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Supervise Machine Learning Model to Predict Mercury Adsorption of Inverse Vulcanized Copolymer

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ABSTRACT

This study proposes a novel approach for improving mercury adsorption predictions using Gaussian Process Regression (GPR), a supervised machine learning technique. By leveraging experimental data on mercury adsorption, including key parameters such as initial mercury concentration, adsorption time, and pH of wastewater, a GPR model was developed to predict mercury removal efficiency. The optimization of hyperparameters, such as the choice of kernel functions and sigma values, was carried out to improve the model's predictive accuracy. The model achieved high performance, with R^2 values of 0.90 for training and test datasets. Additionally, a comprehensive hyperparameter optimization process led to an optimized model with R^2 values of 0.98 and a low mean square error, demonstrating the model's potential for practical, scalable applications in wastewater treatment. This study highlights the promising role of machine learning in enhancing environmental remediation technologies, offering a more efficient and cost-effective alternative for mercury removal.

1. Introduction

The global water crisis, exacerbated by both the scarcity of clean water and contamination from toxic substances, is an escalating challenge, with heavy metals being one of the most significant pollutants. Anthropogenic activities such as gold mining, coal combustion, waste incineration, and industrial processes in sectors like textiles and petrochemicals contribute heavily to this issue [1–5]. Among the toxic heavy metals, mercury is considered one of the most dangerous due to its severe health and environmental impacts. Recognized by the World Health Organization (WHO) as a priority

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pollutant, mercury predominantly exists in its inorganic form, Hg^{2+}, in water [6,7] . However, through a methylation process, it can be transformed into methylmercury (CH_3Hg^+), a far more toxic compound. This transformation is driven by methyl donors in the environment, enabling mercury to enter the food chain through bioaccumulation and biomagnification [8,9].

Mercury's ability to accumulate in organisms, travel long distances through the atmosphere, and persist in ecosystems makes it particularly hazardous. Even at low concentrations, methylmercury poses significant health risks, particularly to the nervous system, kidneys, and other vital organs [10,11] . Chronic exposure can lead to irreversible damage, including neurological disorders such as Minamata disease. Consequently, mercury is a regulated substance in many countries, with guidelines in place to limit its presence in drinking water and wastewater [12-14].

To mitigate mercury pollution, various water treatment technologies have been developed, including chemical precipitation, membrane filtration, ion exchange, and adsorption [15,16] . Among these, adsorption is widely regarded as one of the most effective methods due to its simplicity, cost-effectiveness, and high removal efficiency. Activated carbon is one of the most commonly used adsorbents for mercury, owing to its large surface area and porous structure. However, to enhance its mercury removal capacity, activated carbon often requires surface modifications, such as treatment with metal oxides or sulfides, which can increase costs and introduce environmental concerns such as leaching [17].

Recent research has focused on developing alternative adsorbents that are not only effective but also environmentally friendly and affordable. Materials such as metal-organic frameworks (MOFs) [9,18,19], covalent organic frameworks (COFs) [18], layered double hydroxides (LDHs), and even natural and synthetic polymers have been explored for mercury removal. While these materials show promising results, they are often hindered by limitations such as low adsorption capacity, high production costs, and poor reversibility, highlighting the need for more innovative solutions [18]..

In recent years, inverse vulcanized copolymers have gained attention as a novel class of sulfur-rich materials that can be produced using a one-pot, solvent-free method [20,21]. These copolymers, derived from industrial byproducts such as sulfur, exhibit excellent properties for a range of applications, including fertilizers [22-25] , wastewater treatment [26–28], energy storage, and CO_2 capture [29]. The sulfur content in these materials, particularly the high soft Lewis acid affinity of sulfur for Hg^{2+}, makes them promising candidates for mercury adsorption. Several monomers, including limonene, myrcene, dicyclopentadiene (DCPD), and others, have been utilized to create inverse vulcanized copolymers. However, while these materials boast high sulfur content (≥ 40 wt%), their hydrophobicity limits their mercury uptake capacity, which typically remains under 26 mg/g. This hydrophobic nature inhibits their ability to form hydrogen bonds with aqueous Hg^{2+} ions, thus reducing their effectiveness in mercury removal. Some attempts have been made to improve this, such as coating the copolymer with silica gel to enhance kinetics and using catalysed copolymers for higher binding capacities. Recently, efforts to increase adsorption efficiency have involved incorporating hydrophilic monomers like methacrylic acid (MAA) and 2-carboxyethyl acrylate (CEA), resulting in improved mercury uptake. However, these copolymers still contain unreacted sulfur and processing them into usable forms often requires blending with other polymers, such as polyacrylonitrile (PAN), which raises overall costs. Furthermore, no predictive models have been developed to accurately forecast mercury removal from wastewater, a crucial step for scaling up these processes for industrial applications.

Machine learning (ML) offers a powerful alternative to traditional experimental methods by providing a means to model and predict complex systems. In the context of mercury removal, ML can be used to predict adsorption efficiency and optimize treatment parameters in a more cost-effective and efficient manner. By analysing large datasets derived from experimental studies, ML models can

identify key factors influencing mercury removal and help design better-performing adsorbents. Moreover, these models can assist in developing predictive tools for scaling up mercury removal technologies, a critical step toward industrial implementation.

This study proposes the development of a machine learning model aimed at predicting the effectiveness of various mercury adsorbents in wastewater treatment. By leveraging supervised learning techniques and data from previous adsorption experiments, the model will be trained to predict mercury uptake efficiency based on parameters such as adsorbent type, surface area, porosity, and environmental conditions. Additionally, the model will be used to simulate different treatment scenarios, optimizing the conditions for maximum mercury removal while minimizing costs. Ultimately, this machine learning-based approach aims to provide a more robust, scalable, and efficient solution to mercury contamination in water, potentially transforming the landscape of water treatment technology.

2. Methodology

2.1 Machine Learning Model Development

Figure 1 presents the overall flowchart for the machine learning model development. To develop a predictive machine learning (ML) model for estimating mercury adsorption efficiency, a supervised learning approach was implemented, with a particular focus on Gaussian Process Regression (GPR). GPR is a powerful, non-parametric Bayesian technique that offers not only accurate predictions but also uncertainty quantification, which is crucial in modeling environmental and chemical processes that often involve variability, noise, and nonlinear dynamics.

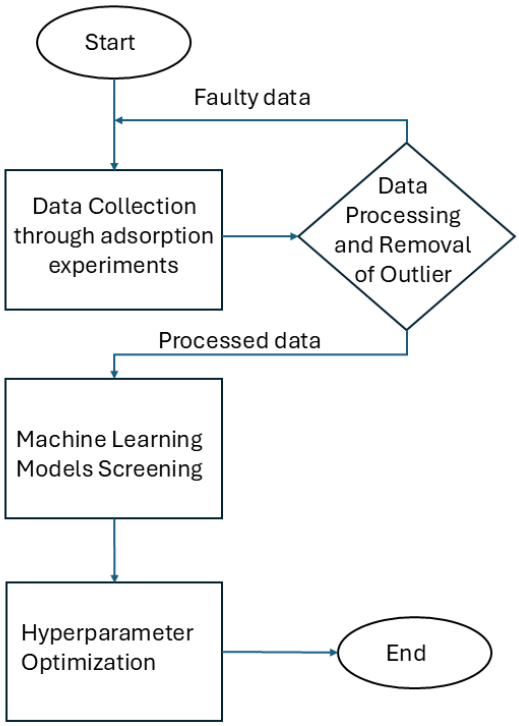


Fig. 1. Overall flowchart of machine model development

Unlike conventional regression algorithms, which fit a single deterministic function to the training data, GPR treats prediction as a distribution over functions. It assumes that the underlying data can be represented as a sample from a multivariate Gaussian distribution, where any finite number of

observed outputs follow a joint Gaussian distribution. The key idea behind GPR is that the output values are not fixed but rather random variables drawn from a Gaussian process, fully defined by a mean function (often assumed to be zero) and a covariance function (kernel). The kernel function encodes assumptions about the smoothness, periodicity, and correlation length of the target function, essentially defining how data points influence each other.

Mathematically, a Gaussian process is defined as Eq. (1):

$$f(x) \sim GP(m(x), k(x, x')) \tag{1}$$

where m(x) is the mean function (often m(x) = 0) and k(x,x′) is the covariance function or kernel, which specifies the similarity between any two-input points x and x'. The predictive distribution at a new point x_* is Gaussian with mean μ(x_*) and variance σ^2(x_*), given by Eq. (2) and (3):

$$\mu(x_*) = K_*^T (K + \sigma_n^2 I)^{-1} y \tag{2}$$

$$\sigma^2(x_*) = k(x_*, x_*) - K_*^T (K + \sigma_n^2 I)^{-1} K_* \tag{3}$$

Here, K is the covariance matrix of the training inputs, K$_*$ is the covariance vector between the new point and training inputs, y is the training target vector, and σ_n^2 is the noise variance. This formulation enables GPR to offer not just a point prediction but also a confidence interval making it extremely useful when modelling complex adsorption phenomena like mercury removal, where experimental uncertainties are common.

In this study, the input features for the GPR model included initial mercury concentration, adsorption time, and solution pH, all of which are known to influence adsorption behavior. These features were selected based on both empirical knowledge and preliminary correlation analysis. The corresponding output was the percentage of mercury removed, which served as the dependent variable.

To ensure that the model accurately captured the underlying physical behavior, hyperparameter tuning was performed. The kernel function, central to GPR performance, was carefully selected and optimized. Different kernels such as squared exponential (RBF), Matern, and rational quadratic were explored. Each kernel brings unique characteristics: for example, the RBF kernel assumes infinite smoothness, while the Matern kernel offers more flexibility and is more robust to noise. Hyperparameters like the length scale and signal variance were adjusted using optimization algorithms to minimize error metrics such as RMSE and MAE.

To combat overfitting a common issue in flexible models like GPR, 5-fold cross-validation was utilized. This method involves partitioning the dataset into five subsets, training the model on four subsets, and validating on the fifth. This process is repeated so each subset serves as validation once, and the average performance is used to estimate model generalization. Furthermore, the full dataset was split into training (90%) and testing (10%) sets. The testing data was withheld during model development and used solely to assess final predictive performance.

All computational tasks, including data preprocessing, model training, kernel selection, and performance evaluation, were conducted in MATLAB R2022b. MATLAB's robust statistical and machine learning toolbox provides a flexible platform for implementing GPR and visualizing uncertainty bands. Its seamless integration of data manipulation, model tuning, and cross-validation procedures made it an effective environment for this project.

3. Results and Discussion

3.1 Gaussian Progression Model Development

Initially, four distinct kernels of Gaussian Process Regression (GPR), a highly flexible and powerful supervised machine learning technique, were investigated to model and predict the mercury adsorption efficiency from wastewater. GPR is a non-parametric, probabilistic model that not only provides point predictions but also offers credible intervals or uncertainty estimates, which is particularly advantageous in modelling complex and nonlinear environmental systems where data variability and measurement errors are common. This feature makes GPR especially valuable in cases like mercury adsorption, where multiple input parameters such as initial mercury concentration, contact time, and pH interact in intricate, nonlinear ways to influence the output variable, namely, the percentage of mercury removed.

The kernels employed in the GPR framework serve as covariance functions that determine how input features relate to one another in the model. Each kernel embodies a different assumption about the underlying function that maps inputs to outputs, thereby influencing how the model interprets patterns in the data. Specifically, the four kernels explored in this study included Exponential, Square Exponential (also known as Radial Basis Function), Matern, and Rational Quadratic kernels. These kernels were selected based on their varied characteristics in capturing different types of trends ranging from smooth and gradual variations to more abrupt changes in data patterns.

To evaluate and compare the performance of each GPR model, three widely used regression evaluation metrics were applied: Root Mean Square Error (RMSE), R-squared (R^2), and Mean Absolute Error (MAE). These metrics provide a comprehensive view of model accuracy, with RMSE and MAE indicating the magnitude of prediction errors, while R^2 explains the proportion of variance in the observed data that is captured by the model. Table 1 presents the results of these performance indicators for both training and testing datasets.

Among the four kernels tested, the GPR models incorporating the Exponential and Square Exponential kernels yielded the most robust predictive performance. Notably, both models achieved a high R^2 value of 0.90 on the test set, suggesting a strong correlation between predicted and actual mercury removal percentages. This high value implies that the models could explain 90% of the variation in the adsorption data, which is indicative of a good model fit. The Square Exponential kernel, in particular, excelled in modelling the smooth and continuous functional relationship between the key input variables (mercury concentration, adsorption time, and pH) and the adsorption performance.

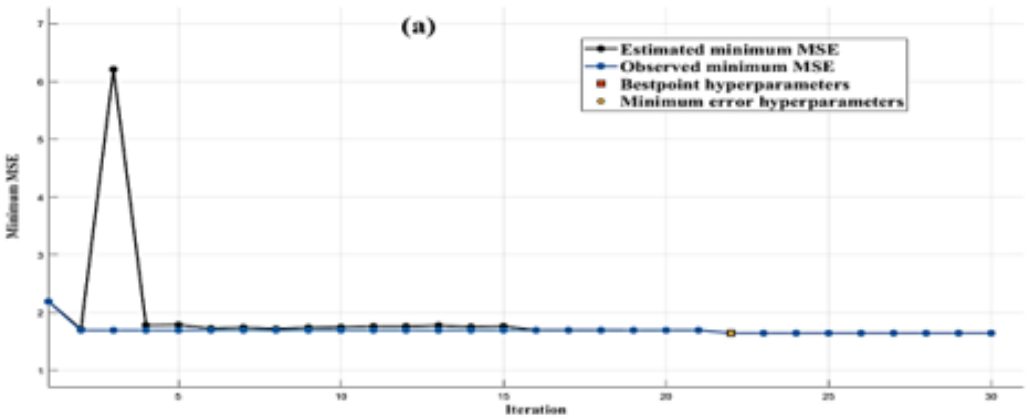
The kernel choice is a fundamental aspect of Gaussian Process Regression, as it essentially governs the shape and behaviour of the functions that the model can learn. The Exponential kernel is well-suited to datasets where sharp transitions or localised variations are expected, making it flexible for capturing short-range dependencies. In contrast, the Square Exponential kernel assumes a more global smoothness in the function and is ideal for modelling gradual and consistent trends across the input space. The superior performance of these two kernels implies that the mercury adsorption process likely involves both local variability and an overall smooth trend, aligning with the chemical and physical nature of the adsorption mechanism. Thus, these kernels are considered appropriate and effective choices for predictive modelling in this environmental context, offering both accuracy and interpretability.

Table 1
Summary of 4 different Kernel of GPR supervised machine learning model

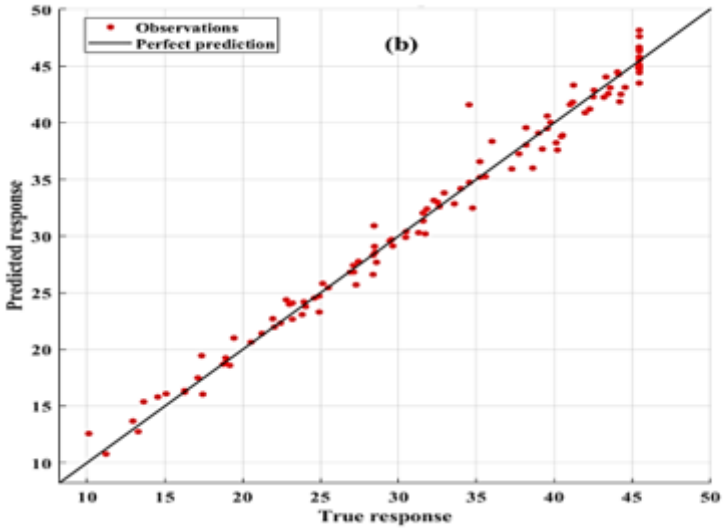
Model Type	RMSE (VAL)	R^2 (VAL)	MAE (VAL)	MAE (Test)	RMSE (Test)	R^2 (Test)
Gaussian Process Regression (Exponential)	2.97	0.91	8.84	1.86	2.47	0.90
Gaussian Process Regression (Squared Exponential)	3.45	0.88	2.46	1.88	2.54	0.90
Gaussian Process Regression (Rotational Quadratic)	2.96	0.91	2.10	2.01	2.63	0.89
Gaussian Process Regression (Matern 5/2)	3.21	0.90	2.29	1.98	2.65	0.89

3.1 Hyperparameter Optimization

To determine the optimal hyperparameters for the Gaussian Process Regression (GPR) model, a comprehensive hyperparameter optimization process was conducted. This process involved systematically exploring specific ranges for each hyperparameter to fine-tune the model for maximum predictive accuracy. The key hyperparameters included sigma, which was varied between 0.0001 and 101.4182, and the basis function, with options such as constant, zero, and linear. The kernel function was explored using a variety of isotropic and non-isotropic options, including Exponential, Matern 3/2, Matern 5/2, Rotational Quadratic, and Squared Exponential. The kernel scale was tested within the range of 0.79 to 790, and the standardization of data was evaluated as either true or false. The goal of the optimization was to minimize the mean square error (MSE), thereby improving the model's prediction accuracy. After conducting multiple iterations, the optimum hyperparameters were identified as follows: a linear basis function, a Nonisotropic Rational Quadratic kernel, a kernel scale of 2.08, sigma = 0.000101, and standardized data = true. These selected values resulted in the most accurate predictions for mercury adsorption, optimizing the GPR model for practical applications in wastewater treatment. Optimized GPR model possesses low mean square error and high R^2 value of 0.98 which shows its robustness to explain the design space as depicted in Figure 2 and Table 2.



(a)



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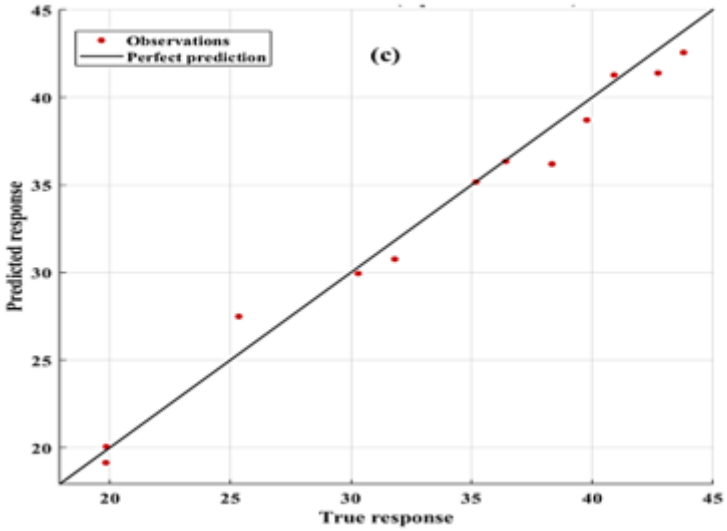

(c)

Fig. 2. (a) Min MSE, predicted vs. actual (b) train and (c) test

Table 2
Error summary of optimized GPR model

Error Index		
RMSE	Train	1.28
	Unseen	1.13
R^2	Train	0.98
	Unseen	0.98
MAE	Train	0.88
	Unseen	0.88

4. Conclusion

The application of Gaussian Process Regression (GPR) has proven to be an effective method for predicting mercury adsorption in wastewater treatment. By incorporating critical parameters such as initial mercury concentration, adsorption time, and pH, the GPR model successfully captured the complex and nonlinear relationships affecting mercury removal efficiency. Initially 4 different kernel

of GPR model were tested, each showed different performance such as the R^2 for exponential, squared exponential, rotational quadratic and Matern 5/2 was found to be 0.90, 0.90, 0.89 and 0.89, which showed that hyperparameter optimization can be done to improve the prediction efficiency of the model. Following an extensive hyperparameter optimization process, the model demonstrated exceptional predictive accuracy, with R^2 values reaching 0.98, indicating its robustness and reliability in real-world scenarios.

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