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RESEARCH ARTICLE / ARAŞTIRMA MAKALESİ

Predictive Modeling of the Syngas Production from Methane Dry Reforming over Cobalt Catalyst with Statistical and Machine Learning Based Approaches

İstatistiksel ve Makine Öğrenmeye Dayalı Yaklaşımlarla Kobalt Katalizör Üzerinden Metan Kuru Reformundan Elde Edilen Sentez Gazının Tahmini Modellemesi

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Abstract
Dry reforming of methane is a promising method to reduce the emission of CO_2 and to use it in various type of Fischer–Tropsch synthesis and production of syngas. In order to obtain desirable products efficiently, the effect of reactants on the products must be known precisely. For this purpose, several studies have published for modeling the dry reforming of methane process with artificial intelligence-based data-driven prediction models. Due to lack of investigating overfitting problem and deficient and/or biased performance evaluations, actual potential of proposed methods have not been revealed for predicting certain outputs of the process. In this paper, we employed three regression methods, i.e., artificial neural networks, support vector machine and polynomial regression to develop prediction models using a dataset with 57 observations. Performance evaluations of the models are performed with 10-fold cross-validation to ensure unbiased results. Proposed methods' both training and testing performances are separately investigated, further applicability is discussed.
Keywords*:* methane, dry reforming, support vector machine, artificial neural networks, polynomial regression, cross-validation

Öz
Metanın kuru reformlanması, CO_2 emisyonunu azaltmak ve çeşitli Fischer-Tropsch sentezlerinde ve sentez gazlarının üretiminde kullanmak için umut verici bir yöntemdir. İstenen ürünleri verimli bir şekilde elde etmek için, reaktantların ürünler üzerindeki etkisi kesin olarak bilinmelidir. Bu amaçla, yapay-zeka bazlı veri odaklı tahmin modelleri ile metan kuru reformunun modellenmesi için çeşitli çalışmalar yayınlanmıştır. Önerilen metotlar, aşırı uyum probleminin araştırılmamasından, eksik ve/veya yanlı performans değerlendirmelerinden dolayı, sürecin belirli çıktılarını tahmin etmek için yetersiz kalmıştır. 57 örnek içeren bir veri seti kullanarak destek vektör makineleri, yapay sinir ağları ve polinom regresyonu olmak üzere üç regresyon yöntemi kullandık ve tahmin modelleri geliştirdik. Modellerin performans değerlendirmeleri, tarafsız sonuçlar elde etmek için, 10 katlı çapraz doğrulama ile gerçekleştirilmiştir. Önerilen yöntemlerin hem eğitim hem de test performansları ayrı ayrı incelenmiş ve pratikte uygulanabilirliği tartışılmıştır.
Anahtar Kelimeler: metan, kuru reformlama, destek vektör makineleri, yapay sinir ağları, polinom regresyon, çapraz-doğrulama

I. INTRODUCTION

Due to the excessive use of fossil-based fuels and urbanization, CO_2 emissions which is the main cause of global warming have increased significantly [1]. With growing awareness about climate change, researchers put effort to develop effective methods to reduce the emission rate. Thus, dry reforming of methane has been proposed as a promising technique to not only reduce CO_2 emission but utilize it to produce syngas which can be processed into useful products such as electricity. Even though there are several catalysts that can be used during the dry reforming of methane such as Ni, Pt, Rh, Ru, Pd and Ir [2–4] , cobalt catalyst gained popularity due to its wide availability and cost efficiency [5]. Certainly, the dry reforming processes must be handled carefully by using precise models to obtain optimal efficiency. Therefore, several studies are published which focusing on kinetic modeling of dry reforming of methane [6] and to best of our knowledge there are limited studies focusing on using artificial intelligence based methods. In Hossain et.al. [7], authors performed dry reforming of methane with $Ni/CaFe_2O_4$ catalyst and obtained a data set with 24 observations to create an artificial neural network model, they

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used feed ratio reaction temperature and metal loading as features and tried to predict CH_4 conversion, CO_2 conversion, H_2 yield and CO yield. They split the dataset into training, testing and validation sets during the training of the model. Even though they obtained satisfactory results, with the use of data set with an extremely low number of observations and a low number of features resulted with inadequacy to explain proposed methods' true potential and generalization ability of the process. In Amin et. al. [8], authors did methane reforming with Rh/MgO catalyst experiment and used their results to develop an artificial neural network model. But the authors used all of the data for training the model and evaluated the performance of the model using the same data set. Thus, the generalization ability of the model is not investigated and proper performance evaluation is not achieved. In Al Ayodele et. al [2] performed methane dry reforming over ceria-supported cobalt catalyst and gathered a data set with 57 observations, they used CH_4 partial pressure, CO_2 partial pressure CH_4 to CO_2 ratio and reaction temperature as features to create an artificial neural network model in order to predict H_2 and CO production rates as well as CH_4 and CO_2 conversion percentages. They also split the dataset into training, testing and validation folds but they only shared the training performance of the model and used a small number of error metrics for evaluation. Due to the lack of test and validation scores and investigation of overfitting, the study wasn't able to explain the accuracy and practical usability of the proposed method. There are limited number of studies have been reported in the literature in order to improve catalyst for dry reforming of methane using artificial neural networks [9,10]. However, prediction error was not given in these studies. Recently, Şener et. al [11] were analyzed the effect of catalyst type, support, preparation method and tried to find the best catalyst for optimum methane conversion. They used 4-fold cross validation method (reserving one fourth of data for testing and repeating this procedure four times to cover entire range) and found RMSE in the range of 4.52 to 8.59. In Huang et. al. [12], authors employed artificial neural networks and genetic algorithm in hybrid fashion. Although their training error was quite low, experimental results had up to %28 error with the predicted values, thus, proposed model showed unreliability in practical applications.

In this paper, we used the same dataset published in [2] to create artificial intelligence-based prediction models. We employed three methods, i.e. polynomial regression, artificial neural networks and support vector regression methods to predict outputs of the dry reforming of methane with a ceria-supported cobalt catalyst. As a contrary to previous studies, performance evaluation of the employed methods is performed with 10-fold cross-validation technique. This approach not only enabled us to explore each methods' generalization performances, also allowed us to make comparison between the employed machine learning methods in unbiased manner. All methods' training and testing performances, as well as further usability, are discussed.

II. MATERIAL AND METHODS

2.1 Dataset Acquisition

In this study, dataset published in [2] is used. The dataset contains 57 observations and four numerical features , x_1, x_2, x_3, x_4 referred to as, CH_4 partial pressure (kPA) , CO_2 partial pressure (kPA), CH4:CO2 ratio, Reaction Temperature (°C), respectively. These features are utilized to develop models to predict four numerical outputs, y_1, y_2, y_3, y_4 i.e., r_{h_2} (mmol/min/g catalyst), r_{CO} (mmol/min/g catalyst), CH_4 conversion (%), CO_2 conversion (%).

2.2 Polynomial Regression

Polynomial regression is one of the regression techniques which represents the relationship between the independent variables (features) and the dependent variable (output) with an n^{th} (n = 2, 3…) degree polynomial equation. The goal of polynomial regression is to find optimal coefficients that can make satisfactory predictions of the output variable in terms of features used in a data set, i.e. good generalization and prediction performance. The choice of the degree and type of the polynomial equation is differing according to the problem which one would like to develop a solution for. In the present paper, the quadratic equation (Equation (1)) is determined to be the most appropriate polynomial equation to apply to the dataset used

$$\hat{y} = \sum_{i=1}^{n}\sum_{j=i}^{n} \beta_{ij} * x_i * x_j + \sum_{k=1}^{n} \alpha_k * x_k \qquad (1)$$

where, $\hat{y}$ is the prediction of the output variable, n is the number of features, β_{ij} and α_k are the coefficients of the polynomial equation. While developing a regression model, a cost function which indicates the prediction accuracy must be selected and minimized during the training phase. For this purpose, commonly used convex cost function, i.e. sum of square errors is selected and used in the present paper. In order to prevent model from learning excessively, which reduces the generalization performance of the proposed method [13], also referred to as overfitting, we add a regularization term to the cost function J (Equation (2)). Minimization of the cost function is performed with Gradient-Descent iterative optimization method [14],

$$J = \sum_{i=1}^{m} (y_i - \hat{y}_i)^2 + \lambda \left(\sum_{j=1}^{n}\sum_{k=j}^{n} {\beta_{jk}}^2 + \sum_{t=1}^{n} {\alpha_t}^2 \right) \qquad (2)$$

where, λ is the regularization parameter.

2.3 Artificial Neural Networks (ANN)

ANN is a machine learning methodology inspired by the human brain and its information processing structure [15]. ANN has found widespread usage across different type of problems in various disciplines due to its proven success for predicting both continuous and discrete variables. ANN consist of neurons which are the primary processing elements, and neuron clusters which are called as layers. Neurons receive inputs from neurons in the previous layer, process the information with its activation function and transmit output for the neurons in next layer [16], this transmitting process starts with input layer and continuous until the neurons in the output layer produces outputs. Every layer between input and output layers are called hidden layer, the number of hidden layers and number of neurons on each hidden layer is hyper-parameters which must be determined beforehand. Training of a neural network is performed by adjusting the weights between each connection which minimizes the difference between predictions and the actual output and widely used backpropagation algorithm is used for training phase [17]. One of the most unique features of the ANNs is the capability of predicting multiple output variable with the same model, because the dataset used in this paper consists of four features and four output variables. We developed a single ANNs model to predict all of the outputs with two hidden layers and nine neurons for each hidden layer as shown in Fig. 1. Sigmoid activation function and linear activation function is used in hidden layers and output layer, respectively.

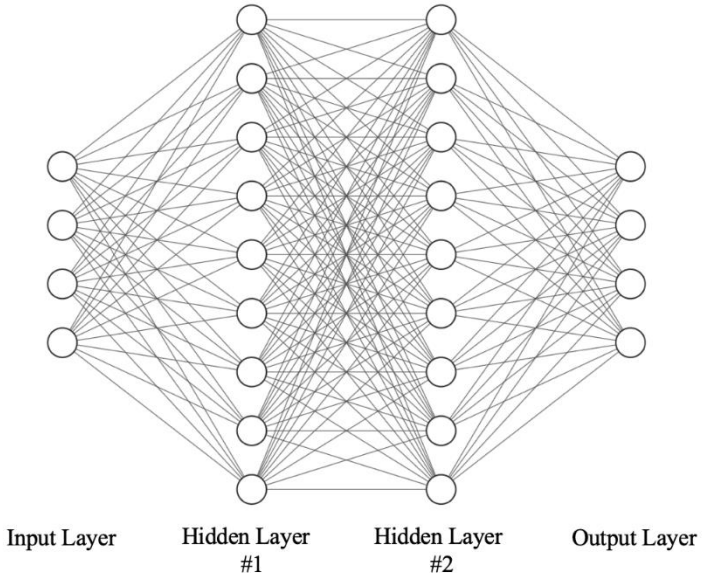

Figure 1. Developed ANNs structure

2.4 Support Vector Regression

Support Vector Machines (SVM) is a popular machine learning technique due to its non-parametric structure and usage of kernel functions. Support Vector Regression (SVR) is a specific type of Support Vector Machines (SVM) technique which is used to predict continuous type variables. Contrary to polynomial regression and ANNs, SVR models are trained by minimizing the generalization error bound instead of prediction error [18]. Therefore, SVR models try to achieve the best generalization performance. The principle of SVR is to transform
data to a higher dimensional feature space with a kernel function and to find an optimal hyperplane which minimizes the selected cost function [19]. Therefore, SVR performs a linear regression in higher dimensions by using a kernel function (K) as shown in Eq. (3)

$$\hat{y} = \sum_{i=1}^{n} \beta_i * K(x_i, x) + c \qquad (3)$$

where, β_i and c are the constant coefficients, $K(x_i, x)$ is the kernel function. In this paper, we used a second-degree polynomial kernel function (Eq. 4). The reason why we selected this kernel function is, linear and gaussian kernel functions were not able to grasp the complexity of the problem, thus, poor prediction performance is obtained by those kernels. When third or higher degree polynomial kernels are used, computational expense significantly increased and performance improvement was negligible and didn't justify the model's enormous training time. During the

training phase of the models, epsilon-insensitive cost function (Eq. 5, Eq. 6) is used and minimized.

$$K(x_i, x) = (x_i' x + 1)^2 \quad (4)$$

$$L_\varepsilon = \begin{cases} 0 & if\ |y - \hat{y}| \le \varepsilon \\ |y - \hat{y}| - \varepsilon & otherwise \end{cases} \quad (5)$$

$$J = \sum_{i=1}^{n} L_\varepsilon(\hat{y}_i, y) + \frac{\lambda}{2} |w|^2 \quad (6)$$

where w is the weight matrix.

III. RESULTS AND DISCUSSION

Development and performance evaluation of the proposed methods are performed in MATLAB environment. Due to the small size of the data set, we implemented 10-fold cross validation technique for and testing. Feature normalization on the data set is performed to prevent suppressing of a feature due to another feature with high magnitude. Prediction models for polynomial regression and SVR develop to predict single output variable, for ANNs all outputs predicted at once. Correlation Coefficient (R^2), Adjusted Correlation Coefficient (Adj.R^2), Root-Mean-Square-Error (RMSE) and Normalized Root-Mean-Square-Logarithmic-Error (RMSLE) metrics are used to show the prediction accuracy of the proposed methods. Each methods' training and test scores of 10-fold cross validation are given in Table 1 and Table 2, respectively.

$$R^2 = 1 - \left[\frac{\sum_{i=1}^{m}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{m}(y_i - \bar{y})^2}\right] \quad (7)$$

each method to ensure unbiased performance evaluation [16,17], the data set was split into ten folds and model is trained with nine of the folds and tested on the remaining one. This process is repeated for each fold, so that, all of the data is used for both training

$$Adj.R^2 = 1 - \left[\frac{(1 - R^2) * (m - 1)}{m - n - 1}\right] \quad (8)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{m}(y_i - \hat{y}_i)^2}{m}} \quad (9)$$

$$RMSLE = \sqrt{\frac{\sum_{i=1}^{m}[log(y_i + 1) - log(\hat{y}_i + 1)]^2}{m}} \quad (10)$$

Table 1. Training performances of proposed methods

Outputs	Polynomial Regression				ANNs				Support Vector Regression			
	R^2	$Adj.R^2$	RMSE	RMSLE	R^2	$Adj. R^2$	RMSE	RMSLE	R^2	$Adj.R^2$	RMSE	RMSLE
y_1	0.87	0.84	0.797	0.294	0.7	0.61	1.253	0.334	0.73	0.64	1.191	0.351
y_2	0.96	0.95	0.365	0.073	0.76	0.69	0.975	0.199	0.95	0.94	0.426	0.097
y_3	0.87	0.83	5.24	0.108	0.94	0.92	3.505	0.079	0.82	0.77	6.067	0.119
y_4	0.78	0.72	8.323	0.179	0.9	0.87	5.555	0.122	0.71	0.66	9.854	0.259

Table 2: Test performances of proposed methods

Outputs	Polynomial Regression				ANNs				Support Vector Regression			
	R^2	$Adj.R^2$	RMSE	RMSLE	R^2	$Adj. R^2$	RMSE	RMSLE	R^2	$Adj.R^2$	RMSE	RMSLE
y_1	0.8	0.73	1.025	0.295	0.61	0.54	1.416	0.428	0.55	0.39	1.554	0.412
y_2	0.94	0.92	0.501	0.073	0.72	0.64	1.065	0.229	0.91	0.89	0.562	0.127
y_3	0.57	0.47	9.692	0.182	0.67	0.56	8.462	0.184	0.65	0.54	8.672	0.165
y_4	0.61	0.52	11.21	0.222	0.42	0.24	13.68	0.287	0.46	0.3	13.15	0.287

One can see that the overall prediction performance of polynomial regression was better compared to other methods in both training and test phases. For output labeled as y_1, all methods performed reasonably in the training phase. In the testing phase, only polynomial regression models were able to make predictions close to its training performance. For output y_2, the performance of polynomial regression and SVR

methods was similar and satisfactory in both training and test phases, but the performance of ANN was significantly lower. For outputs y_3 and y_4, all of the methods showed a major performance decline in the testing phase compared to training. This situation usually indicates overfitting, but during developments of the models, different regularization term constants have been tested for each method to overcome overfitting while avoiding underfitting. Moreover, this performance reduce was due to the low number of observations in the data set which means methods were not able to 'understand' the dynamics of the process with the given number of training data. Therefore, it can be concluded that more observation is required for proposed techniques to make predictions, which can show the true nature of the process. In addition, it should be stated that training performance can be highly inaccurate in the evaluation of the machine learning models. Extremely high performance in the training phase, as shared in [2], can be misleading and developed models may perform unexpectedly worse when models encounter with data they have never seen before, as can be seen in the present paper.

In order to visualize performance evaluations and to have a better understanding of predictions made by polynomial regression, which is the best performing method in this paper, prediction vs actual value graphs are plotted for both training and test phases (Fig. 2 and Fig. 3). Graphs are plotted with a reference line which starts from zero and has slope equals to one. Therefore, any point stays on the reference is predicted correctly, and length between unsuccessful predictions and reference line indicates the prediction error.

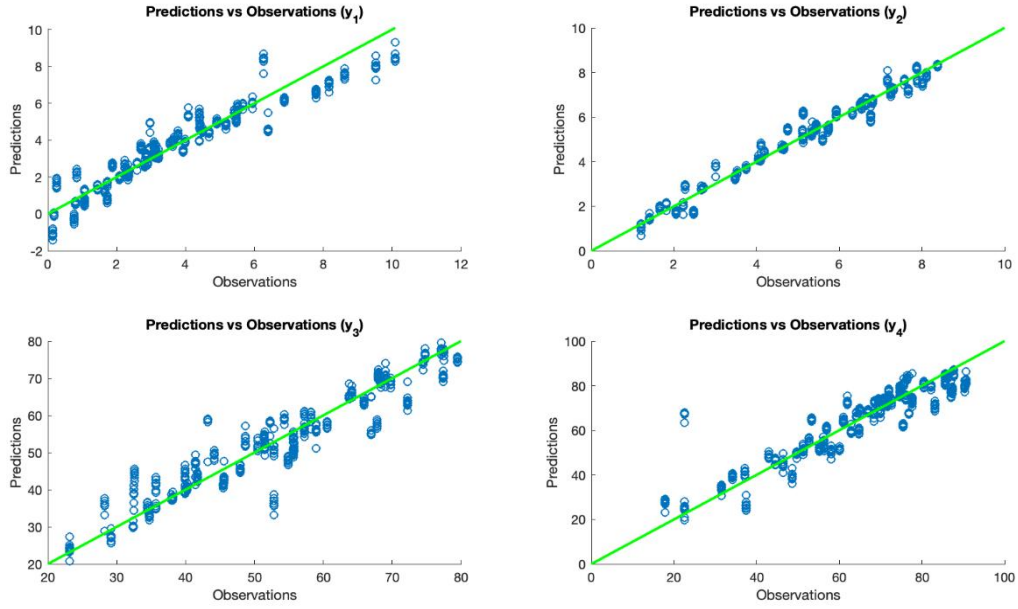

Figure 2. Prediction performance of polynomial regression in training phase

As one can see, a total of 513 data points (57*9) for the training set and 57 data points for the test set are shown in the Figure 2 and Figure 3, respectively. The reason why 513 data points are presented in Figure 2 is due to the nature of 10-fold cross-validation. Every fold is used nine times in the training phase and used as test set once. Therefore, in Fig. 2, every data point which shares the same value in 'Observations' axis is the same prediction of the corresponding observation made by different folds. the small distance between them shows that the desired attributes such as high stability and low variance between folds are achieved in the training phase. When Fig. 3 is examined, predictions follow close to the identical pattern obtained Fig. 2 for outputs y_1 and y_2, which means developed models are successfully avoid overfitting and learned enough information from training data to make accurate predictions. For outputs y_3 and y_4, even the prediction patterns are similar, some of the points are predicted extremely inaccurate which resulted in low prediction accuracy overall. In essence, one can observe polynomial regression and SVR predicted the H_2 and CO production rates with high accuracy in both training and testing phases, on the other hand, ANNs failed to make satisfactory predictions for these outputs. Even though all of the methods performed well in during training phase for CH_4 conversion and CO_2 conversion outputs, they performed significantly worse in the testing phase due to the low number of observations in the dataset. Although ANNs method didn't perform as good as other proposed ones, its performance may increase with the use of different

architectures and hyper-parameters, but when the requirement of high development time and computational resource of ANN is concerned, it may not be the best choice of method for predicting outputs of dry reforming processes. Even though SVR performed better, slightly worse than polynomial regression, it can be considered an overkill due to high memory and computational requirement of the method. Present study shows that polynomial regression's overall performance was best among the proposed methods and with low computational cost of polynomial regression in mind, it can be a good candidate to simulate and make predictions for dry reforming processes with cobalt catalyst especially with larger data sets efficiently.

When the results obtained in this study are compared to similar studies in literature; In [2], authors only provided training error of their proposed prediction models. This practice not only prevent showing the true potential of the methods by not feeding and testing the - unseen data (test or validation data), it also fails to investigate models' generalization ability. In [11], authors evaluated the performance of their models by 4-fold cross validation, even though 10-fold cross validation would be more suitable due to high sample size of the data set they used, generalization ability of proposed models are deeply investigated. They calculated RMSE values between 4.52 to 8.59 which is slightly worse performance compared to this study as shown in Table 2. In [12], authors not only used any kind of cross-validation methods, also they obtained overfitted model with test results up to %28 error shown in the paper. This significantly worse than what we achieved in this paper. As similar studies, in [9] and [10], authors did not provide any kind of error metrics other than cost function of the ANN which does not offer any kind of insight about the prediction ability of the proposed models.

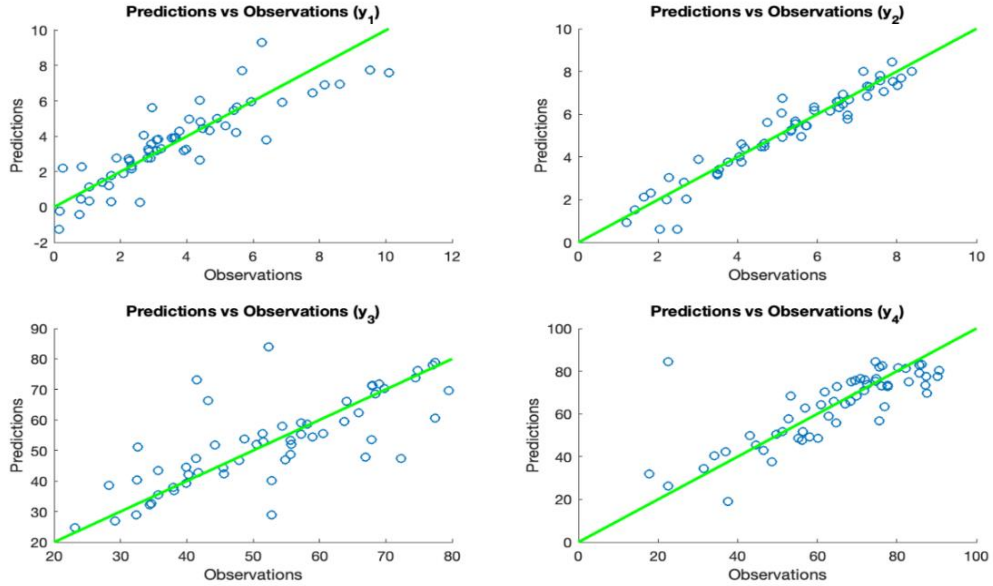

Figure 3. Prediction performance of polynomial regression in testing phase

IV. CONCLUSION

In this paper, polynomial regression, ANNs, and SVR are used to predict four outputs of methane dry reforming processes with cobalt catalyst using four features. It is observed that ANN failed to make accurate predictions compared polynomial regression and SVR. Polynomial regression's performance showed that it is a strong candidate to create prediction models for such problem when the overall accuracy and computational costs are taken into the account. Although, performance of the polynomial regression was better among all proposed methods, still, highly robust predictive models could not be developed for prediction of CH_4 conversion, CO_2 conversion outputs due to the low sample size of the data set used. For future work, we'll focus on implementing the proposed machine learning algorithms on larger datasets and use statistical techniques to determine the impact of each feature which may improve performance of the machine learning methods for the methane dry reforming processes with cobalt catalyst.

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