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# BIOADSORPTIVE BEHAVIOUR OF *Cyperus rotundus* TOWARDS ALUMINUM EXTRACTION: COMPREHENSIVE ISOTHERM-KINETICS, THERMODYNAMIC ANALYSIS AND ANN, SVM, GPR PREDICTION

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**ABSTRACT**

Excessive aluminium concentrations in drinking water have detrimental effects on human health and aquatic ecosystems. While innovative chemical-based methods for aluminium extraction exist, they can be costly. Therefore, a more sustainable approach involves utilising readily available plant materials for aluminium removal. This study investigates the bioadsorption of aluminium using biomass from *Cyperus rotundus* roots (CRR), considering various parameters such as pH, biomass quantity, contact time, initial aluminium concentration, and temperature. The bioadsorption process exhibited an efficiency of up to 97.99% under optimal conditions, including pH 6, 3 g biomass, 90 minutes of contact, and a temperature of 25°C with agitation at 150 rpm. The adsorption process is found to be spontaneous, feasible, and exothermic. Batch adsorption experiments follow the Fritz-Schlunder (five-parameter) isotherm and pseudo-first-order kinetic model. Monolayer adsorption capacities are determined as 148.03 mg/g. Predictive models using Artificial Neural Network (ANN), Support Vector Machine (SVM) and Gaussian Process Regression (GPR) are developed, demonstrating strong abilities in forecasting aluminium removal percentages. The performance of ANN, SVM and GPR models is evaluated through statistical methods such as $R^2$ and MSE, with GPR outperforming ANN and SVM in prediction accuracy. For future applications of CRR bioadsorbents in industrial settings, comprehensive research is recommended, particularly involving real wastewater samples containing aluminium and/or other heavy metals.

**Keywords:** Aluminium Removal, Adsorption Kinetics, ANN, GPR, Non-Linear Regression, Plant-Based Bioadsorbent, SVM

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## INTRODUCTION

Rapid industrialization and urbanization have greatly worsened environmental damage to water bodies due to the discharge of toxic pollutants. Due to the metal pollutants' non-biodegradability, industrial effluents contaminated with heavy metals like Copper, Lead, Iron, Aluminium, Arsenic, Nickel, etc., are considered the most hazardous among chemically intensive industries. Electroplating, mining, tanning, fertilizer, battery manufacturing, pesticide manufacturing, and tanneries are major contributors to heavy metal pollution. Industries like aircraft, motor vehicles, construction of siding, manufacturing of lightweight utensils that use aluminium, and its alloys are recognized as major heavy metal industries, especially aluminium-releasing industries. Aluminium can dissolve in both basic and acidic environments. Aluminium salts are produced when aluminium compounds are broken down with acids. These salts are frequently used in water treatment applications to help coagulate pollutants like sediments, nutrients, and dissolved organic compounds.[2,3] Additionally, as a result of human activity and residence, the soil's aluminium leaches into nearby water bodies. As a result, the toxic Al (III) ions that accumulate in water bodies have an impact on agricultural production. Additionally, municipal water residual aluminium is undesirable.[4,5] Some parts of the states country such as Punjab, Andhra Pradesh, Tamil Nadu, Uttar


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Pradesh, Telangana, are reported to have the presence of aluminium and other heavy metals in excess of the permissible limits.[6-13]

The intake of aluminium by humans came primarily from food, water, and medications. According to epidemiological findings, Aluminium ions are neurotoxins[14], cause harm to fish, zooplankton[15], cyanobacteria[16], algae[17], and water weeds.[18] They also cause kidney issues, Parkinson's and Alzheimer's disease, bone softening[19], pulmonary fibrosis, and anemia. The removal of heavy metals from water and wastewater has become a topic of significant interest in recent years as a result of the stringent laws that have been enacted by various environmental protection agencies in developing countries.[20] Ion-exchange, reverse osmosis, and electro-dialysis are expensive and unlikely to be used in large-scale applications, while precipitation-based conventional methods are ineffective at low concentrations.[21,22] Adsorption is the most effective and widespread physicochemical method used to treat metal-contaminated wastewater. The price of the adsorbent is largely what determines this process's efficiency and economic viability. As a result, the search for an inexpensive and readily available adsorbent material begins. Number of agricultural waste by-products and plant-based adsorbents like *Typha domingesis* leaves powder[23], Curcuma longa powder[24], *Moringa millingotonia* stem and leaves powder[25], *Eucalyptus camaldulensis* bark[3], Rice hull activated carbon[26], Locust bean husk[27], *Cassia Occidentalis* stem powder[28], *Burea monosperma* and *Angel marmelo* [29]etc, have been utilized for the extraction of aluminium ions so far. Thus, use of biomaterials as adsorbents is interesting in this context due to their availability, affordability, little processing, high selectivity towards metal ions, effectiveness and easy regeneration. [30] Polymeric components, including lignin, cellulose, and hemicellulose (collectively called lignocellulosic), are found in plant biomass and its waste.[31] Plant biomass encompasses a wide array of additional components that constitute diverse functional groups, including water, hydrocarbons, lipids, ash, starches, simple sugars, proteins, extractives, and various other compounds. Illustrative functional groups observed in plant biomass and waste encompass amido and acetamido groups, carbonyl groups, phenolic groups, sulfydryl groups, esters, structural polysaccharides, and alcohols, etc.[32] Hence, these compounds could play a role in the binding mechanism of inorganic ions to plant biomass. Alongside their attraction and selectivity, the physicochemical characteristics of lignocellulosic biosorbents also influence their effectiveness in eliminating inorganic contaminants from wastewater.[33] *Cyperus rotundus* (L.) is commonly known as Nut grass or Nagarmotha or purple nutsedge. *Cyperus rotundus* has long been used as a home remedy to treat a variety of ailments.[34] In customary medicine, the tubers of this plant are utilised to treat irritation, Skin diseases, pain, fever, ulcers, pimples, burns and blisters, stomach and digestive illnesses, cholera, fix spasms, loose bowels, increase lactation and period anomalies, etc. It has numerous pharmacological and biological applications, including antiandrogenic and antimicrobial properties, antiallergic activity, antiarthritic activity, anticarcinogenic activity, antidiarrhoeal activity, and anti-inflammatory activity [35-41]. It was found effective in the removal of enteric pathogens from domestic wastewater.[42] It was used to carry out a toxicity test of tempe industrial wastewater, found to be tolerant to it by [43.] Significant reduction in TDS, TSS, BOD and COD was observed in a floating constructed wetland having *Cyperus rotundus* for sewage treatment.[44] It was also found to be effective in the phytoremediation of grey water.[45] Phytoremediation of the heavy metal contamination soil using roots and shoots of *Cyperus rotundus* was found to be effective. Restoration of the diesel-contaminated soil using *Cyperus rotundus* was observed by [41.] Extraction of the Fe, Zn, Cu, Pb, Cd and Cr from crude oil contamination soil was seen by.[46.] Decontamination of Zinc, Cadmium, nickel, and lead from soil was studied by.[47] The removal of As, Cd, Pb, Rb, Sn and Zn from soil using *Cyperus rotundus* phytoremediation was achieved by.[48] It can also be applied as an effective de-fluoridizing (from soil) agent.[49] It was found effective in the elimination of heavy metals like copper and zinc[50], lead[51,] etc., from aqueous solution. The dyes like crystal violet[52] and methylene blue[53] can be efficiently removed using *Cyperus rotundus.* The bioadsorption process is affected by a variety of factors, including surface area, agitation speed, pH, adsorbent dose, contact time, temperature, initial metal concentration, etc. It is challenging to carry out experimental research when all these factors are taken into consideration. It would also be impossible to experiment with the necessary number of groups or under the necessary circumstances; thus, it could be beneficial to develop a quick prediction approach.

As a result, a precise prediction model that might considerably reduce work and operational costs was created. An area of artificial intelligence (i.e., Machine learning techniques) may automatically improve the execution of computer algorithms developed for certain specific tasks. In machine learning research, training data—sample data collected from previously recorded observations or real-time feedback-is used to gain experience. This expertise may be used by machine learning algorithms to develop Predictive /forecasting (mathematical-based) and make choices/ decisions. Machine learning approaches have attracted a lot of attention in recent years for their ability as prediction models. ANN has been used to assess the membrane fouling for drinking water nanofiltration[54], quality of water bodies considering total nitrogen, total phosphorous, dissolve oxygen and pH [55], water quality of downstream of dam[56], estimation of accumulation of heavy metals in urban roads[57], prediction model for removal of amido black 10B dye[58] and heavy metals like Pb[59], Cr[60], Cu[61], Co and Mn[62] etc. Based on an optimisation algorithm, Support Vector Machine (SVM) is a relatively new statistical machine learning technology. SVM has been applied to predict membrane permeability 63, flood forecasting[64], assessment of heavy metal pollution[65] and forecasting of atmospheric contaminants such as CO, $O_3$, PM 2.5.[66,67,68] A probabilistic machine learning approach called Gaussian Process Regression (GPR) is utilised for regression problems. GPR has been used to predict stream water temperature[69], Sulfate content in lakes[70], outlet dissolved oxygen in micro-irrigation sand media filter[71], total dissolved solids in water[72], ground water[73,74], water quality in papermaking wastewater treatment system[75], bioenergy production form fountain grass[76], along with this atmospheric contaminants emission analysis such as $O_3$[77], PM 2.5, NO, $NO_2$, $SO_2$[78], $CO_2$[79], etc. SVM and GPR have several advantages over ANN. When working with high-dimensional data, where the number of features is much more than the number of samples, SVM still works effectively. SVM is computationally effective because it uses support vectors, a subset of training samples, to specify the decision boundary. SVM's structural risk minimisation principle helps prevent overfitting, which occurs when a model performs well on the training data but poorly on unseen data. SVM finds the optimal hyperplane that maximises the margin between classes, promoting generalisation and reducing the risk of overfitting. SVM allows the use of various kernel functions (Gaussian, linear, Quadratic, Cubic) to transform the input space, enabling the modelling of non-linear relationships. GPR offers a probabilistic framework that enables the estimation of prediction uncertainty. Decision-making and risk assessment are aided by information about uncertainty. Even when working with little or sparse datasets, GPR is effective. It enables a reliable estimate and prediction by including a prior distribution across functions and updating it considering the observed data. This is especially useful when gathering significant data is difficult or expensive. Compared to ANN, GPR offers a structure that is easier to understand. The predictions made by the model are based on probabilistic distributions that can be seen and examined. GPR provides an understanding of the underlying patterns, connections, and uncertainties related to the forecast.[76] The current study investigates the untapped potential of *Cyperus rotundus* root powder, a natural substance historically valued for its therapeutic benefits, in the fields of water treatment and environmental remediation. Although many bioadsorbents have been studied in the past for the removal of heavy metal ions, using these organic compounds specifically to remove aluminium ions is a new development in the area. The innovation is in utilising Cyperus rotundus root powder's intrinsic qualities, such as its active functional groups, surface area, and eco-friendliness, to efficiently remove aluminium ions from aqueous solutions. Moreover, this research introduces the utilisation of machine learning technologies to predict the extent of aluminium ion removal, employing techniques such as Artificial Neural Networks (ANN), Support Vector Machines (SVM), and Gaussian process regression (GPR). While machine learning methods have gained widespread application across diverse domains, their application within the realm of bioadsorption research for water treatment stands out as a notable innovation. This study presents an avenue to enhance the efficiency of bioadsorption processes through harnessing the capabilities of computational tools. By offering a fresh and innovative strategy to tackle the challenges associated with aluminium ion contamination in water sources, this work effectively bridges a critical gap in research. The study's principal objectives encompass the comprehensive characterisation of the adsorbent material, optimisation of bioadsorption process parameters, and the systematic assessment of kinetic, isothermal, and thermodynamic parameters through batch experiments.

These empirical investigations provide valuable insights into the performance and efficacy of *Cyperus rotundus* root powder as a potent adsorbent for aluminium ion removal. By harnessing the natural bioadsorptive properties of *Cyperus rotundus* root powder, this work addresses critical challenges in water purification, offering an eco-friendly, cost-effective, and efficient alternative to conventional treatment methods. While underscoring the significance of experimental endeavours, the study places a strong emphasis on the integration of machine learning methodologies to predict aluminium ion removal percentages. Researchers can employ ANN, SVM, and GPR to forecast the efficiency of aluminium ion elimination based on input parameters pertinent to the bioadsorption process. Through this amalgamation of computational techniques, researchers gain a deeper comprehension of the underlying bioadsorption mechanism and can modify process parameters to achieve optimal aluminium ion removal outcomes. The present study contributes directly to Sustainable Development Goal 6: Clean Water and Sanitation by providing an innovative and sustainable approach for the removal of aluminium ions from contaminated water sources. The findings of this study aid in the creation of effective and long-lasting techniques for removing aluminium ions from aqueous solutions. This study offers a thorough and creative solution to the problems related to aluminium ion pollution in water sources by using *Cyperus rotundus* root powder and utilising the power of machine learning. The results bring up new opportunities for utilising computational methods and natural materials in the field of environmental engineering, particularly when it comes to the removal of heavy metals using bioadsorbents.

## EXPERIMENITAL

### Adsorbate Preparation

Stock Aluminium solution was prepared by dissolving 8.791 gm of aluminium potassium sulphate (potassium alum), Al $K(SO_4)_2$. 12 $H_2O$ in distilled water (or di-ionised water) and then dilute to 1000 ml. 1 ml = 500 mg/L.[80] By adding distilled water to the stock Al solution, the requisite initial concentrations of Aluminium ion solution have been obtained.

### Bioadsorbent Preparation

*Cyperus rotundus* roots (CRR), a bioadsorbent, were cleaned and rinsed with distilled water, dried at room temperature, and then oven-dried at 60-80$^0$C. Dried CRR was powdered, sieved (180 μm), and stored and labelled (as CRR Powder) for future experiments.[81]

### Adsorbent Characterization

The CRR biomass's physicochemical properties were analysed using various techniques. To identify functional groups interacting with aluminium ions, Fourier transform Infra-red spectroscopy (FTIR) bands of untreated and metal-loaded biomass were recorded using a Shimadzu IR Affinity with ATR, scanning 400–4000 /cm with a 4 /cm resolution. The Brunauer-Emmett-Teller (BET) surface area analyser (Quantachrome Novae 2200) assessed specific surface area, pore diameter/width and pore volume through $N_2$ adsorption/desorption isotherms at 77.35 K. The surface area was obtained by degassing CRR biomass at 120 $^0$C for 6 h to eliminate any adsorbed contaminants before the $N_2$ adsorption experiment. The zeta potential analyser (Anton Paar litesizer 500) measured particle size and isoelectric point of the CRR biomass. The Scanning Electron Microscope with Energy Dispersive Spectroscopy (SEM-EDS) (ZEISS-Sigma IV) examined the biomass morphology.

### Optimisation of the Factors Affecting the Removal of Aluminium

The bioadsorption of aluminium ions from hydrous solutions was investigated in a batch system. A fixed amount of bioadsorbent in a series of 100 ml iron solutions was used in the batch adsorption experiments. By adding 0.1 N NaOH and 0.1 N HCl solutions, the pH (Labindia PICO + model used to test pH) was adjusted between the range of 2-8, agitated in an orbital shaker for 120 min at 250 °C and at 150 rpm. After removing the biomass from the solution, the UV-VIS spectrophotometer (Shimadzu UV1900) was used to measure the strength of aluminium ions in the solution.[80] Subsequently, the experiment aimed at exploring the effects of contact duration, bioadsorbent quantity, initial metal concentration, and temperature on the removal of aluminium was conducted similarly to the earlier described, except that it employed different contact time intervals (15 to 180 min), bioadsorbent amounts (1 to 6 g/l), initial iron

concentrations (0.005 g/l to 0.05 g/l), and temperatures (25°C to 50°C), correspondingly. For each run, the experiment was carried out thrice, and the average values were considered when analysing the data. The relationship between the adsorption percentage or removal effectiveness of the aluminium metal ion by the powdered roots of *Cyperus rotundus* is given by:

$$\text{Removal efficiency} = e\,(\%) = [(Co\text{-}Ce) / Co] \times 100 \quad (1)$$

Where, Co = Initial concentration of metal (aluminium) ions (mg/l)
Ce = Residual concentration of metal (aluminium) ions (mg/l)

The concentration of metal retained in the sorbent part (i.e. adsorption capability (qe) measured in mg/g) was worked out by applying the formula:

$$qe = (Co - Ce) * V/m \quad (2)$$

Where Co and Ce = initial and final (equilibrium) metal ion concentrations in solution (mg/l),
V = Volume of sample (l)
m = Adsorbent mass (g).

**Kinetics and Isotherm Models**

Kinetic tests were carried out to determine the time of contact essential to reach equilibrium. Predetermined parameters were used for the batch experiments, and samples were taken at predesigned intervals to measure aluminium metal concentrations. In this study, the degree of aluminium adsorption on CRR powder was examined using four kinetic models: pseudo-first order model, pseudo-second order model, Elovich model and power function equation. At varying initial aluminium ion concentrations (between 0.01 and 0.05 g/l), isotherm experiments were also carried out using a batch approach with predefined initial parameters. The concentration of residual aluminium ions was determined as previously stated. There are several models for describing bioadsorption equilibrium that are discussed in the available literature. In this study, Langmuir, Freundlich, Temkin (two parameter), Redlich-Peterson, Sips, Toth (three parameter), Fritz–Schlunder, Baudu (four parameter) and Fritz–Schlunder (five parameter) were used.

**Non-linear Regression and Error Function**

The method of Non-linear regression was employed to assess all model parameters in Origin Pro 2022 software (Version 9.9, Origin Lab Corporation, MA). In this study, the correlation coefficient ($R^2$) and the Sum of squares of errors (ERRSQ), the root mean square error (RMSE), the sum of absolute errors (EABS) and the Relative standard deviation (RSD) were studied for an individual range of experimental observations (Using MS Excel). The minimal difference between model data and experimental data is considered when determining the effective model's fit. The lesser the error function values and the higher the $R^2$ can be considered as best the fitted model.[82] The mathematical expressions for error functions are summarised in Table-1 given in supplementary material.

**Thermodynamic Parameters**

The thermodynamic factors like ΔG(Gibbs' free energy), ΔH(Change in enthalpy) and ΔS(Change in entropy) for bioadsorption of the aluminium ions by CRR powder were determined considering the Standard equations given in the supplementary material.

**Machine Learning Modelling Assumptions and Fundamentals**

**Artificial Neural Network**

One of the most well-liked artificial intelligence techniques is an ANN, commonly referred to as a multi-layer perceptron. The conceptual underpinning is provided by the mathematical mapping of the nervous system.[87] The ANN structure is composed of three distinct layers, the first of which is referred to as the input layer and the third as the output layer. Between them is the hidden layer, which might include one or more. The layers are made up of several neurons stacked in layers as linked nodes. Weights between the nodes may be altered to enhance the execution of the model with the use of the proper cost function.[88]

Rectified linear units (ReLU) and other activation functions, such as the sigmoid, are used in nonlinear computations. The following equation is used for generating the prediction ANN output.

$$y = f(W + B) \quad (3)$$

y represents the output, W is the weight, B stands for bias and f is the activation function.

**Support Vector Machine**

Initially designed for classification, SVM has been enhanced for regression with minimal data by mapping input vectors to a larger feature space, allowing better class separation through a larger margin using various kernel functions.[89,90] The SVM uses the objective function presented in Eqn.-4:

$$min\frac{1}{2}\|w\|^2 + c\sum_{i=1}^{n}(\xi i + \xi i*) \quad (4)$$

Given,

$(w\ \Phi(xi) + b) - yi <= \varepsilon + \xi i$

$yi - (w\ \Phi(xi) + b) <= \varepsilon + \xi i*$

Where $w$ Represents the direction vector, and C denotes the adjustment factor, which is a trade-off between training error and the flatness of the model. $\xi i\ and\ \xi i*$ They are known as slack variables, and Φ(xi) accounts for the higher-dimensional hyperspace for the input vector xi. To achieve its explained variance, the SVM model uses the idea of minimising the sum of errors. The kernel functions of SVMs are used to help move data from a lower-dimensional space into a higher-dimensional domain. The most often used kernel functions include linear, polynomial, Gaussian, radial basis function (RBF), and non-linear functions. Support vector machines (SVMs) offer various benefits over ANN, including the ability to describe nonlinear associations between inputs and outputs and the ability to exclusively employ support vectors as a solution method. For the set of tiny datasets, the generalizability of this method is adequate. SVM has been successfully applied to machine learning with large, high-dimensional datasets; hence, it is a promising approach.[91]

**Gaussian Process Regression**

GPRs play a vital role in modern Bayesian methods for discriminative machine learning (e.g., regression, classification, dimensionality reduction). GPs incorporate prior controls, estimating the likelihood of the latent function's marginal likelihood and potential unseen latent functions.[92] Posterior probabilistic estimates are obtained by conditioning on training data, resulting in multidimensional GPs. Mean function (μ(x)) represents the average forecast, typically centred around zero, but it can be a non-zero constant or function based on prior knowledge. The covariance function (kernel) defines smoothness, variability, and correlation structure between input points (K(x, x')). GPR uses a zero-mean Gaussian prior distribution for underlying functions ($f \sim GP(x), k(x, x')$). The revised belief about the underlying function after seeing training data is represented by the posterior distribution ($p(f \mid X, y)$). GPR prediction aims to estimate the target variable value (y*) using the predictive distribution ($p(y* \mid x*, X, y) = N(y* \mid \mu*, \sigma*)$), where μ* is the mean, and σ* is the standard deviation. Kernel function choice depends on input features and desired underlying function behaviour.

**Data Set Description and Processing of Machine Learning Models**

The data set for machine learning modelling was derived from the experimental results for the removal of aluminium ions using CRR powder. For the machine learning technique (i.e., for ANN, SVM, and GPR models), the following factors were taken into consideration as input variables: pH, dose of biosorbent, time of contact, temperature, initial metal ion concentration, final concentration of aluminium ions in solution, and adsorption capacity. These are arranged in a matrix with seven inputs and one output parameter, totalling 76 points and eight parameters. The dataset was partitioned randomly as 85% for training (66 pt) and 15% for testing (10 pt) for ANN, SVM and GPR models. Neural Network Toolbox (for all ANN calculations) and Regression Learner Toolbox (for all SVM and GPR calculations) from MATLAB Version 9.13 (R2022b) was utilized.

## RESULTS AND DISCUSSION

### Adsorbent Characterization

BET analysis was exploited to compute the surface area and pore size, while, BJH (Barrett-Joiner-Halenda) method is used to obtain the pore volume. $N_2$ adsorption isotherms were measured at 77.35 K adsorption temperature (Fig.-1 of supplementary material). The textural properties of the CRR biomass are listed in Table-2 (provided in supplementary material). CRR biomass's BET surface area and pore volume results demonstrate that the bioadsorbent contains the massive pores needed for adsorption. It has been seen that the surface area for the CRR bioadsorbent in their raw form was higher than compared in the previous report by.[50] For CRR biomass, the mesoporous (w.r.t. IUPAC classification) nature of the material (pore size 3.38 nm) results in better particle dispersion.[93,94] In addition, the pore volume was observed to be 0.027 cc/g. SEM-EDS examined the CRR powder's surface, revealing uneven, mesoporous structures (Fig.-2 provided in supplementary material), consistent with the BET results. This heterogeneous, rough surface supports metal ion bioadsorption, potentially including aluminium ions in varied material areas.[96] EDS analysis (Fig.-S2, A and B) confirmed substantial morphological changes in the biosorbent due to aluminium ion binding, distinctly visible before and after the process. Similar research was conducted by [50] utilising *Cyperus rotundus* powder to extract copper ions and analyse the morphological changes before and after adsorption of copper ions. It is concluded from SEM analysis that this material has a sufficient morphological profile to retain metal ions. EDS findings for CRR powder indicated elemental composition: C (52.13%), O (46.32%), Si (1.04%), Ca (0.47%), and Mg (0.04%) (Fig.-S2, C). Post-bioadsorption (Fig.- S2, D), a new Aluminium peak emerged, confirming its presence on the CRR surface. SEM and EDS results establish the CRR powder's ability to adsorb aluminium ions. The surface charge of the bioadsorbent (CRR powder) was indicated by zeta potential. The surface of CRR biomass was found to be negatively charged. The amplitude of the zeta potential, which causes an electrostatic attraction between the adsorbent and the dispersion medium, determines the stability of bioadsorbents in dispersed media.[95] FTIR analysis (Fig.-3 of supplementary material) characterised CRR biomass pre and post bioadsorption, revealing relevant functional groups (C-O, O-H, C=O, C≡O, C-H) implicated in aluminium ion bioadsorption onto CRR powder (Table-3 provided in supplementary material). The disappearance of the peak at 1242.21 /cm and 1381.09/cm indicates that these may be the potential sites for aluminium ions bioadsorption.[97] No notable alterations in peaks were observed when aluminium ions were introduced to the CRR powder biomass. The chemical composition of the CRR powder biomass remained largely unchanged following aluminium adsorption.[52]

### Influence of pH

The pH of the solution is one of the most important factors because it has such a significant impact on the bioadsorption process. It is important to note that pH can affect the hydration of performing factors (such as carboxyl and amino groups) in biomass, as well as metal chemistry, or its solubility. The pH range of 2–8 was used in the study to determine how pH affected aluminium removal. (Experimental conditions: Bioadsorbent dose = 5 g/l, Initial concentration = 0.01 g/l, contact duration = 120 min, Temperature = $25^0$C, Agitation speed =150 rpm). Adsorption capacities adept their maximum at pH levels of 6. When the pH rises from 2 (28.74%) to 6 (86.98%), the percentage of aluminium ions removed increases (as shown in Fig.-1A). Further rise in pH caused no change in the percentage elimination of aluminium. Analogous results were observed earlier by[29], while examining the adsorption of aluminium ions using the stem powder *of Burea monosperma* and *Angle marmelos*. Proton concentrations surged as the pH decreased, and H+ and metal ions started acting as a counteraction to bind the implicated effective sites on the surface of the biosorbent. Protonated active sites were unable to bind the metal. Free ions continue to exist in the solution as a result. Owing to the medium's increased concentration of hydroxyl anions, aluminium ions precipitated after the original pH of the solution was raised above 6. Therefore, pH 6 was used for all further experiments.

### Influence of Bioadsorbent Dose

Measurement of the consequence of bioadsorbent dose (CRR) on the percentage elimination and adsorption capacity of aluminium (Fig.-1 B) was carried out using 10 mg/l of aluminium concentration of

hydrous solution (Experimental conditions: pH = 6, Contact time = 2 hr, Temperature = 250 °C, Agitation speed =150 rpm). The amount of adsorbent used determines the number of biosorption sites available. Up to a CRR biomass concentration of 3 gm, the bioadsorption of aluminium metal ions increased linearly with increasing biosorbent concentration (with an increase in the biosorbent quantity, an increase in the area of the biomass surface, and the number of potential binding sites). Effectiveness of removal after this CRR dose remains constant (Maximum removal % achieved was 94.95). [2] recorded the like results earlier for Al (III) removal using *Ficus Racemosa* bark biomass (AC). The bioadsorption ability (mg/g) of the CRR decreases from 6.94 to 1.5 as the bioadsorbent dose increases from 1 to 6 gm. While increasing the dosage of the bioadsorbent hurts bioadsorption capacity, it has been recorded that higher adsorbent doses result in a greater proportion of extraction. However, several factors, including intervention between binding sites, electrostatic interactions, and a decreased solvent ratio, cause the diffusion to decrease when the adsorbent dose is increased.[98]

**Influence of Contact Time**

In most cases, bioadsorption exhibits a quick initial metal binding that transitions into a more gradual adsorption stage. The maximum removal of 97.99 % was observed within the 90 minutes of contact time (Fig.-1 C) (Experimental conditions: pH = 6, Initial concentration = 0.01g/l, Dose of bioadsorbent = 3 g/l, Temperature = 250, speed of agitation =150 rpm). In the case of CRR powder, the aluminium removal steadily increased from 50.25 % to 97.99 % as the duration of contact increased from 15 to 90 min. Similarly, bioadsorption potential (in mg/g) reached 3.26 in 90 min. However, despite the increased contact time from 90 to 180 minutes, there was no discernible increase in removal was observed. No additional improvement in bioadsorption capacity occurred after the optimum contact period. Early aluminium extraction was high owing to readily available active sites, but prolonged contact led to diminished effectiveness as these sites became saturated. These findings corroborate those of other researchers, who state that the high availability of empty surface sites at the start of the contact period promotes rapid bioadsorption.[99] As these sites are gradually filled, the adsorption rate diminishes, and the remaining sites are less accessible due to repulsive solute–solute interactions across the solid–liquid interface.[100]

**Influence of Initial Concentration**

Figure-1D depicts that, at constant operational parameters (pH 6, 3 g/l dose, 90 min contact time, 25 °C, 150 rpm), the adsorption capacity of CRR enhances with increasing initial aluminium concentration, while the percentage removal declines. Capacity rose from 3.26 to 13.70 mg/g, accompanied by a reduction in removal from 97.92% to 79.90% as concentration increased from 0.01 to 0.05 g/L. This pattern is attributed to the stronger concentration gradient at higher initial solute levels, consistent with previous studies by.[3]

**Kinetics of Adsorption**

To explore the kinetics of aluminium ion bioadsorption, experimental data can be fitted using various kinetic models (Fig.-2). Model parameters were determined using diverse reaction order kinetics, employing OriginPro 2022 Software (Version 9.9, Origin Lab Corporation, MA) for non-linear regression fitting. Experimental outcomes regarding aluminium ion bioadsorption kinetics onto CRR powder were associated with kinetic models, facilitating parameter determination as outlined in Table-4 of supplementary material. Correlation coefficients together with ERRSQ, EABS, RMSE, and ADD metrics validated the extent of model alignment with the experimental findings. Both the power function and the Elovich equations failed to accurately capture the aluminium adsorption characteristics on CRR powder.

Even so, correlation coefficients greater than 0.990 indicated a high degree of model fit. However, correlation coefficients for aluminium ions are inferior in the cases of the pseudo-second-order (PS2), power function equation, and Elovich model. In contrast, the pseudo-first-order (PS1) model closely fits the bioadsorbent (with an $R^2$ value of 0.9975 and low error function values).

The experimental results align more convincingly with the pseudo-first-order model statistics, which emerges as the most effective in elucidating the bioadsorption kinetics of aluminium ions onto CRR Powder.

**Isotherm Modelling**

Bioadsorption isotherms illustrate the connection between the quantity of the bioadsorbed substances per quantity of bioadsorbent and the concentration of the bioadsorbed constituent in the solution. In order to create new adsorption systems or structures, important facts and statistics can be assembled by estimating equilibrium parameters. The experimental results on aluminium metal ion adsorption on biomass surfaces were preferred to the established adsorption isotherm models. The bioadsorption isotherm was defined using the Langmuir, Freundlich, Temkin (two-parameter), Redlich-Peterson, Sips, Toth (three-parameter), Fritz–Schlunder, Baudu (four-parameter) and Fritz–Schlunder (five-parameter) models because the process was complicated and involved numerous physical and chemical interactions (Fig.-3A, B, C). Table-5 presented in supplementary material compares the experimental results to the models that describe the bioadsorption equilibrium of aluminium ions on CRR powder.

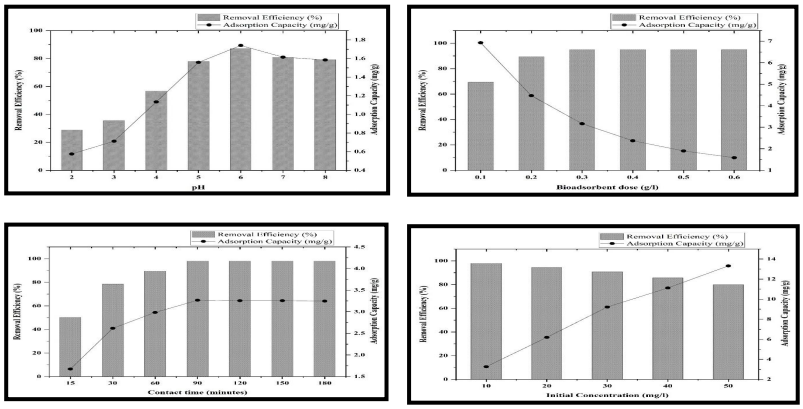


Fig.-1: Effect of Variation of Parameter on the Adsorption of Aluminium by CRR Powder
A: pH B: Contact Time C: Adsorbent Dose D: Initial Metal Concentration

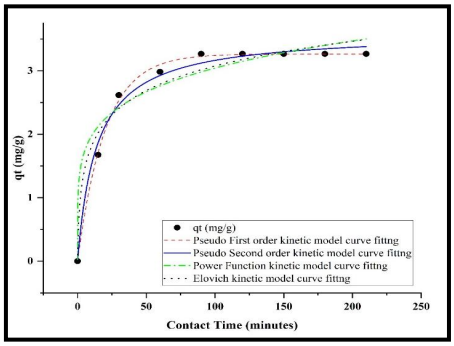


Fig.-2: Bioadsorption Kinetics of Aluminium Ions

The equilibrium constants and adsorption parameters were evaluated through nonlinear regression analysis performed in Origin Pro 2022 (v9.9, Origin Lab Corporation, MA). The error functions, aligned with the slight deviations from foreseen equilibrium data, reveal that the Freundlich isotherm, followed by the Langmuir-Temkin isotherm (among the two-parameter models), offers a close-fit representation. Notably, the Freundlich model excellently captures the experimental observations for aluminium ion bioadsorption onto CRR powder, boasting the lowest error values and a relatively high $R^2$. In the Freundlich equilibrium, the CRR powder's k constants were determined to be 62.08 L/g. The obtained n value (falling between 0 and 10) signifies a relatively strong adsorption of aluminium ions onto the external surface of the CRR adsorbent. The Langmuir isotherm model yielded a monolayer adsorption capacity, q max (in mg/g), of 148.03 for aluminium ions. Conversely, the Temkin isotherm model exhibited low correlation coefficients and higher error values, suggesting its inadequacy in describing this equilibrium. Three-parameter isotherm equations, especially the Redlich-Peterson, Sips, and Toth isotherms, may adequately capture the equilibrium adsorption statics data. These models exhibit strong

coefficients of determination ($R^2 > 0.990$) across all three instances, as indicated in Table S5. Among these models, the Sips isotherm stands out as it provides an excellent and inclusive illustration of the experimental data for bioadsorption isotherms, taking into account the values of the error functions. As derived from non-linear regression, the Toth model parameter (t) ranges from 0 to 1. This suggests a noteworthy similarity between the Toth isotherm and the Langmuir isotherm, implying adsorption on a multifarious surface when t is less than 1. The three-parameter equations yielded the following results for the fit to the bioadsorption isotherm data, in order of accuracy: Toth = Redlich-Peterson < Sips. These three-parameter isotherm models offer a more accurate description of the equilibrium statistics or data compared to their two-parameter models. Furthermore, an exploration was conducted into the adsorption equilibrium statistics utilising four-parameter isotherm models through non-linear regression. The Fritz-Schlunder and Baudu isotherm models, both employing four parameters, were employed to accurately capture the experimental outcomes of bioadsorption isotherms (as depicted in Fig.-3B). Notably, the Fritz-Schlunder and Baudu isotherms exhibit comparable coefficients of determination ($R^2 > 0.990$) and nearly equivalent error function values. It is worth mentioning that the Baudouin isotherm demonstrates a maximal bioadsorption capacity lower than that of both the Langmuir isotherm and its theoretical value. For a more in-depth analysis of the adsorption data, the non-linear variant of the five-parameter Fritz-Schlunder isotherm model was employed. This five-parameter model of Fritz-Schlunder effectively aligns with the experimental observations of bioadsorption isotherms (as illustrated in Fig.-3C). Importantly, the Fritz-Schlunder five-parameter models exhibit superior fitting compared to other isotherm models. This is evident from the remarkably high correlation coefficients (> 0.99) and the notably low error function values associated with this model.

**Thermodynamics Parameters**

Table-6 provided in supplementary material displays the computed thermodynamic parameters, namely ΔG (Gibbs' free energy), ΔH (enthalpy change), and ΔS (entropy change), for the bioadsorption process involving aluminium ions and CRR powder. Additionally, in Fig.-4 (supplementary material), the relationship between 1/T and ln K for CRR powder is graphically represented. The possibility of the reaction is confirmed by the occurrence of negative values in the Gibbs free energy change (ΔG) for the bioadsorption of aluminium ions by CRR powder at various temperatures. This emphasises how the bioadsorption processes are spontaneous, necessitating no external energy input into the system. The adsorption process coincides with an exothermic form of contact, as observed by the negative values of ΔH (enthalpy difference). Moreover, the negative value of ΔS (entropy change) indicates less uncertainty during the bioadsorption of aluminium ions onto the designated adsorbent at the solid-liquid interface. The negative ΔH and ΔS values show that the bioadsorption of aluminium ions onto CRR powder is solely driven by physical interactions and does not include any associated structural alterations.

Table-7 presented in supplementary material provides a comparative assessment of the bioadsorption of aluminium ions by CRR powder in relation to other established biosorbents. The results highlight the potential of the CRR adsorbent for effectively separating aluminium ions from aqueous media. The bioadsorption capacities of certain biological materials may vary due to variations in surface qualities associated with the presence of several functional groups.

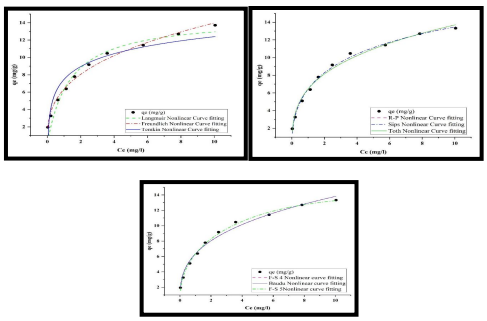

Fig.-3: Bioadsorption Isotherm Model Fitted by A: Two-Parameter Models, B: Three-Parameter Models, C: Four and Five-Parameter Models

**Artificial Neural Network model**

The study proposes a two-layer ANN model for predicting the biosorption of aluminium ions by CRR powder, with a detailed ANN configuration provided in supplementary material Table-8. The dataset was split such that 85% (66 of 76 points) were used for training and 15% (10 points) for testing. By separating the data into training, validation, and test sets, the model's ability to generalise and accurately predict unseen data points was assessed. To identify the most suitable ANN structure, several feed-forward networks with different hidden-layer and neuron combinations were trained, and the architecture yielding the lowest MSE was selected. The optimal model consisted of an FFNN with seven inputs, two hidden layers (10 and 1 neurons), and a single output neuron, denoted as MLP (7:10:1:1). The Levenberg–Marquardt training algorithm was employed to optimise the weights and biases of this network. As depicted in Fig.-4A, the ANN model demonstrated excellent conformity between predicted and experimental values. The model exhibited a strong correlation coefficient of 0.9964, indicating high reliability. Following validation, the prediction accuracy was calculated as 99.65% (Fig.-4B). Evaluation using an independent test dataset yielded an $R^2$ of 0.9933 for aluminium removal performance (Fig.-4C).

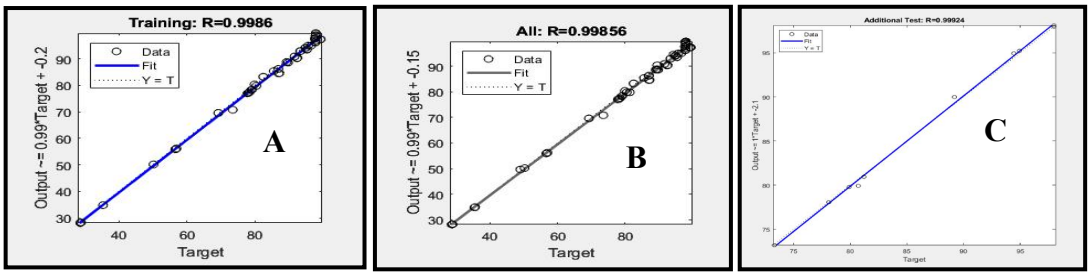


Fig.-4 A: Scatter Plot Indicating Training Efficiency of ANN Model B: Scatter Plot Indicating Overall Efficiency of ANN C: Scatter Plot Indicating Efficiency of Additional Test Data

**Support Vector Machine Model**

An SVM-based predictive model for aluminium ion biosorption onto CRR powder was established in this study. The dataset was partitioned into 85% training (66 points) and 15% testing (10 points). A 10-fold cross-validation procedure was implemented during training. Different kernel functions were assessed, and hyperparameters were optimised using random search, grid search, and Bayesian optimisation. The quadratic kernel produced the most effective model. Figure-5 shows the training and testing results obtained using Bayesian optimisation, and Table-9 (supplementary material) summarises the corresponding model performance and parameters.

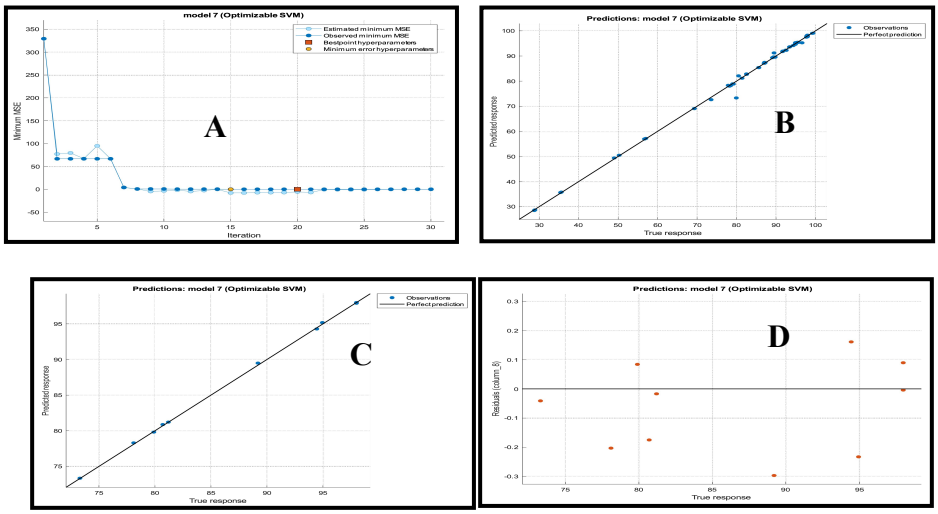


Fig.-5: A: Training SVM using Bayesian Optimiser B: Predicted Vs. Actual Removal Efficiency (%) -SVM Training C: Predicted Vs. Actual Removal Efficiency (%) – Additional testing (SVM) D: Residual Plot for Additional Test Data (SVM)

**Gaussian Process Regression model**

This research aimed to establish a GPR-based model for characterising the biosorption of CRR powder in the removal of Aluminium ions. The model was developed using 85% of the dataset (66 data points) for training, while the remaining 15% (10 data points) were used as a test set, extracted from the original dataset of 76 points. The GPR models were trained with k-folds = 10 cross-validation folds. Multiple kernel functions were examined to identify the most closely operative one for building a robust model. The hyperparameters were fine-tuned during the training process until satisfactory results were achieved. The optimal model, conducive to optimisation, was determined to perform well with the nonisotropic rational quadratic kernel function. The outcomes of training and testing the GPRs using Bayesian optimisers are illustrated in Fig.-6. The performance and parameters of the GPR Bayesian model are outlined in Table- 10 of supplementary material.

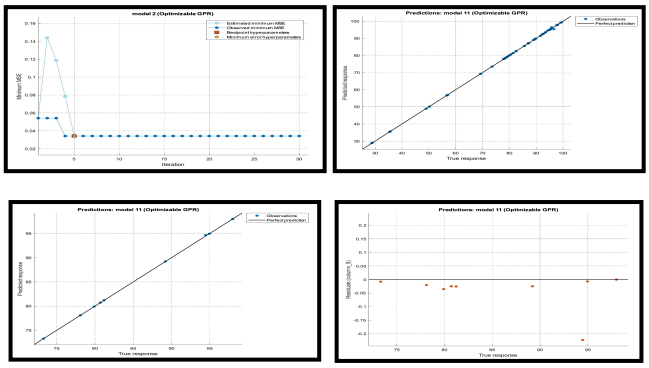

Fig.-6: A: Training GPR using Bayesian Optimiser B: Predicted Vs. Actual Removal Efficiency (%) – GPR Training C: Predicted Vs. Actual Removal Efficiency (%) – Additional Testing (GPR) D: Residual Plot for Additional Test Data (GPR)

**Comparison of Performance of Trained Models**

The predictive performance of the ANN, SVM, and GPR models was assessed using $R^2$ and MSE. For independent validation, ten new experimental trials—unseen during training—were conducted, and the corresponding actual and predicted responses, along with residuals, are reported in Table-1. The GPR Bayesian optimiser generated residuals that were both smaller in magnitude and less variable than those of the ANN and SVM models. Additional statistical comparison using $R^2$ and MSE (Table-11, supplementary material) shows that all three models reached $R^2 \approx 0.99$ for training and testing sets. The GPR Bayesian optimiser, however, showed noticeably lower MSE, confirming its dominance over the ANN and SVM Bayesian optimiser in terms of predictive accuracy and generalisation ability.

Table-1: Statistical Comparison of ANN and SVM Models

| Error Index | Models | | | | | |
|---|---|---|---|---|---|---|
| | ANN | | SVM | | GPR | |
| | Training | Testing | Training | Testing | Training | Testing |
| $R^2$ | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 | 0.99 |
| MSE | 1.538 | 1.609 | 0.917 | 0.160 | 0.184 | 0.07 |

## CONCLUSION

This study delves into the efficacy of Cyperus rotundus root powder as a viable solution for the removal of aluminium ions from aqueous environments. Cyperus rotundus root powder proves to be an environmentally sustainable and economically feasible bioadsorbent for extracting aluminium ions from aqueous streams or media. Optimal conditions, specifically a pH of 6, biomass concentration of 3 gm, contact time of 90 minutes, and a temperature of 25°C at 150 rpm, yielded a remarkable removal efficiency of up to 97.99%. The maximum bioadsorption capacity was determined to be 148.03 mg/g based on the Langmuir model. Nonlinear regression techniques were employed to identify the most suitable isotherm and its associated parameters, both for isotherm and kinetic models. Selection criteria included the coefficient of determination and minimal error function values, ensuring a strong fit for both

models. Kinetic experiments indicated rapid attainment of equilibrium in a short contact time, with the pseudo-first order (PS1) model effectively characterising the kinetics of aluminium ion bioadsorption onto CRR Powder. Through a detailed evaluation of various non-linear isotherm models, the Fritz-Schlünder isotherm emerged as the most closely fitting model for the bioadsorption of aluminium ions onto powdered Cyperus rotundus root. Among the tested models for describing the adsorption equilibrium isotherms of aluminium ions on powdered Cyperus rotundus root powder, the ranking was as follows: Fritz–Schlunder (five-parameters) > Sips > Freundlich > Redlich–Peterson = Toth > Baudu = Fritz–Schlunder (four-parameters) > Langmuir > Temkin. Furthermore, analysis of thermodynamic parameters unveiled the potential, spontaneity, and exothermic nature of the bioadsorption process for aluminium ions. The study employed various machine learning techniques, including ANN (Feed Forward Neural Network), SVM (Bayesian optimiser), and GPR (Bayesian optimiser), to predict the percentage extraction of aluminium ions from aqueous solutions. The predicted biosorption removals from both the ANN, SVM, and GPR models aligned well with the experimental results. To compare the prediction performance of these models, the validation data set was used, and MSE and $R^2$ were utilised as evaluation metrics. The results indicated that the GPR model demonstrated superior prediction precision compared to both the ANN and GPR models. This finding highlights the efficiency and promise of the proposed GPR model for predicting bioadsorption percentage removal. By doing so, this model has the potential to save a substantial amount of time and money. Moreover, the integration of machine learning into this study represents a significant advancement in the field of bioadsorption, enhancing the bioadsorption process and paving the way for future research on the application of computational methods in environmental engineering. This, in turn, may lead to the development of more effective water and wastewater treatment systems. Through the development of a green bioadsorption-based treatment approach, the study provides meaningful progress toward achieving Sustainable Development Goal 6, ensuring cleaner and safer water for communities. Subsequent investigations should prioritise the augmentation or modification of biomass to amplify its efficacy in the removal of aluminium and other heavy metals. The application of CRR for bioadsorptive efficacy within intricate setups, like actual water bodies or wastewater effluents comprising diverse co-ions, demands exploration. Furthermore, possibilities for research encompass column studies, pilot-scale implementation for commercial purposes, desorption investigations, the potential for adsorbent reutilization, and the secure disposal of laden adsorbents.

## CONFLICT OF INTERESTS

The authors have no competing interests to declare that are relevant to the content of this article.

## AUTHOR CONTRIBUTIONS

The present work is related to *SDG-6: Clean water and Sanitation*. All the authors contributed significantly to this manuscript, participated in reviewing/editing and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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