Removal of Fast Green FCF dye from aqueous solutions using Flower Gel as a low-cost adsorbent
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ABSTRACT
The purpose of this study was to investigate the removal of Fast Green FCF dye from aqueous solutions using Flower Gel in a batch adsorption process. The effect of different parameters such as pH, contact time, adsorbent dosage, stirrer speed and temperature were studied, and various isotherms including Langmuir, Freundlich and Tempkin were applied. The adsorbent characteristics were determined by microscopic analysis, Fourier transform infrared spectroscopy (FTIR), X-ray diffraction and ultraviolet-visible (UV-vis) spectrophotometry. The results showed that the equilibrium experimental data fitted well to the Langmuir isotherm and the maximum adsorption capacity for this adsorbent was 58.82 mg/g. The adsorption kinetic data followed the pseudo-second-order kinetic model and the thermodynamic parameters of the adsorption, such as Gibbs free energy ($\Delta G^\circ$), enthalpy ($\Delta H^\circ$) and entropy ($\Delta S^\circ$), showed that the Fast Green FCF adsorption process by Flower Gel was spontaneous and exothermic in nature.

Key words | adsorption, dye effluents, Fast Green FCF, Flower Gel, thermodynamic parameters

INTRODUCTION
Nowadays population growth and industrialization has led to an extensive utilization of colors in various industrial processes, especially in foodstuff all around the world (Banerjee et al. 2016). Dyes are conventionally used in the plastics, leather, textile and food industries, and direct discharge of these dyes to the water creates hazardous wastewaters (Anastopoulos & Kyzas 2014). These effluents can enter human bodies and cause serious illnesses such as cancer and genetic effects (Kundu et al. 2017). Several reports have also shown that with a dye concentration of less than 1 ppm, water bodies can be colored and prevent sunlight penetration (Ogugbe & Sawidis 2017; Geetha et al. 2015). Therefore, removal of these pollutants from aqueous solutions must be done before releasing into rivers (Cengiz & Cavas 2008).

Generally, dyes have been classified as anionic, cationic and non-ionic (Banerjee & Sharma 2015; Mahmoud et al. 2016). Fast Green FCF is an anionic dye which has been generally utilized in coloring foodstuff and is also called as Food Green 3, Green 1724 and Solid Green FCF (Mittal et al. 2009). The specified application of this dye is in histone staining at alkaline pH after acid extraction of DNA, protein staining in electrophoresis and coloring of tinned green peas, fishes, vegetables and desserts at maximum levels of 100 mg/kg (Mittal et al. 2008). It is a highly toxic and allergenic dye that causes eye and skin irritation as well as affecting the respiratory system (Gong et al. 2005; Mittal et al. 2009; Ramalakshmi et al. 2012). Although some developed countries have prohibited its use, Fast Green FCF is one of the most common food dyes which has been used until now. The maximum absorption of this dye is in the range of 622 to 626 nm and its structure is given in Figure 1 (Mittal et al. 2009). Various methods such as chemical precipitation (Seow & Lim 2016), biodegradation (Lin et al. 2016), solvent extraction (Liu et al. 2016), ion exchange (Ladeira & Gonçalves 2007), membrane separation (Liu et al. 2017) and adsorption (Morcali et al. 2014) have been used to remove dye from wastewaters. Among these methods, adsorption is the most efficient method due to its low cost, high efficiency, simple operational features and the ability to remove a wide range of dyes (Abdi et al. 2017).

In previous studies, it has been shown that modified vermiculite (Stawiński et al. 2016, 2017a, 2017b), hydrotalcite-derived mixed oxides (Stawiński et al. 2017c), mango leaf (Uddin et al. 2017), mahogany fruit shell (Sartape et al. 2017),...
and sumac leaves (Dülger et al. 2013) can be used as a low-cost adsorbent for the removal of dye from aqueous solutions. Adsorbents such as de-oiled soya (Mittal et al. 2009), bottom ash (Mittal et al. 2009), peanut hull (Gong et al. 2005), active carbon provided from Gloriosa superba effluent and Alternaria raphani fungal biomass (Ramalakshmi et al. 2012), modified montmorillonite with Crystal Violet (Rytwo et al. 2006), and coffee husk (Ahalya et al. 2014) have been successfully used for the removal of Fast Green FCF from aqueous solutions.

Flower Gel or crystal soil is a biodegradable polymer which does not create any pollution in the environment and is known as the best alternative for replacing garden soil in the modern world.

Flower Gel is usually composed of a hydrophilic polymer such as polyacrylamide. When these polymers soak in water, they adsorb the moisture quickly so that their volume increases 40 to 80 times their initial volume and the average diameter of each ball becomes 1 cm approximately. This can be attributed to the hydrophilic property of this polymer. The most important features of this adsorbent are its cheap price and high durability in water.

According to our knowledge and literature review, there is no study on the removal of dyes using Flower Gel. The objective of this work was to evaluate the removal of Fast Green FCF dye by batch adsorption using Flower Gel as a new low-cost adsorbent, and the effect of parameters such as contact time, pH, adsorbent dosage, stirrer speed and temperature on the adsorption process were investigated. To understand the adsorption mechanism, the equilibrium, kinetic and thermodynamic parameters were studied and the proper models were selected.

**METHODS**

**Materials and methods**

Fast Green FCF with the molecular mass of 808.85 g/mole and molecular formula C$_{37}$H$_{34}$N$_{2}$O$_{10}$S$_{3}$Na$_{2}$ was purchased from Merck (Darmstadt, Germany). Distilled water was used for preparing solutions. This dye was employed without further purification. In this study, adsorption of Fast Green FCF dye was conducted using Flower Gel made in China as the adsorbent.

The adsorption tests were performed in a batch system and concentration of the dye solutions was determined by a UV-Visible spectrophotometer (S2100S model) with a wavelength of 625 nm. The pH values were measured with a pH meter Behineh SAT· 2002. Microscopic analysis was performed by Nikon ECLIPSE E100 microscope and infrared spectrum was recorded with a Shimadzu (8400s) Fourier transform infrared (FTIR) spectrophotometer. The X-ray diffraction (XRD) pattern was conducted using D4Bruker Cu-K$_{α}$ ($λ = 0.15406$ nm). The filter paper used in this study was Whatman No. 42 (125 mm).

**Adsorption studies**

The stock solution of the Fast Green FCF dye was prepared with a concentration of 500 mg/L. After drawing the calibration curve at the wavelength of 625 nm (Figure S1, Supplementary Material, available with the online version of this paper), the amount of adsorbed mass was calculated. In this study, some polymers were soaked in 400 mL of water and left for 4 hours to get the Flower Gel. Finally, the swollen gel that was saturated with water was utilized for the adsorption tests. The effects of pH, contact time, adsorbent dosage, speed of the stirrer and temperature were measured by keeping the other parameters constant.

The removal percentage of the dye from solution and the amount of adsorbed mass were calculated using Equations
\[ \% \text{Removal} = \frac{C_i - C_f}{C_i} \times 100 \]  
\[ q = \frac{C_i - C_f}{M} \times V \]

where \( C_i \) and \( C_f \) are the initial and final concentrations of dye (mg/L) in the solution, respectively; \( M \) is the adsorbent mass in g and \( V \) is the volume of the solution in L. (Karapinar & Donat 2009).

**Adsorption isotherms**

To investigate the adsorption isotherms, 100 mL of the dye solution was made with the concentrations of 25, 40, 55, 70, 85 and 100 mg/L at room temperature and the amount of adsorbed mass was calculated using Equation (2) at pH 3 using 0.04 g of adsorbent and stirrer speed of 480 rpm after 20 min.

**Adsorption kinetics**

In order to determine the kinetics of adsorption, 100 mL of the dye solution with initial concentration 25 mg/L, pH = 3 and containing 0.04 g adsorbent was prepared. Then this solution was shaken at 480 rpm at various contact times (1–30 min). The amount of adsorbed mass was calculated using Equation (2).

**Adsorption thermodynamic**

The thermodynamic parameters of adsorption were estimated using 100 mL of the dye solution with initial concentration 25 mg/L, pH = 3, contact time 20 min and 0.04 g adsorbent at temperatures of 298, 315, 323, 335 and 345 K. The amount of adsorbed mass was calculated with Equation (2).

**RESULTS AND DISCUSSION**

**Characterization of Flower Gel**

**Microscopic analysis**

Due to the jelly structure of the adsorbent, SEM and TEM analysis were limited, so microscopic images with resolution 1x and 10x were taken from the adsorbent before and after the adsorption process. Images are shown in Figure 2(a)–(c). As can be seen in these pictures, after soaking polymer granules in water, spherical swollen gels were formed of the same size. In other words, the polymer granules were saturated with water and this water dissolves Fast Green FCF dye. Therefore dye can go inside the adsorbent structure while the adsorbent stays in the water. It is clear why this adsorbent was useful in the separation of dye from aqueous solutions. Floral Gel is a biodegradable polymer, and when out of water it gradually shrinks.

**FTIR analysis**

The chemical structure of Flower Gel was studied using FTIR spectroscopy within 500–4,000 cm\(^{-1}\) and the results are shown in Figure 3. Bands near 1,108.99 cm\(^{-1}\) were related to the C-O stretching vibration and the aromatic bonds C=C peaked at around 1,402.15 cm\(^{-1}\), 1,556.45 cm\(^{-1}\) and 1,614.31 cm\(^{-1}\). Peaks at 1,666.38 cm\(^{-1}\), 2,948.96 cm\(^{-1}\) and 3,442.70 cm\(^{-1}\) correspond to the carbonyl groups stretching vibrations, C-H stretching vibration, and O-H and N-H stretching vibrations respectively. According to the FTIR spectrum, the presence of the
functional groups, such as hydroxyl and amine, significantly affects the adsorption of Fast Green FCF.

**XRD analysis**

The XRD pattern of Flower Gel is shown in Figure 4. The pattern had no sharp reflection, it only gave a broad reflection at 2θ about 20–40° which can be related to the fully amorphous structure of this adsorbent. It was expected for Flower Gel as a polymeric material.

**UV-visible analysis**

The UV-visible spectrum of the Flower Gel particles had one absorption peak at ∼200 nm and shoulder peaks at ∼236–316 nm. The first corresponded to the π → π* transition of C = C bonds and shoulder peaks were attributed to the n → π* transition of C = O bonds (Figure S2, Supplementary Material, available with the online version of this paper).

**Effect of contact time**

In order to investigate the effect of contact time, 0.02 g of Flower Gel was mixed with 100 ml of Fast Green FCF dye solution with a concentration of 25 mg/L at pH 3 and room temperature. Figure 5 shows the effect of contact time on the removal percentage of Fast Green FCF. As can be seen in this Figure, the removal percentage was increased by increasing contact time and equilibrated after 20 min. The adsorbent was saturated with the dye molecules in that time and hence the removal percentage became almost constant. The ability of this adsorbent for Fast Green FCF adsorption is very fast in comparison with other adsorbents (Gong et al. 2005; Ramalakshmi et al. 2012; Ahalya et al. 2014).

**Effect of pH**

Since pH plays an important role in adsorption processes, first the pHzpc point was determined according to previous works (Cerović et al. 2007; Fiol & Villaescusa 2009; Malekbala et al. 2012). At this pH, the net charge on the surface is zero, in other words, the number of negative and positive charges on the adsorbent are equal. For this purpose, the initial pH of the solutions containing 0.01 M NaCl was adjusted to 3–10 and after addition of 0.15 g of the adsorbent, the suspensions were shaken for 24 hours at 200 rpm. The pH of the solutions was controlled by adding the necessary amounts of hydrochloric acid and diluted sodium hydroxide. The final pH was measured after withdrawing the adsorbent using a filter paper.

Figure 3 | FTIR spectrum of Flower Gel.

Figure 4 | XRD pattern of Flower Gel.

Figure 5 | Effect of contact time on the removal percentage of Fast Green FCF dye (m = 0.02 g adsorbent, pH = 3, 100 ml of the dye solution with the initial concentration of 25 mg/L, T = room temperature).
point of zero charge (PZC) point was determined from the intersection of pHf versus pHi and pHi = pHf curves. At pH < pH_{pzc} acidic conditions are dominant and the surface charge of the adsorbent is positive. Conversely, the charge of the adsorbent is negative due to the presence of OH⁻ species at pH > pH_{pzc}. As can be seen in Figure 6(a), the pH_{pzc} obtained was 6.2. In order to study the effect of pH on the removal percentage of the dye, 0.02 g of adsorbent was mixed with 100 mL of the solution with the concentration of 25 mg/L at different initial pH (from 3 to 9) at room temperature for 20 min. Then the mixture was filtered using a filter paper. The amount of the dye in the filtrate was determined using the spectrophotometer. The results of the initial pH effect on the removal percentage of the dye are shown in Figure 6(b). As can be seen, the removal percentage of the dye was significantly reduced by increasing the initial pH from 3 to 9 because the density of positive charges was increased on the surface of the adsorbent at acidic pH and the removal percentage of Fast Green FCF dye as an anionic dye was increased. These results were in good agreement with Figure 6(a) where the optimum pH for the adsorption of this anionic dye was less than pH_{pzc}. At high pH, there was a competition between OH⁻ ions and the dye for adsorption on the adsorbent and the removal percentage was decreased. A similar trend was reported for the adsorption of Fast Green FCF on de-oiled soya and bottom ash (Mittal et al. 2009) and coffee husk (Ahalya et al. 2014).

**Effect of adsorbent dosage**

The effect of adsorbent dosage on the removal efficiency and the amount of adsorbed mass of the dye is shown in Figure 7. For this purpose, the experiments were done using different amounts of the adsorbent in the solution with pH = 3 for 20 min. By increasing the adsorbent dosage, from 0.01 to 0.04 g, the dye removal efficiency increased because the number of available adsorption sites was increased. In doses higher than 0.04 g, the solution came to equilibrium and the removal efficiency remained almost constant. As the maximum removal of the dye occurred using 0.04 g of adsorbent, the optimum adsorbent amount was selected as 0.04 g.

**Effect of stirrer speed**

The effect of the stirrer speed on the removal percentage of the dye is shown in Figure 8. The adsorption tests were performed at the optimum contact time, pH and adsorbent dosage. The removal percentage of the dye was increased by increasing the stirrer speed from 240 to 480 rpm, but then the removal percentage was decreased by increasing the stirrer speed to 720 rpm. The reason for this phenomenon can be attributed to desorption of the dye from the adsorbent. In other words, the high speed of the agitator caused the dye to come out of the adsorbent pores.

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**Figure 6**  | (a) Determination of pH_{pzc}, (b) effect of pH on the removal percentage of the dye (m = 0.02 g adsorbent, t = 20 min, 100 mL of the dye solution with the initial concentration of 25 mg/L, T = room temperature).

**Figure 7**  | Effect of adsorbent dosage on the removal percentage and the amount of adsorbed mass of the dye (pH = 3, t = 20 min, 100 mL of the dye solution with the initial concentration of 25 mg/L, T = room temperature).
Effect of initial Fast Green FCF concentration

The effect of Fast Green FCF concentration was investigated using six different concentrations. Their values were varied from 25 to 100 mg/L at a constant mass of adsorbent 0.04 g, pH 3 and contact time 20 min. The removal of Fast Green FCF decreased as the dye concentration increased, which may be due to the saturation of adsorbent. Also, the amount of adsorbed mass of Fast Green FCF increased because the concentration driving force had been increased (Figure S3, Supplementary Material, available online). The maximum adsorption capacity for this adsorbent obtained was 58.82 mg/g.

Effect of temperature

To examine the effect of temperature, tests were performed under similar conditions to the previous tests at temperatures of 298, 315, 323, 335 and 345 K. The removal percentage of the dye was reduced with increasing the temperature from 298 to 345 K, indicating that the dye adsorption is an exothermic process on Flower Gel (Figure S4, Supplementary Material, available online).

Adsorption isotherms

In this study, Langmuir, Freundlich and Tempkin isotherms were examined and the results are shown in Table 1.

Langmuir isotherm

The linear form of the Langmuir isotherm is given in Equation (3):

$$\frac{C_e}{q_e} = \frac{1}{bQ_0} + \frac{C_e}{Q_0}$$

where $q_e$ is the amount of material adsorbed by the adsorbent (mg/g), $C_e$ is the ion concentration at equilibrium (mg/L), $Q_0$ is the maximum adsorption capacity (mg/g) and $b$ is the Langmuir constant (L/mg) (Ho et al. 2002). A straight line is obtained by drawing $C_e/q_e$ versus $C_e$ of which the slope and y-intercept are $(1/Q_0)$ and $(1/bQ_0)$, respectively (Figure S5, Supplementary Material, available online). The maximum adsorption capacity for this adsorbent obtained was 58.82 mg/g.

Freundlich isotherm

The linear form of this isotherm is given in Equation (4):

$$\log q_e = \log K_f + \frac{1}{n} \log C_e$$

where $q_e$ is the amount of material adsorbed by the adsorbent (mg/g), $C_e$ is ion concentration at equilibrium (mg/L), $K_f$ is the Freundlich constant and $n$ is the intensity of the adsorption (Copello et al. 2012). A straight line is obtained by drawing $\log q_e$ versus $\log C_e$ of which the slope and y-intercept are $(1/n)$ and $\log K_f$, respectively.

Tempkin isotherm

The linear form of the Tempkin isotherm is given in Equation (5):

$$q_e = \frac{RT}{b_T} \ln A_T + \left(\frac{RT}{b_T}\right) \ln C_e$$

where $q_e$ is the amount of material adsorbed by the adsorbent (mg/g), $C_e$ is the ion concentration at equilibrium (mg/L), $R$ is the universal gas constant (8.314 J/mol.K), $T$ is the temperature (K), $A_T$ is the binding constant associated with the maximum bond energy (L/g) and $b_T$ is the Tempkin constant (Foo & Hameed 2010). A straight line is obtained by drawing $q_e$ versus $\ln C_e$ and $b_T$ and $A_T$ are the slope and y-intercept, respectively. The constants and correlation coefficients related to each isotherm are given in Table 1. The correlation coefficient can be a proper criterion for selecting the most appropriate isotherm. According to Table 1, Langmuir isotherm with a correlation coefficient of 0.977 was consistent with the experimental data. This means that the adsorption involved the formation of a monolayer on a homogeneous surface. In addition, in order to validate this study, the maximum adsorption capacity of Flower Gel was compared with other work in
Table 2. As can be seen in this table, the maximum adsorption capacity of Flower Gel for this dye is comparable to other adsorbents.

Adsorption isotherm  | Model parameters  
---|---
Langmuir  | $Q_0$ (mg/g) 58.820, $b$ (L/mg) 0.085, $R^2$ 0.977  
Freundlich  | $n$ 4.386, $K_f$ (mg/g) (L/mg)$^{1/n}$ 18.980, $R^2$ 0.951  
Tempkin  | $b_T$ 257.170, $A_T$ (L/g) 2.536, $R^2$ 0.918

### Adsorption kinetics

In this study, the experimental data were fitted with pseudo-first-order and pseudo-second-order kinetic models and the results are shown in Table 3.

**Pseudo-first-order kinetic model**

The non-linear form of this equation is as follows:

$$q_t = q_e (1 - e^{-k_1 t})$$  \hspace{1cm} (6)

In this equation, $q_e$ and $q_t$ (mg/g) are the amount of material adsorbed by the adsorbent at equilibrium time and at time $t$, respectively. Moreover, $k_1$ is the constant of the pseudo-first-order kinetic model (Cui et al. 2012). The calculated values and constants of this model are presented in Table 3.

**Pseudo-second-order kinetic model**

The pseudo-second-order model is presented as follows:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e}$$  \hspace{1cm} (7)

In this equation, $q_e$ and $q_t$ (mg/g) are the amount of material adsorbed by the adsorbent at equilibrium time and time $t$, respectively. $K_2$ is the constant of pseudo-second-order model for the adsorption reaction (Visa & Duta 2013) that is obtained from the linear plot ($t/q_t$) versus $t$ (Figure S6, Supplementary Material, available online). As can be seen in Table 3, the pseudo-second-order kinetic model ($R^2 = 0.997$) was consistent with the experimental data.

### Adsorption thermodynamics of Fast Green FCF dye

The thermodynamic behavior of Fast Green FCF adsorption on Flower Gel was estimated using the following equations:

$$\Delta G^o = -RT \ln K_c$$  \hspace{1cm} (8)

$$\Delta G^o = \Delta H^o - T\Delta S^o$$  \hspace{1cm} (9)

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Table 3. Constant values and correlation coefficients for pseudo-first and second-order kinetic models

| Kinetic model          | Model parameters  
---|---
Pseudo-first-order  | $K_1$ (1/min) 1.746, $q_e$ (mg/g) 57.935, $R^2$ 0.716  
Pseudo-second-order  | $K_2$ (g/(mg·min)) 0.015, $q_e$ (mg/g) 66.225, $R^2$ 0.997

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Table 1. Constant values and correlation coefficients for Langmuir, Freundlich and Tempkin adsorption isotherms

| Adsorption isotherm | Model parameters  
---|---
Langmuir  | $Q_0$ (mg/g) 58.820, $b$ (L/mg) 0.085, $R^2$ 0.977  
Freundlich  | $n$ 4.386, $K_f$ (mg/g) (L/mg)$^{1/n}$ 18.980, $R^2$ 0.951  
Tempkin  | $b_T$ 257.170, $A_T$ (L/g) 2.536, $R^2$ 0.918

Table 2. Comparison of maximum adsorption capacity of different adsorbents for removal of Fast Green FCF dye

| Adsorbent                      | Maximum adsorption capacity (mg/g) | Reference  
---|---|---
Peanut hull  | 15.60  | Gong et al. (2005)  
Active carbon from *Alternaria raphani* fungal biomass  | 35.59  | Ramalakshmi et al. (2012)  
Active carbon from *Gloriosa superba* effluent  | 51.02  | Ramalakshmi et al. (2012)  
Modified montmorillonite with Crystal Violet  | 80.88  | Rytwo et al. (2006)  
Coffee husk  | 96.6  | Ahalya et al. (2014)  
Bengal gram husk  | 56.06  | Kanamadi et al. (2006)  
Tamarind shell  | 101.58  | Kanamadi et al. (2006)  
Flower Gel  | 58.82  | This work
where $K_c$ is the solute distribution coefficient between the adsorbent and solution ($q_C/C_0$) and $R$ is the universal gas constant. $T$, $ΔH^o$, $ΔS^o$ and $ΔG^o$ are the temperature in kelvin, enthalpy changes, entropy changes and Gibbs free energy changes, respectively. Equations (8) and (9) can also be written as Equation (10):

$$\ln K_c = \frac{ΔH^o}{RT} + \frac{ΔS^o}{R}$$

where enthalpy and entropy are obtained by drawing $\ln K_c$ versus $1/T$ (Khani et al. 2008). The results of these calculations are shown in Table 4. The negative values of $ΔH^o$ showed an exothermic adsorption process, the negative values of $ΔG^o$ indicate a spontaneous adsorption process and the negative values of $ΔS^o$ represent a decrease in the irregularity at the solid-liquid interface during the adsorption process (Wang et al. 2009).

### CONCLUSION

In this study, Fast Green FCF dye was adsorbed using Flower Gel, and effect of parameters such as pH, contact time, adsorbent dosage, stirrer speed and temperature were investigated. The structure of the adsorbent was investigated by microscopic analysis, FTIR, XRD and UV-visible spectrophotometry. The results showed that the maximum adsorption amount was obtained at pH 3 and the optimum amount of adsorbent and contact time were 0.04 g and 20 min, respectively. The equilibrium data of the adsorption followed the Langmuir isotherm ($R^2 = 0.977$) and the maximum adsorption capacity calculated for this adsorbent was 58.82 mg/g. The kinetic studies indicated that equilibrium data followed the pseudo-second-order kinetic model, and the thermodynamic studies on the adsorption of Fast Green FCF showed an exothermic and spontaneous adsorption process. This work shows that Flower Gel can be used for efficient and economic removal of Fast Green FCF dye from aqueous solutions.

### ACKNOWLEDGEMENTS

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### REFERENCES


### Table 4 | The thermodynamic parameters of Fast Green FCF adsorption on the Flower Gel

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<th>$T(K)$</th>
<th>$ΔG^o$ (kJ mol$^{-1}$)</th>
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