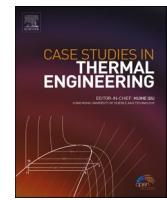


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## Machine learning-based optimization for catalytic sulfur removal: Computational modeling and analysis of fuel purification for reduction of environmental impacts

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### ABSTRACT

Hydrodesulfurization (HDS) process is an important process for separation of sulfur compounds from petroleum-based products due to operational and environmental problems that the sulfur compounds can cause. In this study, this process was evaluated to optimize its performance in removing sulfur compounds from petroleum to reduce its adverse effects. Multiple machine learning models were implemented for optimization of HDS process considering several inputs/outputs. Each data point has four input parameters: pressure, temperature, initial sulfur content of petroleum, and dosage of catalyst in the reactor. Sulfur concentration (ppm), SO<sub>2</sub> emission percentage (%), and HDS cost (\$) are also outputs to be optimized by the machine learning models. Multi-layered perceptron (MLP), Multi-task Lasso (MTL), and Gaussian process regression (GPR) are core models in this study developed for the first time for HDS process. These models were optimized utilizing Artificial Bee Colony (ABC) and applied on cleansed and normalized dataset. According to assessments done on final models, sulfur concentration, emission %, and HDS cost are predicted by R<sup>2</sup>-scores of 0.983, 0.999, and 0.990 respectively using models proposed in this study. Also, absence of overfitting can be guaranteed using these models according to analysis done in results section.

### 1. Introduction

Data-driven models have recently attracted much attention in modeling and optimization of different processes in engineering applications. These models basically rely on the availability of dataset to train a suitable algorithm, and then the model is tested using the data that has not been employed in the training step. Moreover, optimization and classification tasks can be carried out using data-driven models [1–3]. These models are not of physical meaning unlike mechanistic models [4–6]; however, they have superior advantages in process modeling in terms of fitting accuracy [7,8]. The models of data driven have been also used in process control in which online monitoring and model predictive control (MPC) can be implemented to control the process parameters [9–11]. These models can be also used for process optimization in which one or more targets are set, and the process is optimized by tuning the input parameters. An application of the data-driven model is in process optimization for environmental applications in which the emission of harmful gases such as CO<sub>2</sub>, SO<sub>2</sub>, etc. can be minimized.

The method of data driven has been reported in refinery of petroleum to optimize the removal sulfur compounds from hydrocarbons. Process of hydrodesulfurization (HDS) is reported as the major process for sulfur removal and optimized to decrease the

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**Table 1**

The HDS process dataset [12,13].

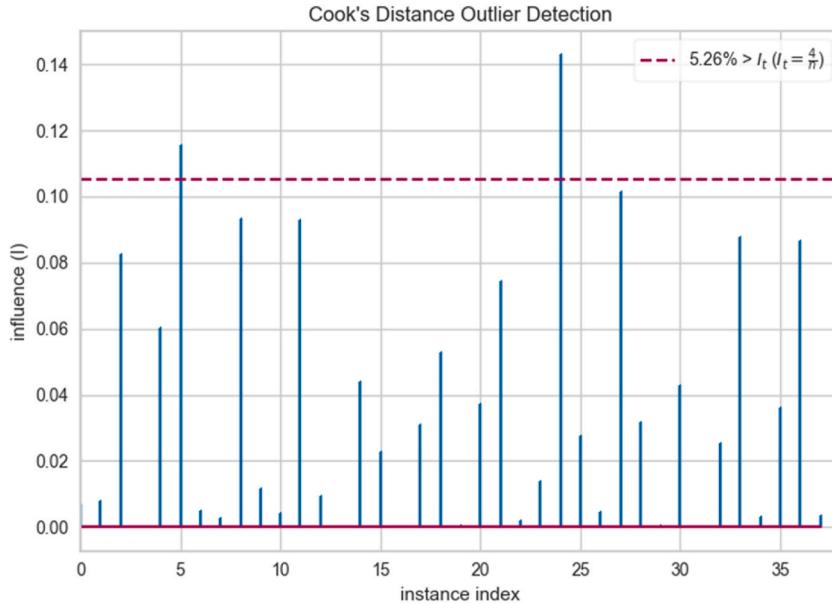
Num	Input variables				Target outputs		
	Temperature (°C)	Pressure (bar)	Dosage (g)	Initial sulfur (ppm)	Sulfur concentration (ppm)	Emission (%)	HDS cost (\$)
1	50	20	0.2	4000	3480	69.6	15
2	50	20	1	500	300	48	12
3	50	20	1	2250	1988	70.7	15.5
4	50	60	0.2	500	330	52.8	18
5	50	60	0.2	4000	3180	63.6	25
6	50	60	1	2250	1805	64.2	25.5
7	50	100	0.2	500	400	64	28
8	50	100	1	500	150	24	32
9	50	100	1	2250	1640	58.3	35.5
10	250	20	0.2	2250	1530	54.4	15.5
11	250	20	0.2	4000	2650	53	19
12	250	20	1	500	110	17.6	16
13	250	20	1	2250	1500	53.3	19.5
14	250	20	1	4000	2600	52	23
15	250	60	0.2	500	150	24	22
16	250	60	0.2	2250	1360	48.4	25.5
17	250	60	0.2	4000	2300	46	29
18	250	60	1	500	75	12	26
19	250	60	1	2250	1315	46.8	29.5
20	250	60	1	4000	2130	42.6	33
21	250	100	0.2	500	245	39.2	32
22	250	100	0.2	2250	1080	38.4	35.5
23	250	100	0.2	4000	1800	36	39
24	250	100	1	500	55	8.8	36
25	250	100	1	2250	850	30.2	39.5
26	450	20	0.2	500	140	22.4	16
27	450	20	0.2	4000	850	17	23
28	450	20	1	500	28	4.5	20
29	450	20	1	2250	290	10.3	23.5
30	450	20	1	4000	650	13	27
31	450	60	0.2	500	7	1.1	26
32	450	60	0.2	2250	289	10.3	29.5
33	450	60	1	500	0	0	30
34	450	60	1	2250	95	3.4	33.5
35	450	60	1	4000	106	2.1	37
36	450	100	0.2	500	5	0.8	36
37	450	100	0.2	2250	180	6.4	39.5
38	450	100	0.2	4000	380	7.6	43
39	450	100	1	500	0	0	40
40	450	100	1	2250	28	1	43.5

process cost and increase the sulfur separation efficiency of process [12,13]. A multi-objective data-driven model based on support vector machine (SVM) was developed for optimization and modeling of hydrodesulfurization process to decrease the level of sulfur compounds in a petroleum feed, and the model was validated through comparing with experimental measurements [12]. It was indicated that the method of SVM, and its combination of Genetic Algorithm (GA) outperformed in comparison with other predictive models in optimization of HDS process.

Indeed, data-driven models and especially machine learning models can be well applied for process modeling and optimization in petroleum engineering area to improve the process efficiency [14]. The method of machine learning needs some experimental data to be developed and the tested model can be used for design space and description of the process [15]. The main disadvantage of the machine learning models in petroleum refinery modeling is that these models do not offer mechanistic understanding of the process and chemical reactions in the desulfurization process. Nonetheless, the non-mechanistic models such as data-driven ones have been successful in statistical analysis of different processes and optimization provided that enough measurements are available to build the models [16–18].

It should not be unexpected that machine learning (ML) has made significant progress in the scientific field in recent years. There has already been evidence that this collection of statistical models can greatly speed up both fundamental and practical research. The development and integration of machine learning (ML) into a wide variety of scientific subjects and domains is gaining momentum at the moment [19–23].

A neural network that consists of many layers of perceptrons is referred to by its acronym MLP. Forward-feeding artificial neural networks are what are known as MLPs. At a minimum, an MLP will have three layers total, comprised of an input, an output, and at least one (generally multiple) hidden layer. Instead of being triggered, input layer nodes just serve as a representation of the data point. For starters, there's the input layer. The input layer will have  $d$  extra nodes if the vector reflecting the data point has  $d$  dimensions [24, 25].



**Fig. 1.** Cook distance analysis of HDS data.

When the likelihood follows a normal distribution, the posterior also has an analytic form, which makes GPR a generalizable and analyzed probabilistic model. This model is a non-parametric model with complete Bayesian features. It provides a posterior for the predicted outcomes [26].

LASSO regression, also known as least absolute shrinkage and selection operator, is an example of a linear model that makes use of regularization and is one of the models utilized in this investigation. Lasso models predict sparse coefficients linearly. This method selects solutions with smaller non-zero coefficients, minimizing the features the supplied solution depends on. Consequently, there are fewer features overall. As a consequence of this, Lasso and the several iterations that exist are crucial in the field of compressed sensing. Under certain conditions, it is able to extract the complete set of coefficients that are greater than zero [27,28].

In this research we developed a new methodology based on machine learning for the first time to study a petroleum unit operation for removal of sulfur compounds from a feed. The new methodology can contribute to the optimization of HDS process for fuel purification in petroleum industry. The methods of computation are based on Multi-layered perceptron (MLP), Multi-task Lasso (MTL), and Gaussian process regression (GPR), which are optimized via the method of Artificial Bee Colony (ABC). In this work, we will introduce the problem statement in section 2, then the ML models will be explained in section 3 along with optimization method, and finally, the results will be validated through comparing with experimental data and evaluated consequently.

## 2. Problem statement

The problem under investigation in this study is a batch process at laboratory scale designed for desulfurization of a petroleum feed via HDS process. The experimental data has been collected from a source, and detailed explanation of the process and experiments can be found in Ref. [12]. Each data point contains 4 input parameters including pressure, temperature, catalyst amount, and initial amount of sulfur [13]. Moreover, three outputs including HDS cost, sulfur concentration, and emission of SO<sub>2</sub> were considered for the modeling of process [13,29]. The dataset is listed in Table 1 indicating the inputs/outputs parameters for modeling the process. A Cook's distance analysis of the dataset is visualized in Fig. 1, which shows only 2 data points are outliers and should be omitted in training phase. Modeling is carried out using three machine learning models to estimate the target parameters as outlined in Table 1.

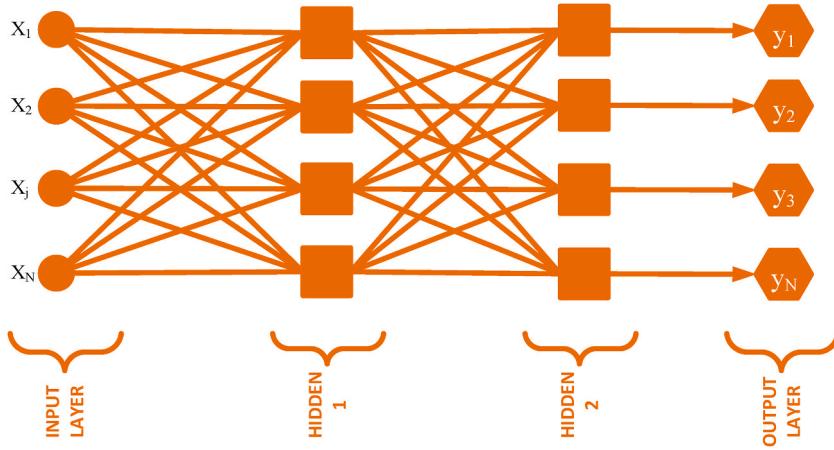
## 3. Machine learning methods

### 3.1. Artificial Bee Colony (ABC)

D. Karaboga [30] proposed the Artificial Bee Colony (ABC) method in 2005. In several fields, including image processing, robot navigation, and parameter extraction it has proven to be an effective solution [31]. The process by which a bee colony gathers food can be thought of as an analogy for the ABC method. In this analogy, the position of each food source stands for a candidate solution, and the value of nectar that can be found in each food source is the fitness or objective of the corresponding solution [30].

This algorithm employs multiple groups (types) of bees (individuals or population members):

- Employed bees that search problem space for solutions (food sources).
- Onlookers bees that wait for a solution to be selected by employed bees.
- Scout bees that conduct a random search of other member's food sources.



**Fig. 2.** The network structure of MLP designed and used in modeling HDS process in this work.

**Table 2**  
Optimized hyper-parameters of MLP models.

Output	Hidden layer sizes	Activation	Solver	Tolerance
<b>Sulfur concentration</b>	94	relu	<i>lbfgs</i>	0.00051
<b>emission</b>	251	relu	<i>lbfgs</i>	0.022
<b>HDS cost</b>	23	identity	<i>lbfgs</i>	0.00041

**Table 3**  
Optimized hyper-parameters of GPR models.

Output	Alpha	Number of restarts optimizers
<b>Sulfur concentration</b>	$5.0 \times 10^{-6}$	5
<b>emission</b>	$4.99 \times 10^{-6}$	4
<b>HDS cost</b>	$1.80 \times 10^{-9}$	1

**Table 4**  
Optimized hyper-parameters of MTL models.

Output	Fit intercept	Tolerance	Alpha
<b>Sulfur concentration</b>	True	0.00058	3.96864
<b>emission</b>	True	0.036	0.00732
<b>HDS cost</b>	True	0.019	0.00212

**Table 5**  
Scores error rates final models.

Output Models	Sulfur concentration		Emission		HDS cost	
	Train	Test	Train	Test	Train	Test
MLP	0.999	0.983	0.999	0.971	1.0	1.0
GPR	0.999	0.942	0.999	0.864	0.999	0.990
MTL	0.757	0.790	0.818	0.738	1.0	0.999

**Table 6**  
Error rates final models.

Output Models	Sulfur concentration		Emission		HDS cost	
	MAE	RMSE	MAE	RMSE	MAE	RMSE
MLP	$7.69 \times 10^1$	$9.05 \times 10^1$	3.41	4.23	$5.57 \times 10^{-1}$	$6.58 \times 10^{-1}$
GPR	$1.23 \times 10^2$	$1.59 \times 10^2$	7.58	9.27	$1.69 \times 10^{-4}$	$1.58 \times 10^{-4}$
MTL	$3.11 \times 10^2$	$3.76 \times 10^2$	8.10	9.58	$1.38 \times 10^{-2}$	$1.63 \times 10^{-2}$

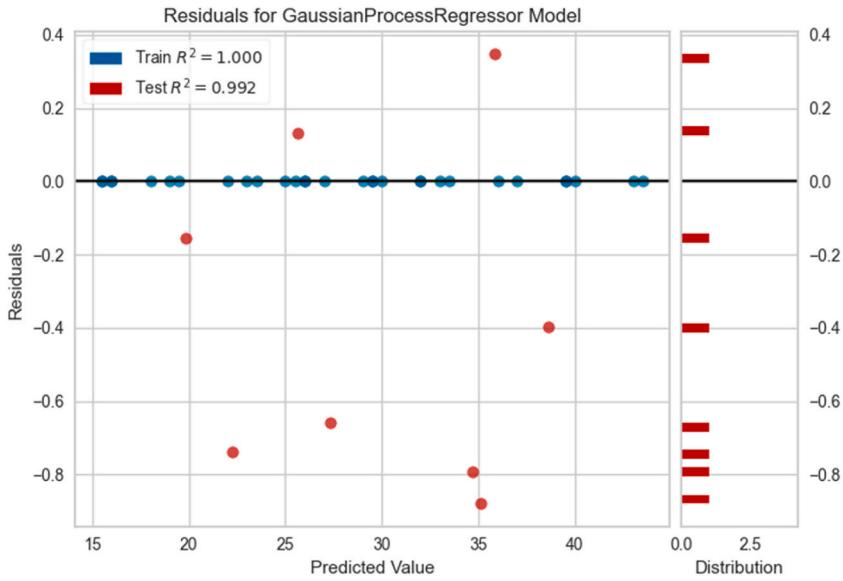


Fig. 3. Residuals of final model on HDS cost.

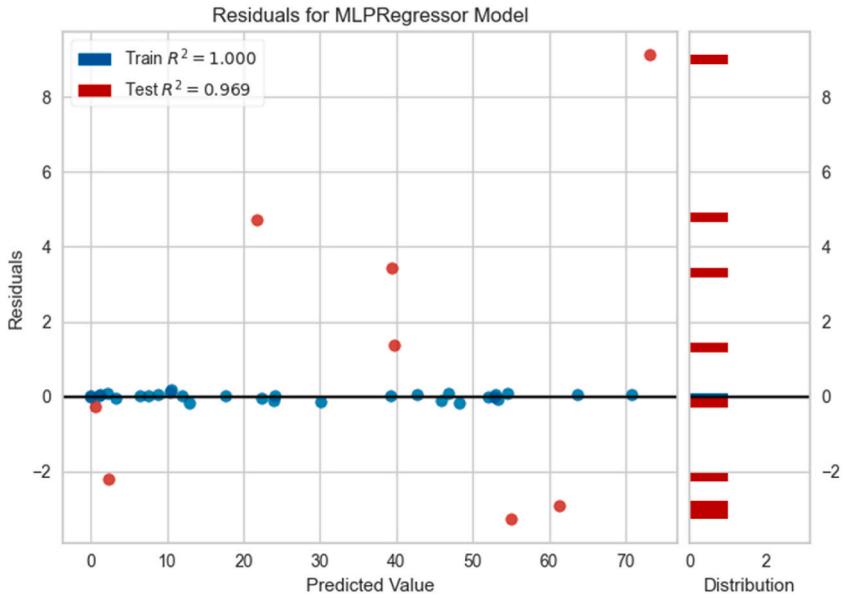


Fig. 4. Residuals of final model on emission percentage of SO<sub>2</sub>.

At the beginning of the process, half of the bees in the colony function as observers, and the other half as workers. Let  $N_p$  and  $D$  represent the overall population size and problem dimensionality, respectively. The count of busy bees is directly proportional to the total count of potential food sources that can be found in the area that is being investigated.

The following are the steps of this optimization algorithm [32]:

1. Initialize each bee colony solution ( $x_i$ ) and calculate the corresponding fitness values ( $f_i$ ) of them, where  $i=\{1,2 \dots, n\}$  and  $n$  denotes the quantity of utilized bees.
2. Using the following equation, generate a new solution,  $y_{ij}$ , given by the utilized bee in the neighborhood of  $x_i$  [32]:

$$y_{ij} = x_{ij} + \varphi(x_{ij} - x_{kj})$$

Where,  $k \in \{1, \dots, N_p\}$ ,  $j \in \{1, \dots, D\}$  are indices selected at random, and  $\varphi$  is a randomly selected number between  $-1$  and  $+1$ . Although  $k$  is chosen at random, it differs from  $i$  whereas  $x_k$  is the position randomly selected neighborhood of  $x_i$ .

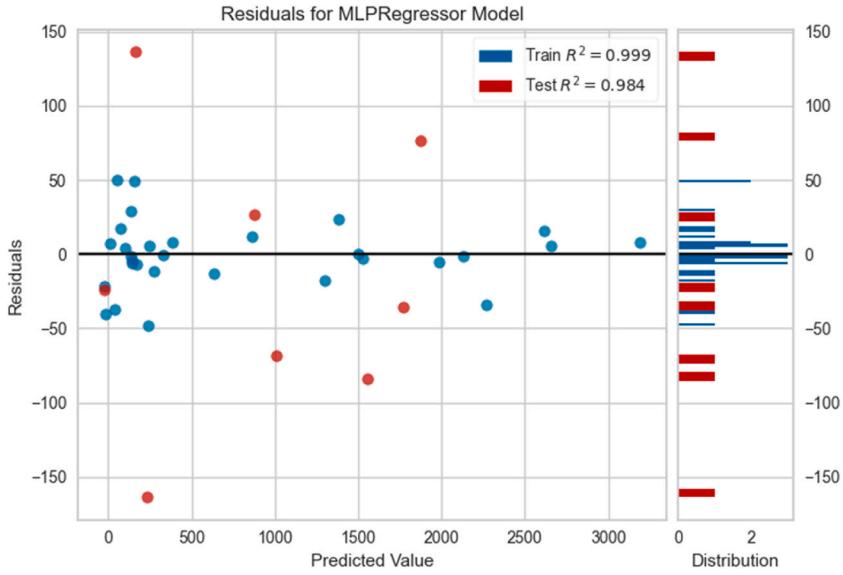


Fig. 5. Residuals of final model on Sulfur concentration in the final product.

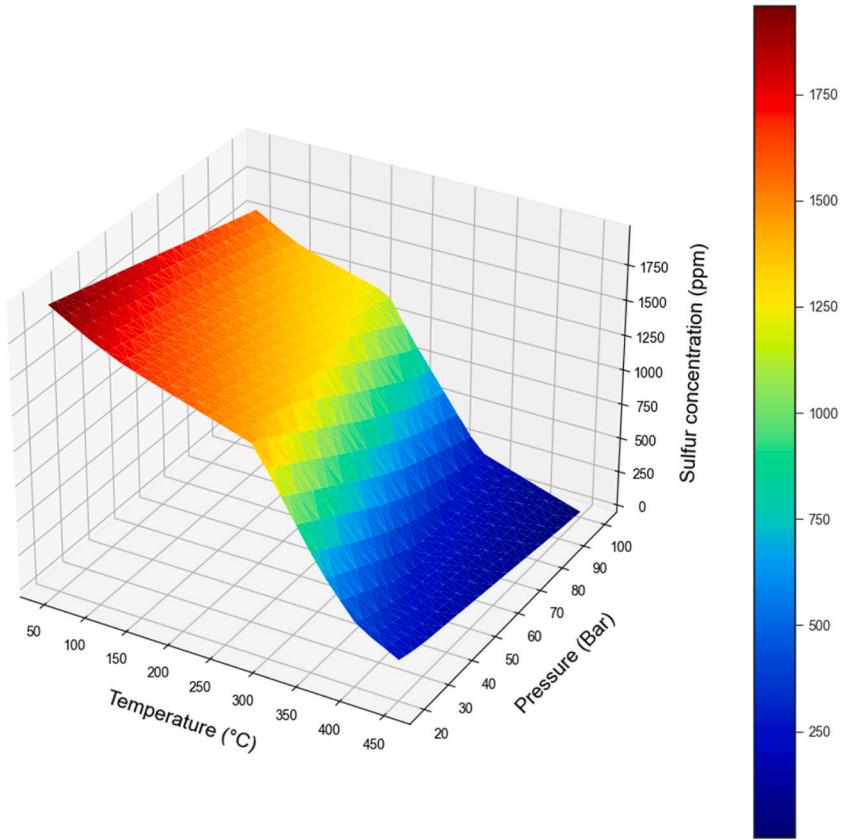
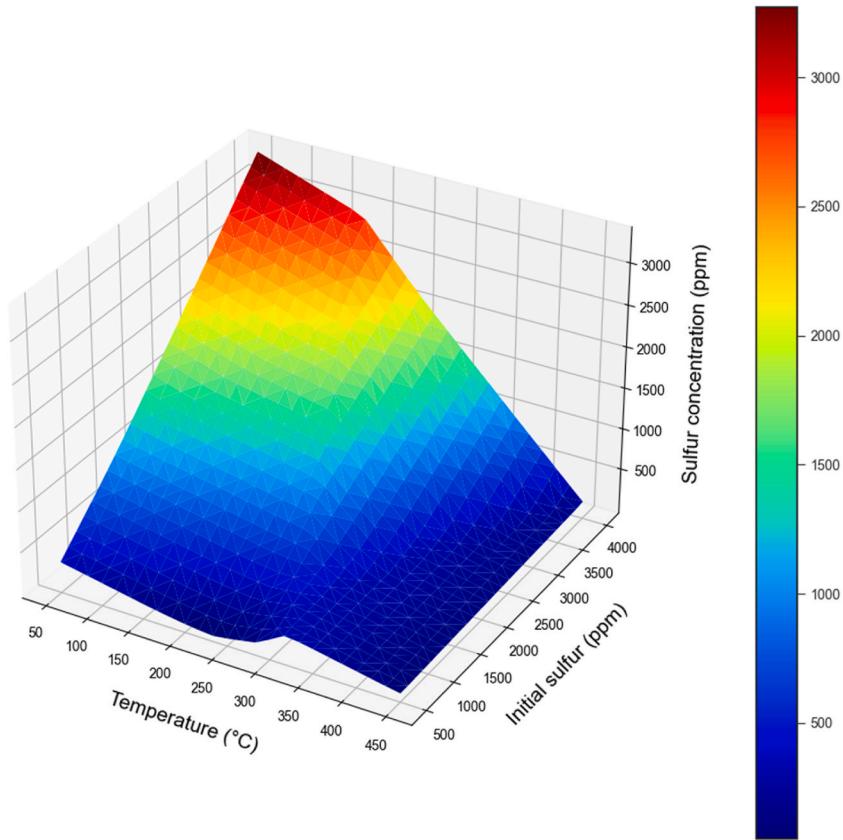


Fig. 6. 3d surface plot of Sulfur concentration variations versus Temperature and Pressure of reactor.

3. Replace  $x_i$  with  $y_i$  if the value of fitness for the new solution  $y_i$  is greater than the fitness of  $x_i$ .
4. Create a candidate solution depending on the nearby onlooker bees. Based on nectar data collected by all worker bees, an onlooker bee selects a food source with a probability  $\pi_i$  that is directly proportional to the value of nectar it provides [32]:



**Fig. 7.** 3d surface plot of Sulfur concentration (in the product) variations versus Temperature and Initial sulfur content.

$$p_i = \frac{f_i}{\sum_{i=1}^{S_N} f_i}$$

Here,  $S_N$  denotes the quantity of food sources and  $f_i$  represents the solution  $x_i$ 's fitness.

5. If the position fitness cannot be improved after a specified number of cycles (iterations), it will be abandoned, and the related employed bee will be promoted to scout bee.
6. Save the best fitness and its associated solution.
7. Return to step 2 if the desired fitness or stopping criteria is not reached.

As the main innovation aspect of this research, we have employed this algorithm to tune hyper-parameters and optimize them [33]. In fact, a food source (solution) is considered a combination of different values of hyper-parameters of the models, and optimization is done based on minimizing the error rate with the help of cross-validation.

### 3.2. MLP method

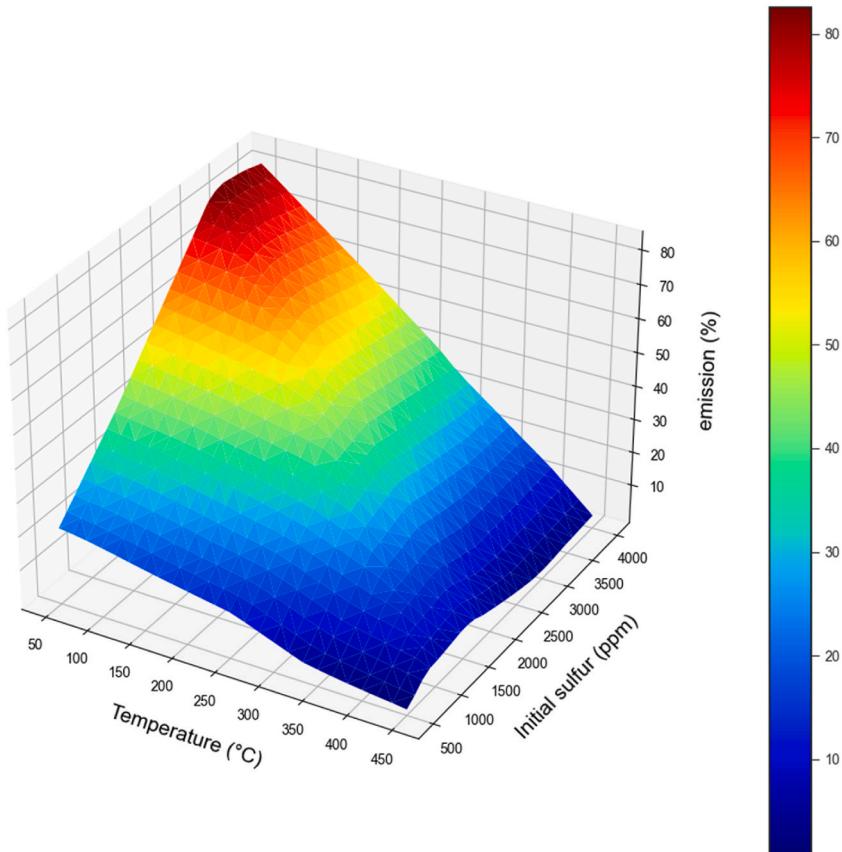
Similar to real-life neurons, MLP is a model used to predict neural activity. From a single-layer NN base, at least one hidden layer is added. In Fig. 2 we see the basic structure of an MLP network.

Input layer, hidden layer, and output layer are typical components of an MLP, and their interconnections are depicted in Fig. 1. Each node is a nonlinearly activated neuron, except for the input layer. MLP can approximate any nonlinear function by tuning the weights and activation functions. As part of its training and learning process, MLP uses Adam or lbfgs to iteratively tune the connection weight and offset in an effort to reduce the loss function.

The important hyper-parameters of MLP model for all three outputs of the dataset are listed in Table 2. These values are obtained using ABC algorithm.

### 3.3. Gaussian process regression (GPR)

Gaussian process regression (GPR) is a supervised learning approach that analyzes the functional space corresponding to the regression model as a Gaussian process:  $y = f(X, w)$ ,  $f \sim \mathcal{GP}(\mu, \kappa_\theta)$ , estimating the regression model parameters by learning the samples. As a non-parametric model with complete Bayesian features, Gaussian process regression model provides a posterior for the



**Fig. 8.** 3d surface plot of  $\text{SO}_2$  emission variations versus Temperature and Initial sulfur concentration.

predicted outcomes; when the likelihood follows a normal distribution, the posterior also has an analytic form, making GPR a generalizable and analyzed probabilistic model [26]. On top of that, GPR is a universal estimator for any function under certain criteria involving the kernel function and the exponential set. GPR has been applied to challenges in text mining, image processing, and lots of regression problems in scientific fields because of the easy nature of Gaussian processes and their kernel functions. GPR is a computationally intensive approach that is typically employed in regression task with small data sizes, however expanded algorithms exist for big sample and high-dimensional applications [34–36]. Table 3 lists the critical GPR model hyper-parameters for all three dataset outputs. These values were obtained by employing the ABC method.

#### 3.4. Multi-Task Lasso

The following equation can be used as an expression of the general linear regression model [37].

$$Y = \omega X + \epsilon, i = 1, 2, \dots, m$$

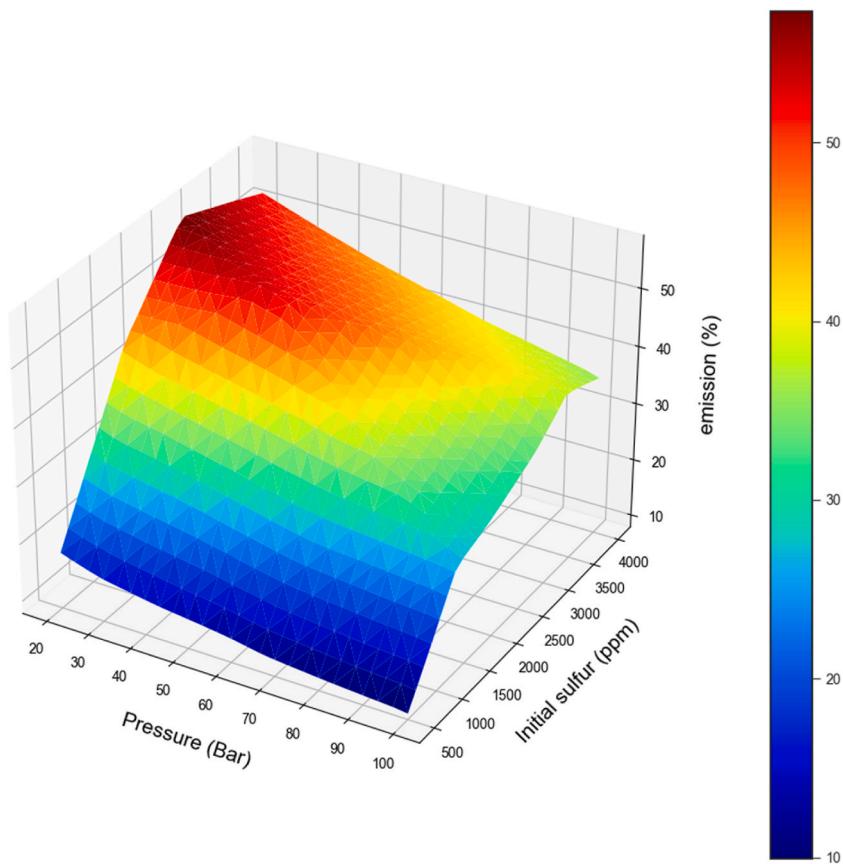
The variables  $m$ ,  $\omega$ , and  $\epsilon$  in the aforementioned equation all refer to the sample size, the weight, and the random error, respectively. As a standard technique, Ordinary Least Squares (OLS) can be used to build a loss function for linear regression and find values for the parameters. This function of loss is represented by the equation below [38].

$$\min_{\omega} \|\omega X - Y\|_2^2$$

Multi-task Lasso regression is a linear model trained with a mixed  $\ell_1 \ell_2$ -norm for regularization that takes into account simultaneous fitting of multiple linear regressions. Here is what we mean by the objective function [38]:

$$\min_{\omega} \frac{1}{2m} \|\omega X - Y\|_{\text{Fro}}^2 + \alpha \|\omega\|_{21}$$

The above formula can be optimized using the ABC method, where  $\|\omega X - Y\|_{\text{Fro}}$  Frois Frobenius Norm of the first equation,  $\|\omega\|_{21}$  is the sum of the root squares of the columns of  $\omega$ ,  $\alpha$  are the hyper-parameters. Table 4 lists the significant hyper-parameters of the MTL model for all three outputs of the dataset. These values were determined by the ABC algorithm.



**Fig. 9.** 3d surface plot of SO<sub>2</sub> emission variations versus Pressure and Initial sulfur concentration.

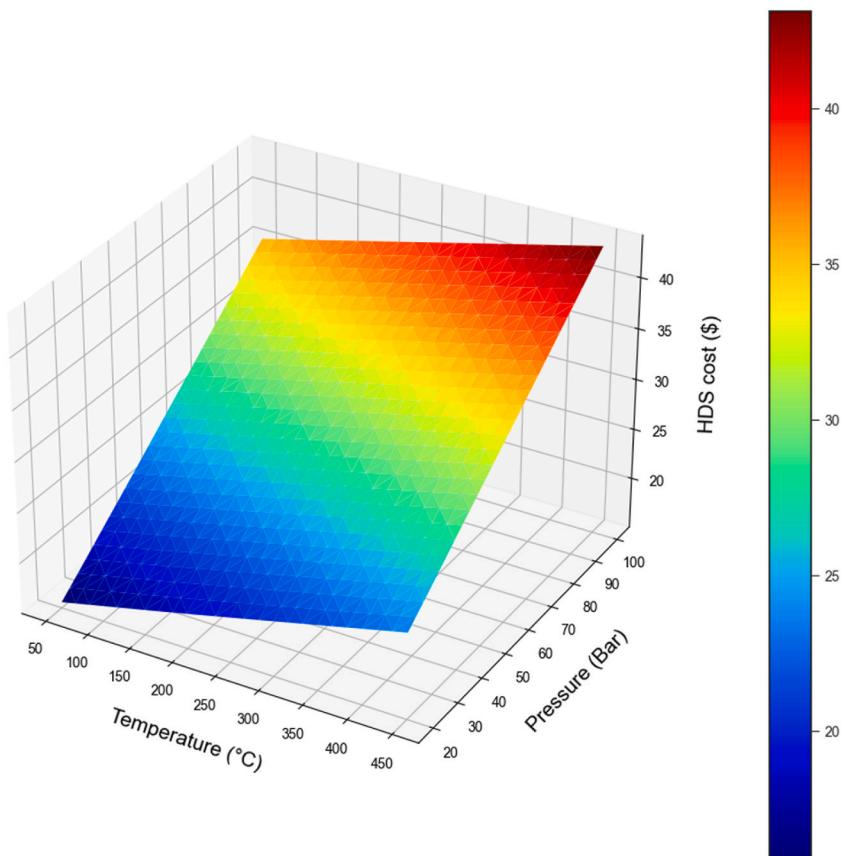
#### 4. Results & discussions

The models selected in this research have been implemented with the optimized values that are mentioned separately in the previous sections and have been evaluated and compared for all three outputs, the results of which are shown in [Tables 5 and 6](#). The modeling was conducted using Python package for the ML and optimization as well.

Based on these results, the MLP model has clearly shown the best accuracy for all three outputs as indicated by the statistical parameters listed in [Tables 2 and 3](#). But due to the higher complexity of this model, this model is somewhat suspicious of overfitting due to the higher error rate at the cost of HDS. Therefore, due to the relative closeness of the R<sup>2</sup> score of this model with GPR, as well as the lower error rate of GPR, this model has been selected for the analysis of the process.

In general, the GPR model has been chosen for the HDS cost and the MLP model for the other two outputs, and the residuals of the final models for all three outputs are shown in [Figs. 3–5](#). It is clearly observed that the deviations from the target points are not significant revealing the robustness and accuracy of the tuned ML models in prediction of the response parameters.

The developed and validated models were finally applied for design space creation and analysis of the inputs/output's parameters. The objectives of the models were to analyze the process and optimize the separation process which have been obtained using the developed ML models. The results of the 3D surface plot are illustrated in [Figs. 6–10](#). As seen, temperature has more impact on the separation efficiency of the HDS process, and the percentage of sulfur removal has been increased with enhancing the temperature of the reactor for desulfurization process. Increasing the reactor's temperature would enhance the chemical reaction rate between the sulfur and hydrogen in HDS process, which in turn results in consumption of more sulfur and its removal from the petroleum feed. On the other hands, it is known that increasing the temperature would decrease the physical solubility of gas in the liquid. Indeed, considering both effects, it is revealed that the chemical reaction is more dominant such that the solubility is enhanced with increasing the reactor's temperature. On the other hand, mass transfer needs to be taken into account for deeper analysis of the process due to the impact of temperature and pressure on the molecular diffusivity of gas in the gas phase as well as liquid phase diffusivity. It is perceived that both diffusion and convection have contributions to the total mass transfer of the species in the process. Furthermore, temperature and pressure both have a direct impact on the enhancement of process cost (See [Fig. 10](#)). Indeed, increasing temperature and pressure means more energy consumption in the HDS process, which finally would enhance the cost of sulfur separation [29].



**Fig. 10.** 3d surface plot of HDS cost versus Temperature and Pressure.

## 5. Conclusions

- HDS process was modeled in this work using multiple machine learning models to enhance the efficiency of process in desulfurization for petroleum industry. The process of HDS is utilized for purification of petroleum products such as fuels for protection of plant as well as environment.
- Based on the collected experimental data, some parameters were set as inputs to define the optimization task in this study for the process.
- The parameters of reactor pressure (bar), temperature (°C), initial sulfur content (ppm), and dosage of catalyst (g) are the four input features for each data point of HDS process.
- Outputs included sulfur concentration (ppm), emission (%), and HDS cost (\$).
- The core models in this study are multi-layered perceptron (MLP), Multi-Task Lasso (MTL), and Gaussian process regression (GPR). These models were created using an optimized Artificial Bee Colony (ABC) and were tested on a cleansed and normalized dataset.
- Sulfur concentration, emission, and HDS cost are predicted by  $R^2$  scores of 0.983, 0.999, and 0.990, respectively, using models proposed in this study, according to final model assessments. According to the analysis performed in the results section, these models can also guarantee the absence of overfitting.

## Author statement

Qikun MA: Conceptualization; Writing – original draft, Formal analysis, Methodology, Writing – Review & Editing, Investigation, Validation.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

All data are available within the published paper.

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