Preparation, Characterization and Application of Magnetic Fe₃O₄-CS for the Adsorption of Orange I from Aqueous Solutions

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Abstract
Fe₃O₄ (Fe₃O₄-CS) coated with magnetic chitosan was prepared as an adsorbent for the removal of Orange I from aqueous solutions and characterized by FTIR, XRD, SEM, TEM and TGA measurements. The effects of pH, initial concentration and contact time on the adsorption of Orange I from aqueous solutions were investigated. The decoloration rate was higher than 94% in the initial concentration range of 50–150 mg L⁻¹ at pH 2.0. The maximum adsorption amount was 183.2 mg g⁻¹ and was obtained at an initial concentration of 400 mg L⁻¹ at pH 2.0. The adsorption equilibrium was reached in 30 minutes, demonstrating that the obtained adsorbent has the potential for practical application. The equilibrium adsorption isotherm was analyzed by the Freundlich and Langmuir models, and the adsorption kinetics were analyzed by the pseudo-first-order and pseudo-second-order kinetic models. The higher linear correlation coefficients showed that the Langmuir model (R² = 0.9995) and pseudo-second-order model (R² = 0.9561) offered the better fits.

Introduction
Currently, dyes are widely used in many industrial applications, including textiles, printing, plastics, food, leather and papermaking, among others [1–4]. In China, large amounts of dye wastewater are directly discharged into natural water courses without treatment, particularly in the rural area of China [5]. Wastewater containing dyes and dyed products has caused pollution in many areas [6–10]. Due to the complex structure of their aromatic molecules, most azo dyes biodegrade slowly if at all, which causes a natural unbalance in the environment [11–13]. Some dyes containing special compounds are considered toxic to both human and animals even at very low concentrations, generally most of these compounds cause mutagenic, teratogenic and carcinogenic effects which subsequently lead to the generation of health disorders such as dysfunction of the kidney, reproductive system, liver, brain, and central nervous system [14]. The improper management of industrial water containing dyes is a source of pollution.

Orange I belongs to the family of azo dyes, which represent around 50% of all dyes used in textile industry [15–16]. Because it contains an -N=N- chromophore group, Orange I is highly toxic and causes various diseases [17–18], such as nausea, carcinogen, dermatitis, methemoglobinemia, tumors and allergies [19–20]. Being anionic in nature, orange I has high potential to leach into the soil profile and to contaminate ground water [21]. Wastewater containing Orange I can seriously harm human health if discharged into freshwater. The orange coloration is also a type of environmental pollution [1]. Environmental restrictions have been established by many local governments to control the quality of colored effluents and force industries to decolorize their effluents before discharging them [22]. Therefore, it is necessary to remove Orange I from wastewater before it is discharged into bodies of freshwater.

Several methods of dye removal have been developed during the global environmental movement, including photocatalysis [23], coagulation [24], and adsorption [25]. Among the processes for treating colored wastewater, adsorption is the best choice [26–27], because it is a low cost and easy to implement method [28]. Normal adsorbents such as active carbon and alumina have been used to adsorb dyes in wastewater, but their adsorptive capacity is not as high as expected [1], and they are expensive. It is therefore necessary to find a cheaper and more effective adsorbent substitute for the normal adsorbents.

Chitosan (CS) is a natural biopolymer obtained from the process of alkaline deacetylation of chitin [29]. It is hydrophilic, biocompatible, biodegradable and antibacterial. Chitosan is an ideal adsorbent because of its functional groups. Each glucosamine unit has one amine group (–NH₂) and two hydroxyl groups (–OH) [30]. Under acidic conditions, the amine groups of chitosan become protonated, and the positive group (–NH₃⁺) can adsorb some negative ions through electrostatic interaction. The adsorption capacity of chitosan could also be strengthened by forming a hydrogen bond between the hydroxyl groups (–OH) and the adsorbed molecules. However, pure chitosan does not have optimal adsorption because it easily dissolves in acidic solution and has weak chemical resistance [31]. Blending chitosan with magnetic Fe₃O₄ can effectively avoid the chemical weakness of...
chitosan, the hydroxyl groups on the surface of Fe₃O₄ can interact with amine groups and hydroxyl groups of chitosan through hydrogen-bond interaction to keep chitosan stable under acidic condition. The obtained adsorbent is also easily prepared, inexpensive and has high adsorptive capacity. Compared with other magnetic adsorbents, the cost of the adsorbent prepared is about 1/2 of amino-functionalized silica-coated Fe₃O₄ [32] and 1/3 of amine-modified silica magnetite [33]. The magnetic adsorbent thus obtained was used to process wastewater containing the acid dye orange I.

Commonly, magnetic adsorbents are prepared by two-steps method [34–35], which has a complicated preparation process and low production. In this study, Fe₃O₄ (Fe₃O₄-CS) coated with magnetic chitosan was prepared by a one-step method. The resulting Fe₃O₄-CS was characterized by Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD). The morphology of Fe₃O₄-CS was examined by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The chitosan content was measured by thermal gravimetric analysis (TGA). Orange I was adsorbed from an aqueous solution at room temperature (25°C), and the adsorbent could be easily be separated from the Orange I solution by magnetism. The effects of pH, initial concentration, and contact time were investigated. The adsorption isotherm and adsorption kinetics were studied for a comprehensive understanding of the adsorption process.

**Methods**

2.1 Chemical and materials

Chitosan with a 95% degree of deacetylation, FeCl₃ (97%) and FeSO₄·7H₂O (99%) were purchased from Sinopharm Chemical Reagent Co. Ltd. Orange I (C₁₆H₁₁N₂NaO₄S, MW = 350.32) was purchased from Aladdin Chemistry Co. Ltd. A stock solution was prepared by dissolving 0.4 g Orange I in 500 mL of distilled water, which was diluted to approximate concentrations. Other reagents used in this study were all analytical grade, and all solutions were prepared by using deionized water.

2.2 Preparation of magnetic Fe₃O₄-CS

First, 2.17 g of FeCl₃ and 0.77 g of FeSO₄·7H₂O were dissolved in 50 mL of deionized water in a 250 mL flask. The mixture was vigorously stirred in a water bath at 313 K for 30 min. Then 0.35 g of chitosan was dissolved in 100 mL of 1% (v/v) acetic acid. The chitosan solution was added to the flask and vigorously stirred for 2.0 h. Then, 48 mL of NH₃·H₂O was added dropwise over 2.0 h, and the solution was vigorously stirred for another 1.0 h. Throughout the process, the temperature was maintained at 313 K, and the whole process was conducted under protection of N₂ gas. After the reaction, the product was filtered and washed with distilled water and ethanol 3 times. Then, the precipitate was dried in a vacuum oven at 333 K. The obtained product was Fe₃O₄-CS.

2.3 Characterization of Fe₃O₄-CS

FTIR spectra of Fe₃O₄-CS were recorded on a Bruker VECTOR-22 IR spectrometer. KBr and the sample (approximately 1% mass of KBr) were mixed together, and then the precipitate was dried in a vacuum oven at 333 K. The obtained product was Fe₃O₄-CS.

![Figure 1. FTIR spectra of CS and Fe₃O₄-CS.](doi:10.1371/journal.pone.0108647.g001)

![Figure 2. XRD patterns of Fe₃O₄-CS.](doi:10.1371/journal.pone.0108647.g002)
mixture was pestled and pressed into a tablet. The spectra were collected over the spectral range of 400–4000 cm$^{-1}$.

XRD patterns were recorded on a Rigaku D/max 2500 kV PC X-ray diffractometer operating at 40 kV. The scan angle $2\theta$ varied from 10° to 80° and the scan speed was 0.03° s$^{-1}$.

SEM images were taken on a Quanta 200 (Philips- FEI, Holland). The SEM images were taken by applying 10 kV voltage with various magnification times for the observation of the surface.

TEM was obtained with a JEM-2100F microscope using an accelerating voltage of 200 kV. The samples were lightly ground and then dispersed ultrasonically in ethanol. A drop of the suspension was evaporated on a ‘holey’ carbon film and pre-deposited on 200-mesh copper grids.

TGA were conducted on a Perkin–Elmer Diamond TG/DTA Instrument with a heating rate of 10°C min$^{-1}$ under a nitrogen flow at temperatures ranging from 25 to 800°C. During the TGA measurement, the ratio of chitosan content in the Fe$_3$O$_4$-CS was obtained.

### 2.4 Batch adsorption experiments

First, 0.05 g of Fe$_3$O$_4$-CS and 40 mL of Orange I aqueous solution were added into a 50 mL conical flask. After agitation for

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**Figure 3.** SEM images (a, b, c) and TEM image (d) of Fe$_3$O$_4$ and Fe$_3$O$_4$-CS. 
**Figure 4.** TGA curve of Fe$_3$O$_4$-CS. 
**Figure 5.** UV-vis spectra of Orange I solution before and after adsorption.
3 h at a rate of 225 rpm, the flask was placed on a magnet, and the Fe$_3$O$_4$-CS was aggregated on the bottom by magnetic force. One milliliter of supernatant was diluted in a 50 mL volumetric flask to obtain a measurable absorption. The concentration of the Orange I in the solution was immediately determined via UV-vis spectroscopy (Purkinje General, TU-1901) at an optimal wavelength of 476 nm, which corresponds to the maximum absorbance for Orange I. The adsorption ability was calculated using absorbance values measured before and after adsorption according to the following equation:

$$q_e = \frac{C_0 - C_e}{M} \times V$$  \hspace{1cm} (1)

Where $q_e$ is the amount of dye adsorbed by the adsorbent (mg g$^{-1}$), $C_0$ is the initial dye concentration (mg L$^{-1}$), $C_e$ is the dye

![Figure 6. Effect of pH on Orange I adsorption by Fe$_3$O$_4$-CS.](doi:10.1371/journal.pone.0108647.g006)

![Figure 7. Schematic illustration for adsorption of Orange I by Fe$_3$O$_4$-CS.](doi:10.1371/journal.pone.0108647.g007)

![Figure 8. Effect of initial concentration on Orange I adsorption by Fe$_3$O$_4$-CS.](doi:10.1371/journal.pone.0108647.g008)

A Highly Efficient Adsorbent
Results and Discussion

3.1 Characterization of the Fe₃O₄-CS adsorbent

The FTIR spectra of chitosan and Fe₃O₄-CS are shown in Fig. 1. As shown, the spectra of Fe₃O₄-CS are almost consistent with the spectra of chitosan. The adsorption at approximately 3440 cm⁻¹ reflects the overlapping of the stretching vibration of the O-H groups and N-H groups. The adsorption at 2927 cm⁻¹ and 2860 cm⁻¹ is ascribed to the C-H stretching vibration of the -CH₂ groups in chitosan. The adsorption at 1643 cm⁻¹ is attributed to the deformation vibration of primary amine, the one at 1580 cm⁻¹ is attributed to the N-H deformation vibration of -NH₂ groups, and the one at 1423 cm⁻¹ is attributed to the C-N stretching vibration. The FTIR spectra in Fig. 1 clearly demonstrate the existence of chitosan in the Fe₃O₄-CS.

The XRD pattern of Fe₃O₄-CS is shown in Fig. 2. There are characteristic peaks at 2θ = 30.1°, 35.4°, 43.1°, 53.4°, 56.9° and 62.5°, which correspond to the (220), (311), (400), (422), (511) and (440) crystal planes of Fe₃O₄ respectively; these peaks are consistent with the PDF card in the database (PDF No. 19-0629). This indicates the existence of Fe₃O₄, and the obtained adsorbent can be separated from aqueous solutions by magnets [36].

Pure Fe₃O₄ was prepared in order to make a comparison with Fe₃O₄-CS on the surface structure. The surface structure of the pure Fe₃O₄ and synthesized magnetic Fe₃O₄-CS is shown in SEM images of Fig. 3. In Fig. 3(a), the surface of pure Fe₃O₄ is rough and irregular. After being coated with chitosan, the surface of the resulting Fe₃O₄-CS becomes smooth, and the folding structure can be clearly observed in Fig. 3(b). In Fig. 3(c), the folds on the surface of the Fe₃O₄-CS adsorbent are more distinct. They are formed by the coating of organic chitosan on the surface of Fe₃O₄.

Fig. 4(d) is the TEM image of Fe₃O₄-CS. In Fig. 3(d), the nanoparticles, which have a spherical structure and uniform particle size, could be observed. The color of the center of the sphere is darker, which is ascribed to the existence of Fe₃O₄. In contrast, due to the coating of organic chitosan, the color of the edge of the sphere is lighter. Through the SEM and TEM measurements, the microtopography of Fe₃O₄-CS could be clearly observed, and the nanoparticle size is approximately 20 nm.

Fig. 4 shows the TGA curve of Fe₃O₄-CS in the temperature range of 303 K-1000 K at a heating rate of 10 K min⁻¹. The weight lost from the Fe₃O₄-CS adsorbent was divided into three different temperature ranges. A 6.5% loss in the first stage was ascribed to the loss of absorbed and bound water between 303 K (30°C) and 423 K (150°C). Approximately 20% of the weight loss in the temperature range occurred between 463 K (190°C) and 603 K (330°C) in the second stage, which was due to the degradation and deacetylation of chitosan. Approximately 1% of the weight loss was in the third stage, which was due to further degradation of chitosan at 653–773 K (380–500°C). The output of Fe₃O₄-CS was 1.40 g and the theoretical output of Fe₃O₄-CS was 1.60 g. According to the TGA analysis, water content in Fe₃O₄-CS was about 6.5%, then the pure Fe₃O₄-CS obtained was about 1.31 g, the productivity was about 81.8%, the high productivity make the adsorbent economical and practical.
3.2 Effect of pH
UV-vis spectra of Orange I solution (400 mg L\(^{-1}\), pH 2.0) before and after adsorption is presented in Fig. 5. The absorbance after adsorption decreases sharply compared with absorbance before adsorption, which indicates a much lower concentration after adsorption. The effect of pH on adsorption is shown in Fig. 6. The adsorption amount decreases with increasing pH in the pH range from 1.0 to 9.0. The mechanism of adsorption relies on the ionic interactions between amino groups (\(-\text{NH}_3^+\)) and sulfonate groups (\(-\text{SO}_3^2-\)) of Orange I (shown in Figure 7). At pH 2.0, the adsorption amount is 178 mg g\(^{-1}\), and it decreases to 5.6 mg g\(^{-1}\) at pH 9.0. At pH 1.0, the adsorption amount does not increase obviously compared with adsorption amount at pH 2.0. This can be attributed to the protonation of the \(-\text{NH}_2\) groups into \(-\text{NH}_3^+\) in the presence of H\(^+\) ions under acidic conditions. At pH 2.0, the \(-\text{NH}_2\) groups were already completely protonated into \(-\text{NH}_3^+\), and the electrostatic interaction between \(-\text{NH}_3^+\) and \(-\text{SO}_3^2-\) was strengthened. With increasing pH, the amount of \(-\text{NH}_3^+\) decreased, and when the pH passes the isoelectric point of chitosan, the \(-\text{NH}_3^+\) groups deprotonated to the form of \(-\text{NH}_2\) groups, and the electrostatic interaction disappeared. Therefore, the optimal pH of adsorption is 2.0.

3.3 Effect of initial concentration
The effect of the dye concentration on the adsorption amount is shown in Fig. 8. It is clear that the adsorption of Fe\(_3\)O\(_4\)-CS increases with the initial concentration of Orange I in concentrations between 50 and 400 mg L\(^{-1}\). The maximum adsorption amount appears at a concentration of 400 mg L\(^{-1}\). Between 400 and 800 mg L\(^{-1}\), the adsorption amount remains constant at approximately 180 mg g\(^{-1}\), and no increase is observed.

The decoloration rate is an important parameter in the practical process of wastewater treatment. The decoloration rate of Orange I by Fe\(_3\)O\(_4\)-CS is shown in Fig. 9. It is clear that the decoloration rate decreases with increasing initial concentrations. When the concentration is in the range of 50–150 mg L\(^{-1}\), more than 94% of Orange I is removed. The optical concentration range for the decoloration of Orange I is 50–150 mg L\(^{-1}\). At this concentration, both the adsorption amount and adsorption efficiency of the Fe\(_3\)O\(_4\)-CS adsorbent are high enough for practical applications.

3.4 Effect of contact time
The effect of contact time on the adsorption of Orange I is shown in Fig. 10. Adsorption is fastest in the early stages of adsorption process. The adsorbent interacts with dye molecules through electrostatic attraction once the adsorbent is added to the dye solution. After 5 minutes of adsorption, the adsorption amount was over 100 mg g\(^{-1}\). This rapid uptake is due to high availability of vacant sites on the surface of the adsorbent [37]. The maximum adsorption amount was observed after 30 minutes. At this point, the vacant sites were all occupied by dye molecules, and saturation was reached. The optical contact time for the adsorption of Orange I was 30 minutes. This short contact time is feasible for practical applications.

3.5 Isotherm study
The equilibrium adsorption isotherm is an important parameter in an adsorption system. The Langmuir and Freundlich models were used to describe the equilibrium characteristics of Orange I adsorption onto Fe\(_3\)O\(_4\)-CS.

3.5.1 Langmuir Isotherm. The Langmuir model is based on the assumption of monolayer adsorption without interactions between the adsorbed molecules. The equation can be expressed as:

\[
\frac{C_e}{Q_e} = \frac{1}{K_L Q_0} + \frac{C_e}{Q_0}
\]

where \(C_e\) is the equilibrium concentration of Orange I solution (mg L\(^{-1}\)), \(Q_e\) is the adsorbed value of Orange I at the equilibrium concentration (mg g\(^{-1}\)), \(Q_0\) is the maximum adsorption amount (mg g\(^{-1}\)), and \(K_L\) is the Langmuir binding constant. The plot of \(C_e/Q_e\) versus \(C_e\) is a straight line in Fig. 11(a). The correlation coefficient is \(R^2 = 0.9995\). The value of \(Q_0\) obtained from the

| Table 1. Langmuir and Freundlich parameters for adsorption of Orange I by Fe\(_3\)O\(_4\)-CS. |
|---------------------------------------------------------------|
| **Langmuir model**                              | **Freundlich model**                     |
| \(R^2\) | \(Q_0\) (mg g\(^{-1}\)) | \(K_L\) (L mg\(^{-1}\)) | \(R^2\) | \(K_F\) | \(b_F\) |
|------------------|------------------|------------------|------------------|------------------|------------------|
| 0.9995          | 180.8            | 0.1760           | 0.8174           | 57.47            | 0.2025           |

Figure 11. Plots of Langmuir (a) and Freundlich (b) isotherms. doi:10.1371/journal.pone.0108647.g011
Langmuir isotherm is 180.8 mg g\(^{-1}\), which is perfectly consistent with the experimental data. It also indicates that the adsorption process was mainly monolayer.

### 3.5.2 Freundlich Isotherm

The Freundlich model is based on the assumption of adsorption on a heterogeneous surface. The equation can be expressed as:

\[ \ln Q_e = b_F \ln C_e + \ln K_F \]  \hspace{1cm} (3)

where \( Q_e \) is the adsorbed value of Orange I at the equilibrium concentration (mg g\(^{-1}\)), \( b_F \) is a constant describing the adsorption intensity, \( K_F \) is the Freundlich constant, and \( C_e \) is the equilibrium concentration of Orange I solution (mg L\(^{-1}\)). Plots of \( \ln Q_e \) versus \( \ln C_e \) are presented in Fig. 11.(b). The correlation coefficient is \( R^2 = 0.8174 \), which indicates that the adsorption isotherm does not fit the Freundlich model very well. The value of \( b_F \) is lower than 1, suggesting a normal Langmuir isotherm [38]. The adsorption isotherm parameters are presented in Table 1.

### 3.6 Adsorption kinetics

To understand the mechanism of adsorption kinetics, pseudo-first-order and pseudo-second-order kinetic models were used to analyze the experimental data. The pseudo-first-order kinetic model can be expressed as [39]:

\[ \frac{dq_t}{dt} = K_1 (q_e - q_t) \]  \hspace{1cm} (4)

Under the conditions \( q_t = 0 \) at \( t = 0 \), and \( q_t = q_e \) at \( t = t \), the Equation can be converted into a linear kinetic equation:

\[ \ln (q_e - q_t) = \ln q_e - K_1 t \]  \hspace{1cm} (5)

where \( q_t \) (mg g\(^{-1}\)) is the amount of Orange I absorbed at time \( t \) (min), \( q_e \) is the amount of adsorbed dye at equilibrium (mg g\(^{-1}\)), and \( K_1 \) (min\(^{-1}\)) is the equilibrium rate constant of pseudo-first-order kinetic model. The plot of \( \ln (q_e - q_t) \) through time is presented in Fig. 12.(a).

The pseudo-second-order process can be written as follows [40]:

\[ \frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \]  \hspace{1cm} (6)

where \( q_t \) (mg g\(^{-1}\)) is the amount of Orange I absorbed on the adsorbent at time \( t \) (min), \( q_e \) is the amount of adsorbed dye at equilibrium (mg g\(^{-1}\)), and \( k_2 \) (g mg\(^{-1}\) min\(^{-1}\)) is the equilibrium rate constant of pseudo-second-order model. The plot of \( t/q_t \) over time is presented in Fig. 12.(b). The Orange I pseudo-first-order and pseudo-second-order correlation coefficients are 0.8664 and 0.9561, which illustrates that the pseudo-second-order mechanism offered a better fit than the pseudo-first-order mechanism. The kinetics parameters and rate constants are presented in Table 2.

### Conclusions

In this study, magnetic Fe\(_3\)O\(_4\)-CS adsorbent was prepared by a one-step method for the adsorptive removal of Orange I from aqueous solutions. High adsorption capacity was achieved through the ionic interactions between protonated amino groups (–NH\(_3^+\)) of chitosan and sulfonate groups (–SO\(_3^−\)) of Orange I. The pH, initial concentration and contact time played a significant role in the dye adsorption capacity of Fe\(_3\)O\(_4\)-CS. The maximum adsorption amount reached 183.2 mg g\(^{-1}\) at a concentration of 400 mg L\(^{-1}\) at pH 2.0. Isotherm modeling revealed that the Langmuir equation could better describe the adsorption of Orange I on the Fe\(_3\)O\(_4\)-CS as compared to Freundlich model and pseudo-second-order kinetic model fitted with experimental data well. The fast uptake and magnetic separation gives the Fe\(_3\)O\(_4\)-CS adsorbent a high potential for effective removal of Orange I in water treatment.

| Table 2. Kinetics parameters for the adsorption of Orange I by Fe\(_3\)O\(_4\)-CS. |
|---------------------------------------------------------------|
| **pseudo-first-order model** | **pseudo-second-order model** |
| \( R^2 \) | \( k_1 \) (min\(^{-1}\)) | \( R^2 \) | \( k_2 \) (g mg\(^{-1}\) min\(^{-1}\)) | \( q_e \) (mg g\(^{-1}\)) |
| 0.8664 | 1.178 x 10\(^{-1}\) | 0.9561 | 5.277 x 10\(^{-4}\) | 224.2 |

Figure 12. Plots of pseudo-first-order (a) and pseudo-second-order (b) kinetic models.
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Author Contributions
Conceived and designed the experiments: YKD MSP FQY. Performed the experiments: YKD YJH. Analyzed the data: YKD LYW WJG FQY.

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