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Research Article

Zeolite Used for Optimized Removal of Radioactive Cobalt with Response Surface Methodology

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ABSTRACT

Radioactive cobalt is one of the most abundant radionuclides in radioactive waste. This study investigated the removal of radioactive cobalt (⁶⁰Co) by adsorption with zeolite 3A and 5A from aqua solution. The response surface methodology was employed to constitute the predictive regression model to guess the decontamination factor for radioactive cobalt removal. The experimental maximum decontamination factor 30.37 and 15.9 were obtained for zeolite 3A and zeolite 5A, respectively. The calculated model was significant for both zeolite 3A and 5A ($p < 0.05$). The predicted maximum decontamination factor was 30.05 and 15.19 in optimum conditions for zeolite 3A and zeolite 5A, respectively. Zeolite 3A has a higher adsorbent capacity than zeolite 5A for the removal of radioactive cobalt from aqueous solution.

Keywords: Radioactive cobalt, Adsorption, Response surface methodology, Zeolite 3A, Zeolite 5A

Zeolit Kullanılarak Uzaklaştırılan Radyoaktif Kobaltın Cevap Yüzey Yöntemi ile Optimize Edilmesi

ÖZET

Radyoaktif kobalt, radyoaktif atıktaki en bol bulunan radyonüklidlerden biridir. Bu çalışmada, radyoaktif kobaltın (⁶⁰Co) su çözeltisinden zeolit 3A ve 5A ile adsorpsiyon yoluyla uzaklaştırılması araştırılmıştır. Cevap yüzey yöntemi, radyoaktif kobaltın uzaklaştırılmasında dekontaminasyon faktörünü tahmin etmek için öngörücü regresyon modelini oluşturmada kullanılmıştır. Deneysel maksimum dekontaminasyon faktörü zeolit 3A ve zeolit 5A için sırasıyla 30.37 ve 15.9 olarak elde edilmiştir. Hesaplanan model zeolit 3A ve 5A için anlamlıydı ($p < 0.05$). Zeolit 3A ve zeolit 5A için tahmin edilen maksimum dekontaminasyon faktörü, optimum koşullarda sırasıyla 30.05 ve 15.19'dur. Sulu çözeltiden radyoaktif kobaltın uzaklaştırılmasında zeolit 3A, zeolit 5A'dan daha yüksek adsorban kapasitesine sahiptir.

Anahtar Kelimeler: Radyoaktif kobalt, Adsorpsiyon, Cevap yüzey yöntemi, Zeolit 3A, Zeolit 5A

I. INTRODUCTION

The radioisotopes are delivered into the environment through the release of nuclear waste sewage [1]. A large number of nuclear wastes was produced by nuclear facilities. These wastes are substantial risks to the environment [2]. Many radioactive elements have a long time of half-life. They are not going to be neutralized easily [3]. Reduction of radioactive waste is necessary owing to human health problems accompanied by the aggregation of radioactive elements [4].

^{60}Co has many beneficial applications such as materials check and sterilization. It is used to treat cancer and to sterilize medical equipment in medicine. The half-life of ^{60}Co is 5.27 years. It is the longest-lived isotope of cobalt. Owing to its long half-life, the removal of radioactive cobalt is getting more attention from researchers [5]. Elimination of ^{60}Co from aqueous media has received great interest recently. There are many techniques to dispose of radionuclides from wastewaters [6].

One of the most effective strategies to minimize nuclear waste volumes is the selective separation of radioisotope from nuclear wastes [2]. Adsorption is favorable amid the methods owing to its easiness, thermal resistance, specific cation selectivity, and low cost [1]. Adsorbents such as zeolite have become interesting for radioactive waste management recently [7].

Zeolites are hydrated aluminosilicate minerals with crystalline microporous. Zeolite contains enormous alkaline metal ions. They have microporous and their frameworks are charged. Their anionic tetrahedral framework structures made them extremely hydrophilic sorbents. Their cavities are occupied by water molecules and charge balancing cations [2, 8].

Response surface methodology (RSM) is a famous experimental design method for optimization. It is a mathematical system for statistical modeling of problems [9]. It is utilized to examine the interaction between the independent variable and the dependent variable [10]. It is used for the optimization of each input variable for optimal conditions. RSM is convenient to optimize the adsorption experiments [10-12].

The ever-increasing pressure to diminish the amount of radioactivity in the environment requires constant improvement of processes for the treatment of radioactive waste. The toxic nature of radioactive cobalt has been a public health problem for many years. Thence, the studies of the removal of radioactive cobalt from wastewater are important [13]. A systematic understanding of how the amount of sorbent and radioactivity participate in the adsorption process of zeolite in radioactive wastewater is still lacking [14]. The non-radioactive cobalt adsorption on zeolite 3A and 5A were studied previously [15]. This study aims to show how effectively zeolite can absorb ^{60}Co with different concentrations of solids and radioactivity. This research is focused on the thermally activated zeolite applied for effective disposal of radioactive cobalt ions from aqua solution. The adsorption process of radioactive cobalt by zeolite 3A and 5A adsorbents were exploited under experimental conditions. RSM was used to examine the relationships between input variables and response. The input variables are the amount of adsorbent and the initial activity of a solution.

II. MATERIALS AND METHOD

3A and 5A zeolites were obtained from Sigma-Aldrich. Both of them (3A and 5A) were powder. The molecular formula of zeolite 3A and 5A are shown as a fellow (respectively):



Zeolites were activated with temperature for two hours at 873 K to remove water from them. Thereafter, samples were added to the radioactive cobalt and suspensions were mixed for 4 h. Afterward, the mixtures were filtered with a syringe filter. The activities of radioactive cobalt were measured with Polon Warszawa Analyzer (A-22p HT Power supply ZW N-21M HT Control 0/2000V). A volume of 10 mL solution was used for all liquid radioactive cobalt reading. The radioactive measurements were repeated 10 times. The experimental decontamination factor was presented as:

$$DF = A_0 / A_f \quad (3)$$

where A_0 was the initial activity of radioactive cobalt feed solution (Bq/L) and A_f was the final activity of radioactive cobalt (Bq/L).

More information related to the experiment can be seen in previous studies [16, 17].

RSM was employed for analysis and modeling. RSM is a helpful statistical and mathematical technique as indicated by previous studies in detail [16-19].

In the RSM model, response (radioactive decontamination factor) can be described which is influenced by controllable various input factors.

The general form of RSM can be shown as:

$$\hat{DF} = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{12} x_1 x_2 \quad (4)$$

$$\beta = [\beta_0 \beta_1 \beta_2 \beta_{11} \beta_{22} \beta_{12}]^T \quad (5)$$

$$\beta = (X^T \cdot X)^{-1} \cdot X^T \cdot DF \quad (6)$$

where: β – present regression coefficients;
 X – input variables;
 DF – decontamination factor (response).

Adsorption of radioactive cobalt onto zeolite 3A;

- regression model with coded variables:

$$\hat{DF}_{3A} = 12.36 + 6.86x_1 - 2.20x_2 + 6.74x_1^2 - 1.51x_2^2 - 3.40x_1x_2 \quad (7)$$

subjected to: $-1 \leq x_i \leq +1, \quad \forall i=1,2$

- regression model with actual variables:

$$\hat{DF}_{3A} = -1.8 - 198SD + 0.00360A_0 + 2696SD^2 - 0.01792SDA_0 \quad (8)$$

valid for the range: $0.05 \leq SD \leq 0.15$ (%w/v); $7600 \leq A_0 \leq 15200$ (Bq/L)

Adsorption of radioactive cobalt onto zeolite 5A;

- regression model with coded variables:

$$\hat{DF}_{5A} = 7.995 + 4.280x_1 + 1.090x_2 + 0.599x_1^2 + 0.630x_2^2 + 0.368x_1x_2 \quad (9)$$

subjected to: $-1 \leq x_i \leq +1, \quad \forall i=1,2$

- regression model with actual variables:

$$\hat{DF}_{5A} = 6.44 + 15.6SD - 0.00090A_0 + 240SD^2 + 0.00194SDA_0 \quad (10)$$

valid for the range: $0.05 \leq SD \leq 0.15$ (%w/v); $7600 \leq A_0 \leq 15200$ (Bq/L)

The modeling details can be found in previous studies [16-20]. Minitab 19 software was utilized for all calculations.

III. RESULTS AND DISCUSSION

The ANOVA was used to examine the accuracy of the calculated model. According to the results, the model was compatible where the p-value was 0.034 ($p < 0.05$) for zeolite 3A. This means that the calculated model is significant for zeolite 3A. F value was 7.87, it was shown that the experimental decontamination factor obtained by changing the factor levels were statistically meaningful at the 91% confidence limit. R^2 value should be close to 1 for a good statistical model. The mathematical model is adequate for the prediction radioactive cobalt removal by zeolite 3A sorption since $R^2 = 0.91 > (0.75)$. Lack of fit F and p values are 6.93 and 0.271 ($p > 0.05$), respectively.

For zeolite 5A calculated model p-value was 0.015 ($p < 0.05$) means that the model is significant. The model F value was 12.67, it was shown that the experimental decontamination factor obtained by changing the factor levels were statistically meaningful at the 94% confidence limit. The mathematical model is adequate for the prediction radioactive cobalt removal by zeolite 5A since $R^2 = 0.94 > (0.75)$. Lack of fit F and p values are 37.39 and 0.120 ($p > 0.05$), respectively. Lack of fit was not significant and this means that the model is good [20, 21].

Table 1. Radioactive cobalt experimental design for zeolite 3A

| Run number (N) | Factors (input values) | | | | Response | | |
|-------------------|-----------------------------------|------------------------------------------------|---------------------------------|--------------------------------------------|--------------------------------|-----------|-----------|
| | Amount of sorbent (g/100ml) | Initial activity of radioactive cobalt Bq/L | | Final activity Bq/L | Decontamination Factor (DF) | | |
| | | Experimental | Predicted | | | | |
| | Sorbent Dosage (% w/v) | level^a x₁ | A₀ (Bq/L) | level^a x₂ | | DF | DF |
| 1 | 0.15 | 1 | 15200 | 1 | 952.98 | 15.95 | 18.83 |
| 2 | 0.05 | -1 | 15200 | 1 | 1374.32 | 11.06 | 11.93 |
| 3 | 0.15 | 1 | 7600 | -1 | 250.25 | 30.37 | 30.05 |
| 4 | 0.05 | -1 | 7600 | -1 | 640.81 | 11.86 | 9.53 |
| 5 | 0.15 | 1 | 11400 | 0 | 399.86 | 28.51 | 25.95 |
| 6 | 0.05 | -1 | 11400 | 0 | 1057.51 | 10.78 | 12.24 |
| 7 | 0.1 | 0 | 15200 | 1 | 1226.80 | 12.39 | 8.64 |
| 8 | 0.1 | 0 | 7600 | -1 | 731.47 | 10.39 | 13.05 |
| 9 | 0.1 | 0 | 11400 | 0 | 887.16 | 12.85 | 12.36 |
| 10 | 0.1 | 0 | 11400 | 0 | 1058.50 | 10.77 | 12.36 |

^a -1 = low. 0 = center. +1 = high.

Table 2. Radioactive cobalt experimental design for zeolite 5A

| Run number (N) | Factors (input values) | | | | Response | | |
|----------------|-----------------------------|---------------------------------------------|-----------------------------------|---------------------|-----------------------------|-----------------------------------|-----------------|
| | Amount of sorbent (g/100ml) | Initial activity of radioactive cobalt Bq/L | | Final activity Bq/L | Decontamination Factor (DF) | | |
| | | Sorbent Dosage (% w/v) | level ^a x ₁ | | A ₀ (Bq/L) | level ^a x ₂ | Experimental DF |
| 1 | 0.15 | 1 | 15200 | 1 | 1000.55 | 15.19 | 14.96 |
| 2 | 0.05 | -1 | 15200 | 1 | 3188.35 | 4.77 | 5.67 |
| 3 | 0.15 | 1 | 7600 | -1 | 572.85 | 13.27 | 12.05 |
| 4 | 0.05 | -1 | 7600 | -1 | 1761.01 | 4.32 | 4.22 |
| 5 | 0.15 | 1 | 11400 | 0 | 997.81 | 11.42 | 12.87 |
| 6 | 0.05 | -1 | 11400 | 0 | 2227.51 | 5.12 | 4.31 |
| 7 | 0.1 | 0 | 15200 | 1 | 1463.86 | 10.38 | 9.71 |
| 8 | 0.1 | 0 | 7600 | -1 | 1221.93 | 6.22 | 7.53 |
| 9 | 0.1 | 0 | 11400 | 0 | 1401.32 | 8.14 | 7.99 |
| 10 | 0.1 | 0 | 11400 | 0 | 1341.07 | 8.50 | 7.99 |

^a -1 = low. 0 = center. +1 = high.

Table 3. Analysis of variance (ANOVA) for the response for zeolite 3A

| Source | Sum of Squares | df | Mean Square | F-value | p-value | |
|--------------------|----------------|----|-------------|---------|---------|-----------------|
| Model | 463.84 | 5 | 92.77 | 7.87 | 0.0339 | significant |
| A-Sorbent Dosage | 10.55 | 1 | 10.55 | 0.8950 | 0.3977 | |
| B-Initial Activity | 11.30 | 1 | 11.30 | 0.9581 | 0.3831 | |
| AB | 46.38 | 1 | 46.38 | 3.93 | 0.1184 | |
| A ² | 106.02 | 1 | 106.02 | 8.99 | 0.0400 | |
| B ² | 5.35 | 1 | 5.35 | 0.4538 | 0.5375 | |
| Residual | 47.16 | 4 | 11.79 | | | |
| Lack of Fit | 45.00 | 3 | 15.00 | 6.93 | 0.2706 | not significant |
| Pure Error | 2.16 | 1 | 2.16 | | | |

The model F-value of 7.87 implies the model is significant for zeolite 3A. There is just a 3.39% chance that an F-value this large could occur due to noise. The lack of fit F-value of 6.93 implies the lack of fit is not significant relative to the pure error. There is a 27.06% chance that a lack of fit F-value this large could occur due to noise (Table 3).

Table 4. Analysis of variance (ANOVA) for the response for zeolite 5A

| Source | Sum of Squares | df | Mean Square | F-value | p-value | |
|--------------------|----------------|----|-------------|---------|---------|-----------------|
| Model | 119.59 | 5 | 23.92 | 12.63 | 0.0146 | significant |
| A-Sorbent Dosage | 0.0649 | 1 | 0.0649 | 0.0343 | 0.8621 | |
| B-Initial Activity | 0.7076 | 1 | 0.7076 | 0.3737 | 0.5740 | |
| AB | 0.5402 | 1 | 0.5402 | 0.2853 | 0.6215 | |
| A ² | 0.8400 | 1 | 0.8400 | 0.4436 | 0.5418 | |
| B ² | 0.9261 | 1 | 0.9261 | 0.4891 | 0.5229 | |
| Residual | 7.57 | 4 | 1.89 | | | |
| Lack of Fit | 7.51 | 3 | 2.50 | 38.63 | 0.1176 | not significant |
| Pure Error | 0.0648 | 1 | 0.0648 | | | |

The model F-value of 12.63 indicates the model is significant for zeolite 5A. There is just a 1.46% chance that an F-value this large could occur due to noise. The lack of fit F-value of 38.63 implicates the lack of fit is not significant relative to the pure error. There is an 11.76% chance that a lack of fit F-value this large could occur due to noise (Table 4).

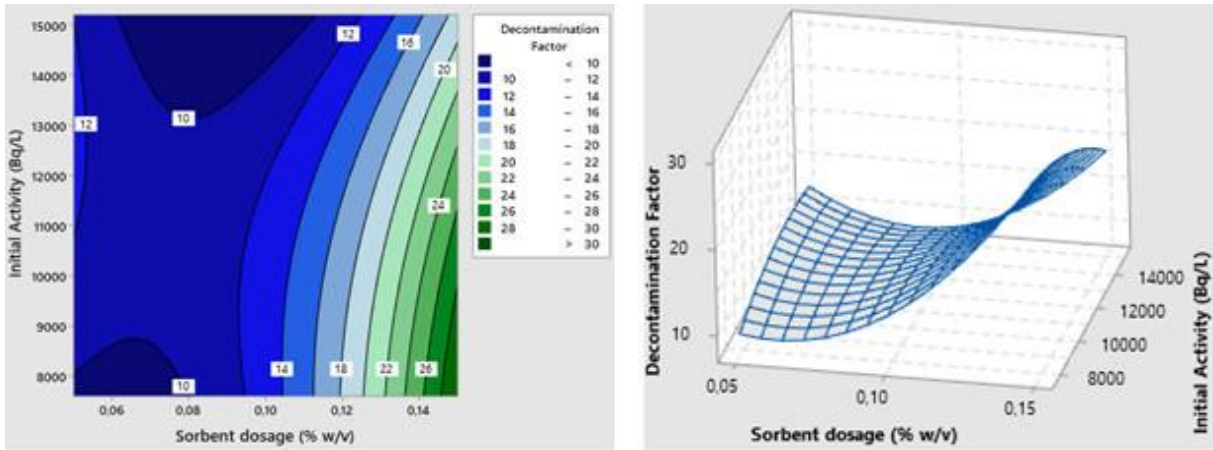


Figure 1. Zeolit 3A - radioactive cobalt contour plot and response surface plot for decontamination factor.

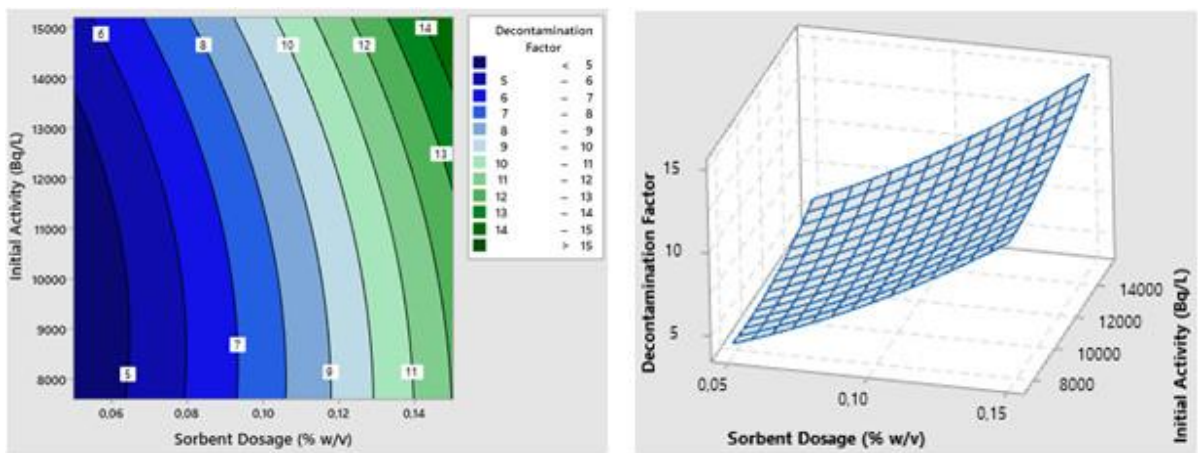


Figure 2. Zeolit 5A - radioactive cobalt contour plot and response surface plot for decontamination factor.

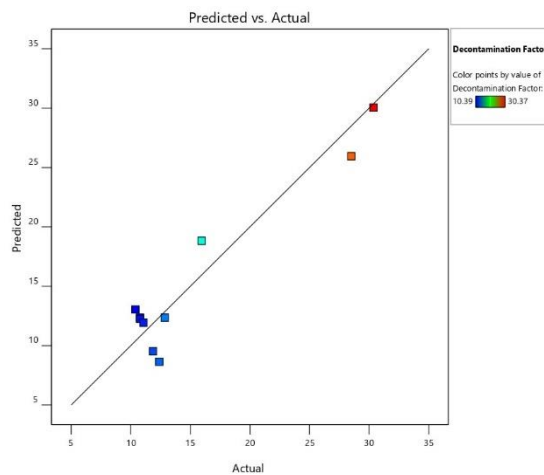


Figure 3. Predicted and actual decontamination factor for zeolite 3A

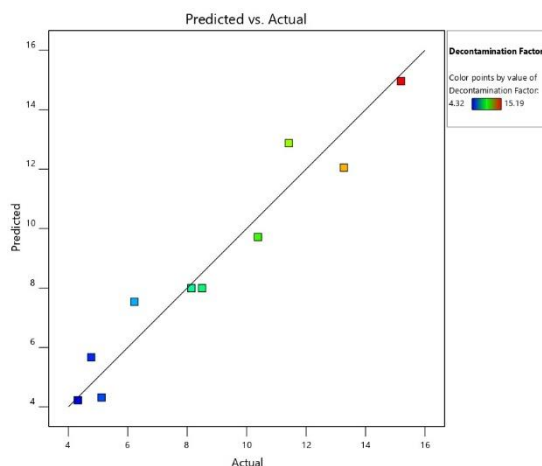


Figure 4. Predicted and actual decontamination factor for zeolite 5A

Figure 1 shows the radioactive cobalt contour plot and response surface plot for the decontamination factor for zeolite 3A. The counter map demonstrates the decontamination factor interconnection amid the sorbent quantity and initial radioactive cobalt concentration. The increasing of sorbent dosage will give enhanced values of the decontamination factor. However, increasing of initial cobalt concentration will diminish the decontamination factor for zeolite 3A. Decontamination factor is growing for higher sorbent dosage (i.e. $SD > 0.1\%$ w/v). The interaction among the input variables and response (decontamination factor) can be seen in the three-dimensional response surface plot for radioactive cobalt adsorption in figure 1.

The optimal condition was observed to be $A_0 = 7600$ Bq/L and $SD = 0.15$ w/v for radioactive cobalt adsorption on zeolite 3A. The maximum computed decontamination factor was 30.05 in optimal condition. The experimental corresponding value (decontamination factor) was 30.37 for the same condition (Table 1).

Figure 2 shows the radioactive cobalt contour plot and response surface plot for the decontamination factor for zeolite 5A. The decontamination factor is increasing while sorbent dosage is rising. The decontamination factor is growing while the initial cobalt concentration is increasing for higher sorbent dosage (i.e. $SD > 0.1\%$ w/v).

For the 5A sorbent, the optimal condition was determined to be $A_0 = 15200$ Bq/L and $SD = 0.15$ w/v. The maximum decontamination factor was predicted as 14.96 in this condition. The experimental decontamination factor corresponds to 15.19 for the same condition (Table 2).

The experimental minimum decontamination factor was 10.39 for 0.1 % w/v sorbent dosage and 7600 Bq/L initial cobalt activity for zeolite 3A. The predicted value in the same input variables corresponds to 13.05. In the case of zeolite 5A, the experimental minimum decontamination factor was 4.32 for 0.05 g sorbent dosage and 7600 Bq/L initial cobalt activity.

Figures 3 and 4 demonstrate the predicted and actual decontamination factor for zeolite 3A and 5A, respectively. As seen from the figures predicted values are very close to actual values (Figures 3 and 4).

The calculated value is 4.22 (predicted) in the same condition for zeolite 5A. The sorbent dosage has a certain effect on response (decontamination factor) for both sorbents (zeolite 3A and zeolite 5A). According to our experimental result, decontamination capacity of zeolite 3A is higher than the zeolite 5A.

Due to their high radiation stability and extreme selectivity, zeolites are useful for the removal of radioisotopes from nuclear wastewater. The more the adsorbent dosage will result the better the

decontamination factor. Determination of the optimum quantity of adsorbent is a key parameter and affects the whole adsorption process [2, 3]. The main adsorption mechanism is that cations in the zeolite structure can exchange with cations in the radioactive solutions freely through the cavities [14]. RSM is an effective mathematical tool to assess the effect of the input variables and their interactions on the dependent variable (response) [12].

IV. CONCLUSIONS

In this study, we investigated the ability of removal of the radioactive cobalt by zeolite 3A and 5A in aqua solution. The maximum experimental decontamination factor was 30.37 and 15.9 for zeolite 3A and zeolite 5A, respectively. Zeolite 3A demonstrates stronger adsorption than zeolite 5A because the network-like structure contains small pores. RSM was used to describe input variables (Sorbent dosage and initial activity) relationship with the response (Decontamination factor). Our results denoted that molecular sieves lead to a large amount of removal of radioactive cobalt ions from aqua solution. We presented that both zeolite 3A and 5A might be used as an adsorbent for the removal of radioactive cobalt in radioactive wastewater. It was found that zeolite showed much better performance for cobalt removal from aqua solution as compared to other adsorbents such as pumice. Response surface methodology is high accuracy and great prediction performance.

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