. Dependent variables were estimated utilizing an equation. The experimental run trails of 13—that comprise the pH, coagulant dosage, and settling time —were applied to the JMP DoE software.[3]

Table 1: Custom design trails-RSM method-JMP Pro 17

Trail No	pH	Coagulant Dosage (g/L)	Settling time (min)
1	8.4	9.14	20
2	7	6.5	100
3	12	1	20
4	7	1	180
5	7	12	180
6	2	1	20
7	2	12	66.4
8	12	12	100
9	12	6.5	180
10	7	1	100
11	8.4	9.14	20
12	7	6.5	100
13	2	6.5	180

Results and Discussion

Scanning and Electron Microscopy

SEM is an advanced imaging technique that provides high-resolution visualizations of material surfaces. By using a focused electron beam, SEM reveals topographical features, particle morphology, elemental composition, and crystallographic information. It plays a crucial role in material characterization, nanomaterials research, and understanding sample properties at the nanoscale. Proper sample preparation is essential for accurate SEM results. The FESEM technique, was used to identify the outermost morphology of the rice husk powder was studied. The image generated at 2000× magnification is displayed in Figure 2. It is possible to see pores on the rice husk's surface, which suggest possible places for particle adsorption.[10]

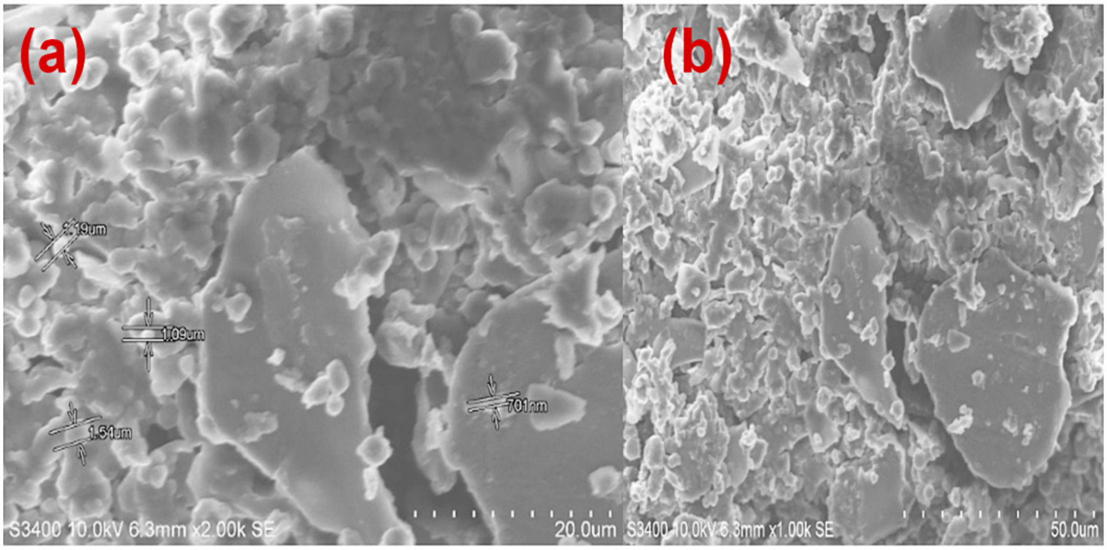

Fig 2 (a)&(b): FESEM microphotography of rice husk powder under 2000x magnification

Table 2: Percentage of fluoride removed by the rice husk in water sample

Trail No	pH	Catalyst Dosage (g\L)	Settling time (mins)	Final Fluoride values ("Absorbance Units" or AU)	Fluoride removal efficiency (%)
1	8.4	9.14	20	0.335	51%
2	7	6.5	100	0.306	55%
3	12	1	20	0.328	52%
4	7	1	180	0.292	57%
5	7	12	180	0.381	44%
6	2	1	20	0.284	59%
7	2	12	66.4	0.217	68%
8	12	12	100	0.319	53%
9	12	6.5	180	0.412	40%
10	7	1	100	0.314	54%
11	8.4	9.14	20	0.599	13%
12	7	6.5	100	0.318	54%
13	2	6.5	180	0.309	55%

After conducting trail 7, the optimal fluoride dosage was found to be 12 g/L. The results show that the ideal pH range is 2, and that rice husk powder has 68% fluoride reduction efficiency. The rate at which rice husk removed fluoride decreased significantly as pH increased from 2.0 to 12.[28]

Rsm and Graphical Representation

At each design step, fluoride removal was optimized using RSM analysis, which involves for three independent parameters and thirteen different trails. The percentages of fluoride removed are shown in table 2.

Responses on Reduction of Fluoride by using Rice Husk

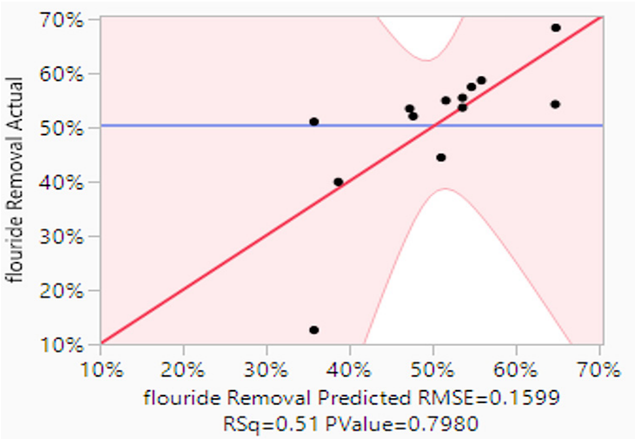

Fig 2: Predicted Vs Actual Removal efficiency of Fluoride by rice husk

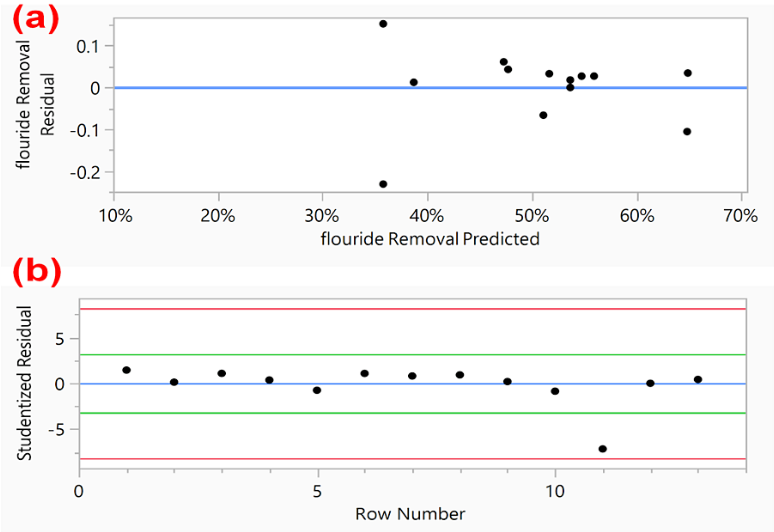

Fig 4: (a) and (b) Residual versus predicted graph Studentized residual versus row number

The R(square) value for fluoride reduction is approximately 0.999. Moreover, the model's suitability is evaluated using f-value, p-value, and accuracy. Root mean square error is 0.1599, and the probability value is 0.7980.

Individual limits are shown in green, and externally studentized residuals with 95% simultaneous ranges (Bonferroni) are shown in red.

The residual value indicates how much a data point deviates vertically from the regression line. While some data points may fall exactly on the line, others do not. Figure 14(a) displays a scatter plot of residual fluoride reduction versus predicted fluoride reduction. In most cases, the residual reduction values closely matched the planned reduction values. Since most of the data points align to create a straight line and the larger data points have residual values near zero, the line fit is seen as favorable. Furthermore, a strong correlation was observed, with a coefficient of determination, R(square) of 0.99905, between the predicted values and the experimental results. Figure 4(b) illustrates the relationship between the row number and the studentized residuals.[22]

Table 3: Analysis of Variance

Source	DF	Sum of Squares	Mean Square	F Ratio
Lack Of Fit	2	0.02841644	0.014208	0.3846
Pure Error	2	0.07389000	0.036945	Prob > F
Total Error	4	0.10230644		0.7222
				Max RSq
				0.6471

Source	Log worth	PValue	
Settling time*Settling time	0.518	0.30337	
PH (2,12)	0.470	0.33876	
Catalyst Dosage*Catalyst Dosage	0.310	0.48940	
Catalyst Dosage (1,12)	0.220	0.60292	^
Settling time (20,180)	0.207	0.62080	^
PH*Catalyst Dosage	0.156	0.69860	
Catalyst Dosage*Settling time	0.070	0.85054	
PH*Settling time	0.024	0.94601	

Fig 5: Quadratic components: Logworth and p-value of the respective parameter

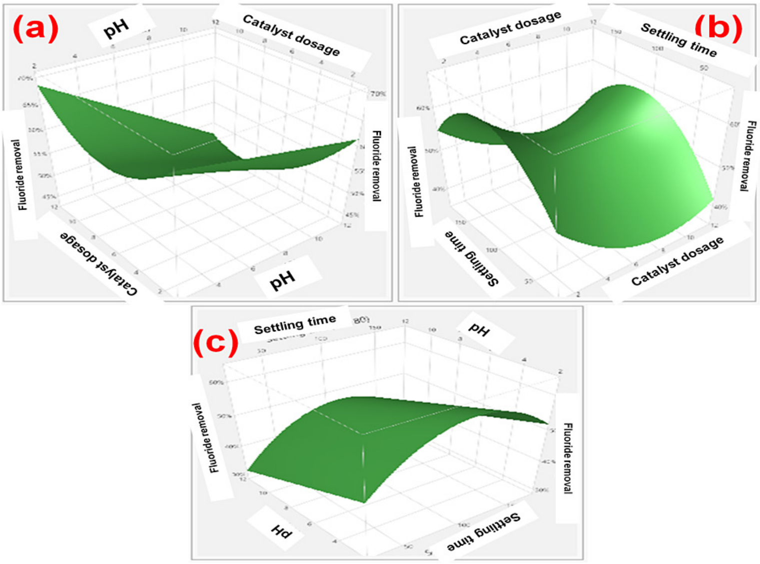

Fig 6: Response surface showing the effects of the independent variables on fluoride removal, a) effects of PH and catalyst dosage b) effects of catalyst dosage and settling time c) effects of settling time and pH

The constructed model's predictions' dependability was verified using an ANOVA-based study. ANOVA yielded significant statistical measures such as degrees of freedom (DF), sum of squares, mean square, F ratio, and p-value, with Table 3 providing more detailed information. Mean square values are derived by dividing the total squares of all components by the appropriate model degrees of freedom and the error variance. The model's relevance is assessed using the F-value and the p-value. A significant model in statistics is defined by a high p-value (0.7222) and a low F-value (0.3846). The model's fit accuracy is further supported by the coefficient of determination (R square). According to Figure 5 coefficient of determination, R(square) = 0.51, the suggested model can explain 50.90% of the variation in the outcome. Furthermore, the large coefficient of determination (R square) = 0.51 suggests the model's great validity. The model is considered suitable for this investigation since the regression coefficient is near to one, suggesting that there are little disparities between the actual and anticipated findings.[9, 20]

Table 3: Comparative study with earlier research work with rice husk

Adsorbent	Target	Efficiency (%)	Reference
Vetiveria Zizanioides	Fluoride	93%	1
rice husk	Fluoride	98%	8
Al/Fe oxides onto tea waste	Fluoride	87%	2
rice husk	Fluoride	67%	Present study

In the proposed model, Figure 5 illustrates the importance of every parameter. The LogWorth variation, represented as − log10(p-value), modifies p-values to form a suitable scale for visualization. A p-value below 0.01 typically provides strong support for the null hypothesis. The reference blue line corresponds to the Log Worth [log10(0.01)] at two. Consequently, if the Log Worth value of a parameter is less than two, it is considered significant. The factor that has the most significant impact on reducing fluoride is associated with a p-value of 0.33876 and a Log Worth value of 0.518. Other factors that need to be considered include the amount of catalyst used, the combination of pH and catalyst dosage, and the combination of pH and contact time. According to this research, the reduction of fluoride levels is mainly influenced by the quantity of catalyst used, the fluoride concentration, and quadratic factors such as pH × catalyst dosage and pH × contact time.[17]

In Figures 6(a), (b), and (c), you can see the response surface plots showing the relationship between pH and catalyst dosage, as well as catalyst dosage and settling time. Figure 6(a) illustrates the impact of pH and catalyst dosage on fluoride removal, indicating that both factors play a significant role. The graph demonstrates that increasing the pH from 2 to 12 led to a noticeable enhancement in fluoride reduction, suggesting that acidic conditions are optimal for using coconut fiber to remove fluoride. At a pH of 2, 68% of the fluoride was removed. In Figure 6(b), the lowest fluoride content (0.5 ppm) corresponds to the greatest reduction. The decrease in reduction percentage is noticeable as the concentration of fluoride increases. This could be due to the high concentration of fluoride molecules, which may be obstructing access to the active sites of the coconut fiber. The p-value calculated for the data in Fig. 11 suggests that fluoride reduction is more sensitive to changes in fluoride concentration. The highest reduction is achieved when the catalyst dosage is 12 g\L and the settling time is 66.4 minutes. Figure 6(c) illustrates the response surface plots of settling time versus pH. The correlation between the reaction time of 20–180 minutes is similar. Correspondingly, as the pH of the solution shifts from acidic to alkaline, the rate of fluoride reduction also declines.[21]

The study investigated the fluoride removal capacity of natural adsorbents, such as rice husks, due to their wide surface area and porous nature. Doses of 1 to 12 g/L of rice husk were used to remove 67% of the fluoride. pH 2 and dosages of 1 g/L and 6.5 g/L produced the best outcomes. The stirring rate was 10 minutes of rapid mixing at 100 rpm after

30 minutes of gentle mixing at 40 rpm throughout a contact period of 20 to 180 minutes. Table 3 shows the Comparative study with earlier research work with rice husk. Adsorption in acidic conditions can occur primarily due to electrostatic attraction between positively charged adsorbent surfaces (resulting from the presence of hydrogen ions in the acidic solution) and negatively charged adsorbate molecules, essentially acting like a magnet to draw the adsorbate to the surface.

Conclusion

The research evaluated the efficacy of natural absorbents, specifically rice husk, in removing fluoride from groundwater with the use of JMP software. The absorbent exhibited significant potential in adsorbing fluoride ions; rice husk demonstrated effectiveness across a broader pH range (2–12), at acidic condition that is PH 2 the fluoride removed efficiently. It was observed that factors such as temperature, stirring duration, pH, and adsorbent concentration have an impact on the efficiency of fluoride removal. In general, the results indicate the potential of rice husk as cost-effective and efficient biosorbent for treating fluoride-contaminated groundwater. Further optimization of these parameters using JMP software could enhance their application in practical water treatment systems, effectively addressing environmental and public health concerns associated with fluoride contamination. The potential applications are varied and include environmental remediation, cost-effective and eco-friendly water purification, incorporation into technology, and the improvement of community health and welfare. By employing bio-based remedies to protect the environment and improve human health, this approach addresses current water quality issues while also advancing sustainable development objectives.

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Ethics Statement

This research did not involve human participants, animal subjects, or any material that requires ethical approval.

Informed Consent Statement

This study did not involve human participants, and therefore, informed consent was not required.

Permission to Reproduce Material from other Sources

Not Applicable

Author Contributions

- **Rajashekara Rakshitha:** Conceptualization, Methodology, Writing – Original Draft– Review & Editing.
- **Doddapaneni Nitisha:** Data Collection, Analysis, Writing
- **Nagaraju Pallavi:** Visualization, Project Administration, Supervision.

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