Article

Preparation and Removal Properties of Cimetidine from Aqueous Solution by Waste Bricks Incorporated with Different Iron Oxides

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Abstract: In this study, to investigate the elimination of micropollutant from aqueous solutions by construction waste, waste red brick (WRB) with and without incorporated of iron species (goethite and hematite) were prepared by a simple method. The prepared materials were systematically characterized and batch experiments were conducted to study the elimination of typical micropollutant cimetidine from the water environment. Results showed that both two iron species could be successfully incorporated onto WRB and the main structure of WRB were maintained. The adsorption process of cimetidine onto bricks was fast due to the microstructure and useful adsorption sites on the surface of bricks. The Langmuir model fitted the experimental data better and the $q_{\text{max}}$ of cimetidine increased about 24.4% and 39.6% for the incorporation of hematite and goethite, respectively. The pH values influenced the adsorption behavior greatly and the favorable pH value was around 6.0. The charge screening effect and competition adsorption may influence the adsorption behavior together. Due to the interaction between cimetidine and bricks, the outer-sphere complexation may be formed in the adsorption process. This study shows new methods for the elimination of the micropollutant from the water environment and offers useful guidelines for the reuse of construction waste.

Keywords: cimetidine; goethite; hematite; iron oxide coated; waste red brick

1. Introduction

For the past decades, water pollution has got greater attentions due to the growing contaminants from traditional pollutants to emerging contaminants [1]. As one of the mostly emerging pollutants, the pharmaceuticals have attracted considerable attention due to the higher usage for both human health and livestock breeding [2]. Cimetidine (as showed in Table 1), a kind of typical pharmaceuticals, is widely used as H$_2$-receptor antagonist in mammals for the treatment of duodenal ulcers and gastric hypersecretion [3,4]. Like other pharmaceuticals, tons of cimetidine are consumed annually and continuously accessed into the environments through various pathways [5]. Numerous researchers have found the occurrence of cimetidine in many kinds of aqueous environments [6–8]. Although the relative concentration of cimetidine in the environment is low, reports showed that cimetidine had short or long-term adverse effects on many aquatic animals, especially altering the capacity of immunity and reproduction [9]. It is urgent to exploit new promising technology for the elimination of cimetidine from the water environment.
In recent years, with the increasing demand of clean water, the requirement is increasing for the development of advanced water treatment technologies, such as advanced oxidation, photocatalytic degradation, membrane filtration, and adsorption [10,11]. The oxidation of cimetidine with many materials (such as TiO₂ or UV/visible light) has been extensively investigated due to the high removal efficiency. Results showed that cimetidine could be removed by reactive oxygen species, which are produced in the advanced oxidation and photocatalytic degradation process [12]. However, the secondary pollutants may be introduced in the degradation process and the adverse effects may be enhanced [11]. Compared with other methods, adsorption remains gained more interest in the elimination of micropollutants from aqueous solutions due to the simplicity, low initial cost, eco-friendliness, and easy possibility of regeneration [13]. As the key factor of adsorption, desired adsorbents should have high surface area, amount adsorption sites, low cost, and easy regeneration.

With the rapid development of urbanization in China, a large number of construction waste produced in recent years [14]. As a typical construction waste, waste bricks could cause an adverse effect on the environment due to the low utilization rate [15]. Recently, waste bricks could be investigated as an adsorbent due to its low cost, stable chemical structure, micropore volume, which showed better performance than conventional adsorbents such as clay, silt, and lime soil [16]. Studies have showed that waste bricks could be used for the removal of dyes, phosphorus, and heavy metals from the water environment [17–19]. However, compared with the commercial adsorbents, the lower adsorption capacity of waste bricks inhibits their application in wastewater treatment [20]. Many physical or chemical methods could be used to improve the removal efficiency of waste bricks [21,22]. A report showed that the incorporation of TiO₂ onto waste brick could enhance the removal efficiency of Pb²⁺ [22]. Furthermore, few reports focus on the removal of micropollutants by waste brick. As we know, iron species showed good efficiency for the elimination of emerging micropollutants from aqueous solutions due to the inner-sphere complexes [13]. Nevertheless, most of these iron species show relatively low surface area, small particle size, and easy agglomeration, which inhibit their application in water treatment [23]. Therefore, we propose that functionalized waste bricks with different iron species may overcome the deficiencies remarked above. Due to the proper characteristics of waste brick, the iron species could be incorporated onto waste brick homogeneous heighten the adsorption efficiency of waste bricks.

In this study, to explore the new adsorbent with low cost and high efficiency for the removal of micropollutants, waste bricks with and without iron species were selected to investigate the removal of typical pharmaceutical-cimetidine from aqueous solutions. The waste red brick (WRB) was selected due to the high production amount in China. In addition, the functionalization of typical iron species (goethite and hematite) onto WRB was conducted to enhance the adsorption capacity of cimetidine. The prepared materials were systematically characterized and the influence of iron species was investigated. Furthermore, the adsorption behavior of kinetics, isotherms, pH, and ionic strength were investigated to study the insight into the adsorption of cimetidine onto waste bricks.

2. Materials and Methods

2.1. Materials and Chemicals

Cimetidine (>99%) was purchased from Tokyo Chemical Industry Corporation (Tokyo, Japan) and used without further purification. The WRB was obtained in Beijing University of Civil Engineering.
and Architecture (Beijing, China). Methanol was HPLC grade and purchased from Fisher Scientific Corporation (Waltham, MA, American). Other chemicals such as NaHCO$_3$, NaCl, NaOH, HF, HClO$_4$, FeSO$_4$·7H$_2$O, and Fe(NO$_3$)$_3$·9H$_2$O were all of analytical grade and purchased from Sinopharm Chemical Reagent Corporation (Beijing, China). All the solutions in this study were prepared by ultrapure water (Milli-Q).

2.2. The Preparation of Adsorbents

Waste red brick (WRB): The obtained waste brick was firstly broken in grains and sieved with the sizes of 0.5 to 1.0 mm. Then, the obtained material was soaked in hydrochloric acid solution with the concentration of 6 mol/L at 90 ± 1 °C for 24 h. The resultant material was recovered through filtered and washed several times with distilled water. Finally, the particles were dried at 105 ± 1 °C overnight and the adsorbent was finally obtained.

Furthermore, the functionalized of goethite and hematite onto WRB were carried out as described by Schwertmann [24].

Goethite coated waste red brick (GWRB): After N$_2$ had bubbled into distilled water for 30 min, 13.9 g FeSO$_4$·7H$_2$O was added the solution. Then 110 mL of 1 mol/L NaHCO$_3$ was added and the air had bubbled into the mixture with a flow rate of 30–40 mL/min. A certain amount of WRB was added into the solutions and the mixture was stirred continuously for 48 h. The pH value in the progress of oxidation is maintained at about 7.0 by the NaHCO$_3$ buffer and the oxidation was complete when the color changed. The mixture was filtered, washed several times with deionized water. The finally products were obtained by freeze-drying and named as goethite coated waste red brick (GWRB).

Hematite coated waste red brick (HWRB): 20 g Fe(NO$_3$)$_3$·9H$_2$O was added in 250 mL distilled water, which was preheated to 90 °C. Then 300 mL 1 mol/L KOH solution and 50 mL 1 mol/L NaHCO$_3$ solution were added into the mixture with the temperature at 90 °C. After a certain amount of WRB was added, the mixed solutions (pH = 8.0–8.5) was hold in a flask at 90 °C for 48 h. The mixture was cooled, filtered, and washed several times with deionized water. The finally products were obtained by freeze-drying and named as hematite coated waste red brick (HWRB).

2.3. The Characterization of Waste Bricks

The surface morphology of the waste bricks was performed by scanning electron microscopy (SEM, Hitachi Limited S-4800, Tokyo, Japan). The chemical analysis of adsorbents was obtained by an X-ray fluorescence (XRF, Shimadzu XRF-1800, Tokyo, Japan). Mineralogy of the incorporated iron oxides was characterized by using an X’pert PRO MPD (X-ray diffraction (XRD), PANalytical, Rotterdam, Holland) with Cu Kα radiation. Scans were conducted from 10° to 90° at a rate of 6° per minute and the Cu tube operated at 35 kV and 30 mA. The nitrogen adsorption isotherms of adsorbents were performed with a Micromeritics ASAP 2020 (Mike, Atlanta, Georgia State, USA). The Fourier transform infrared spectrometer (FTIR) of bricks was determined by Tenson 27 FTIR Spectrometer (Bruker, Berlin, Germany) and the powder samples were analyzed with the wavelength of 500–4000 cm. The adsorbents were digested with the acid mixture of hydrochloric acid, nitric acid, hydrofluoric acid, and perchloric acid compounds (5:5:3:2, v/v/v/v) by the electric heating plate, the iron content was measured by atomic absorption spectrophotometry (Hitachi Z-2010, Tokyo, Japan).

2.4. The Adsorption Experiments of Cimetidine onto Bricks

Batch experiments were used in the adsorption process and the dark condition was maintained in all experiments to inhibit the potential photo-degradation of cimetidine. The prepared WRB, GWRB, and HWRB were used in the adsorption behavior of cimetidine onto waste bricks. The kinetics adsorption processes were conducted as follows: WRB, GWRB, and HWRB were added into the three solutions with the initial cimetidine concentration of 5 mg/L and a dosage of 2 g/L. All the mixed solution was stirred with a speed of 150 rpm. The solution pH values were maintained at about 7.0 ± 0.1 by using 0.01 mol/L HCl and NaOH solution in all experiments. At different time intervals, the samples
were collected and filtrated through a 0.22 μm polytetrafluoroethylene membrane filter (Jinteng, Tianjin, China). The concentrations of cimetidine in the solutions were analyzed. The equilibrium adsorption amount of cimetidine onto bricks was calculated by Equation (1).

\[ q_e = \frac{V(C_0 - C_e)}{m}, \]  

where \( C_0 \) and \( C_e \) are the initial and equilibrium concentration of cimetidine (mg/L), \( q_e \) (ug/g) is the adsorption capacity at equilibrium, \( V \) is the volume of the solution (L), and \( m \) is the mass of the adsorbents (g).

Adsorption isotherms of cimetidine onto waste bricks were conducted by adding 60 mg WRB, GWRB and HWRB into 30 mL solution with the cimetidine initial concentrations were 1, 2, 5, 8, 12, 16, 20, and 40 mg/L. The solution pH values were maintained at 7.0 ± 0.1 by using 0.01 mol/L HCl and NaOH solution. The suspensions were shaken at a speed of 150 rpm for 24 h. Finally, the solutions were filtered through a 0.22 μm polytetrafluoroethylene membrane filter and the concentrations of cimetidine were analyzed.

Furthermore, the influences of pH values and ionic strength were investigated to gain insight into the adsorption properties of cimetidine onto WRB, GWRB, and HWRB. To determine the influence of pH values on the adsorption of cimetidine, a series of 5 mg/L cimetidine solutions were prepared with the pH values range from 4.0 to 10.0. The solution pH values were maintained at desired values by using 0.01 mol/L HCl and NaOH solution in all experiments. Other experimental conditions were the same as the isotherm experiments.

The ionic strength effect on the adsorption of cimetidine onto WRB, GWRB, and HWRB was studied. A series of solutions were prepared with different ionic strength (NaCl, 0.001–0.2 mol/L). The solution pH was maintained at 7.0 ± 0.1 by using 0.01 mol/L HCl and NaOH solution in all experiments. Other experimental conditions were the same as the isotherm experiments.

The concentration of cimetidine was analyzed by ultra performance liquid chromatography (UPLC, Waters) at a wavelength of 225 nm. The mobile phase was a mixture of 0.01% CH₃COOH solutions-methanol (65:35, v/v) and the flow rate was 0.4 mL/min.

2.5. Statistical Analyses

In this study, the adsorption kinetic parameters and the Langmuir parameters were obtained by non-linear least-squares regression analyses. Origin software (edition Origin 9.1, OriginLab, Northampton, MA, USA) was used to analyze all the data in the manuscript.

3. Results and Discussion

3.1. The Physicochemical Characterization of WRB, GWRB, and HWRB

Figure 1 shows the SEM micrographs of waste bricks before and after the deposition of goethite and hematite. As shown in Figure 1a, the WRB exhibited a relative compact appearance with certain grains. After the incorporation of goethite and hematite (Figure 1b,c), the main surface properties of WRB were maintained. Furthermore, new small particles were appeared on the surface of WRB, which revealed the heterogeneity of the iron deposit on the bricks. Table 2 shows the chemical analysis, specific surface area and amount of coated iron oxides on WRB, GWRB, and HWRB. The XRF results showed that the main constituent of three bricks was SiO₂, in addition to smaller amount of other oxides, such as Al₂O₃, iron oxide, CaO, and MgO. These results were consistent with the results reported by others [17]. Furthermore, after the incorporation of goethite and hematite, the main composition nearly unchanged except the slight variations of the oxide contents, which indicated the main structure of brick was not changed. After the digestion by acid, the results showed that the iron content on the GWRB and HWRB is higher than that of WRB. After the calculation, the amount of coated iron on the surface of GWRB and HWRB was 4.28 and 3.41 mg/g, respectively. These results were corresponding to the conclusion shown in the data of XRF. However, after the incorporation of
goethite and hematite, the specific surface area of GWRB and HWRB decreased from 17.46 to 8.2 and 2.2 m²/g, respectively.

Figure 1. The SEM images of waste red brick (WRB; a), goethite coated waste red brick (GWRB; b), and hematite coated waste red brick (HWRB; c).

Table 2. The chemical analysis, specific surface area and amount of coated iron on the surface of adsorbents.

| Sorbents | Chemical Analysis (weight %, X-ray fluorescence (XRF)) | Specific Surface Area (m²/g) | Fe Content (mg/g) |
|----------|-------------------------------------------------------|-----------------------------|------------------|
| WRB      | Si₂O₅Al₂O₃Fe₂O₅CaO MgO Others                         |                             |                  |
| GWRB     | Si₂O₅Al₂O₃Fe₂O₅CaO MgO Others                         |                             |                  |
| HWRB     | Si₂O₅Al₂O₃Fe₂O₅CaO MgO Others                         |                             |                  |
Table 2. The chemical analysis, specific surface area and amount of coated iron on the surface of adsorbents.

| Sorbents | Chemical Analysis (Weight %, X-ray Fluorescence (XRF)) | Specific Surface Area (m²/g) | Fe Content (mg/g) |
|----------|-------------------------------------------------------|-----------------------------|------------------|
| WRB      | Si₂O 70.37, Al₂O₃ 15.87, Fe₂O₃ 4.12, CaO 1.33, MgO 2.03, Others 5.69 | 17.46 | - |
| GWRB     | Si₂O 71.22, Al₂O₃ 15.47, Fe₂O₃ 5.17, CaO 1.73, MgO 1.77, Others 6.32 | 8.20 | 4.28 |
| HWRB     | Si₂O 69.58, Al₂O₃ 15.32, Fe₂O₃ 5.20, CaO 1.56, MgO 1.95, Others 7.54 | 2.20 | 3.41 |

The XRD results of GWRB and HWRB are shown in Figure 2. The two diagrams obtained for uncoated and coated brick are almost identical and characteristic of quartz. However, the XRD pattern of GWRB shows several well-defined characteristic peaks of (110), (120), (111), (121), and (151) [25], suggesting the existence of the goethite structure on the surface of GWRB. The goethite was coated successfully proved by the increase of (110) characteristic peak and the intensity of all other peaks in GWRB. In the same way, the powder XRD pattern of HWRB appears the characteristic peaks of (012), (110), and (024) [26]. The increase of the intensity peaks in this case is associated with the increase quantity of deposited iron oxides on the surface of bricks.

The FTIR of WRB, GWRB, and HWRB were illustrated in Figure 3. For all three bricks, the bands at 3439, 1090, 820, and 590 cm⁻¹ could be ascribed to the stretching vibrations of Si–OH, the asymmetric vibrations and symmetric vibrations of Si–O–Si, the symmetric vibrations of Fe–O–Fe, respectively [27,28]. Compared with results in the WRB, the main peaks of GWRB and HWRB were nearly unchanged, indicating that the incorporation of iron species did not change the main structure of the bricks. However, the intensity peak of Fe–O–Fe increased after the incorporation, suggested that the content of iron oxide on the surface of waste bricks increased.
3.2. The Adsorption Kinetics of Cimetidine onto WRB, GWRB, and HWRB

The adsorption kinetics of cimetidine onto WRB, GWRB, and HWRB are shown in Figure 4. The results showed that the adsorption of cimetidine onto three bricks was fast at the first 1 h then followed by a relatively slow process and finally reached the equilibrium at about 4 h. The rapid adsorption process of cimetidine onto bricks was probably attributed to the microstructure and abundant useful adsorption sites on the surface of bricks [29]. These results could also be observed by other reports, where the adsorption of heavy metal and basic blue onto waste bricks [15,16]. It is interesting to notice that the adsorption capacities of cimetidine on three bricks are different at the same experimental conditions. The equilibrium adsorption capacities of cimetidine onto WRB, GWRB, and HWRB are 112.8, 194.8, and 166.6 µg/g, respectively. The differences in adsorption capacities for the different adsorbents may be related to the physicochemical characteristics of adsorbents.
In order to analyze the adsorption kinetics of cimetidine onto waste bricks further, two typical models were used to fit the experimental data. The pseudo-first-order and pseudo-second-order models were represented by Equation (2) and Equation (3), respectively [30,31].

\[
\frac{dq_t}{dt} = K_1(q_e - q_t), \quad (2)
\]
\[
\frac{dq_t}{dt} = K_2(q_e - q_t)^2, \quad (3)
\]
where \(q_t\) and \(q_e\) were the adsorption capacity at time \(t\) and saturation (ug/g), \(K_1\) (s\(^{-1}\)) and \(K_2\) (ug/g/s) are the adsorption rate constants of two models.

As shown in Table 3, the correlation coefficients for pseudo-second-order model were relatively higher \((R^2 = 0.93–0.97)\), suggesting that the adsorption of cimetidine onto three bricks may be chemisorption. Furthermore, compared with experimental results, the equilibrium adsorption capacities calculated by the pseudo-second-order model were more reasonable than the pseudo-first-order model. Therefore, the adsorption mechanism was mainly attributed to the surface complex reaction between cimetidine and the useful adsorption sites of the bricks. Compared with the \(q_e\) of the three bricks, the incorporation of iron species could increase the amount of useful adsorption sites. The results were in accordance with the adsorption of tetracycline onto iron oxides, which shared the same functional groups (–NH\(_2\) and –CH\(_3\)) as cimetidine [32]. These results suggested that WRB, GWRB, and HWRB could be used as useful adsorbents for the removal of cimetidine from aqueous solutions. Especially for GWRB and HWRB, due to the microstructure and abundant useful adsorption sites, these two adsorbents have the potential for the application in the water treatment.

| Adsorbents | Pseudo-First-Order Model | Pseudo-Second-Order Model |
|------------|--------------------------|---------------------------|
|            | \(K_1\) | \(q_e\) | \(R^2\) | \(K_2\) | \(q_e\) | \(R^2\) |
| WRB        | 6.50  | 104.7  | 0.91   | 80.43  | 11.3   | 0.93   |
| HWRB       | 3.12  | 155.4  | 0.86   | 29.08  | 16.6   | 0.95   |
| GWRB       | 3.13  | 180.5  | 0.93   | 21.30  | 19.5   | 0.97   |

3.3. The Adsorption Isotherms of Cimetidine onto WRB, GWRB, and HWRB

Adsorption isotherms are the basic requirements for the investigation or the design of adsorption systems. The adsorption isotherms of cimetidine onto WRB, GWRB, and HWRB are shown in Figure 5. For all bricks, the adsorption capacity of cimetidine increased with the increasing equilibrium concentration. All three isotherms displayed a sharp slope at the lower initial concentration, which indicated the high adsorption efficiency. This result could be ascribed to the abundant adsorption sites on the bricks. With the equilibrium concentrations of cimetidine increased, the adsorption sites gradually turned into saturated and the adsorption isotherm finally reached a plateau.

To comprehend the adsorption isotherms of cimetidine further, two typical isotherm models were applied to investigate the adsorption properties. The Langmuir model is usually based on the hypothesis that the adsorbate only forms a monolayer around the adsorbent surface. The Freundlich isotherm model is usually used to describe the adsorption characteristics for heterogeneous surface [30,33]. The Langmuir and Freundlich models were represented by Equations (4) and (5), respectively.

\[
q_e = q_{\text{max}}K_LC_e / (1 + K_LC_e), \quad (4)
\]
\[
q_e = K_fC_e^{1/n}, \quad (5)
\]
where \(q_{\text{max}}\) is the maximum adsorption capacity (ug/g), \(K_L\) is the Langmuir constants of adsorption, \(K_f\) is the Freundlich constant related to overall adsorption capacity (ug/g), and \(n\) is the constant related to surface heterogeneity.
The parameters and correlation coefficients of the two isotherms models were shown in Table 4. The results showed that Langmuir model was more appropriate for the description of the adsorption of cimetidine onto WRB, GWRB, and HWRB ($R^2 = 0.98–0.98$), which could be attributed to the monolayer molecular adsorption between cimetidine and bricks. These results were the same as the results that the adsorption of other micropollutants onto adsorbents [22,23]. Furthermore, the values of $1/n$ were close to zero (0.32–0.41), illustrating that the adsorption of cimetidine onto WRB, GWRB, and HWRB was favorable [34].

![Adsorption isotherms of cimetidine onto WRB, GWRB, and HWRB.](image)

**Figure 5.** Adsorption isotherms of cimetidine onto WRB, GWRB, and HWRB.

**Table 4.** The parameters and correlation coefficients of Langmuir and Freundlich adsorption isotherms.

| Adsorbents | Langmuir Model | Freundlich Model |
|------------|----------------|------------------|
|            | $K_L$ | $q_{max}$ | $R^2$ | $K_f$ | $1/n$ | $R^2$ |
| WRB        | 0.12  | 172.1    | 0.99  | 34.2  | 0.41  | 0.96  |
| HWRB       | 0.20  | 240.3    | 0.99  | 61.8  | 0.36  | 0.87  |
| GWRB       | 0.23  | 214.1    | 0.98  | 66.7  | 0.32  | 0.93  |

Furthermore, as showed in Table 4, the calculated $q_{max}$ of three bricks followed by the order of GWRB > HWRB > WRB, suggesting the adsorption efficiency of waste bricks was heightened by the incorporation of iron species. Compared with hematite, results showed that goethite could offer more useful adsorption sites for cimetidine removal. Furthermore, due to the –OH group of goethite, the GWRB was more easily formed complex with functional groups (–NH$_2$ and –CH$_3$) of cimetidine and heightened the adsorption efficiency of waste bricks. The same results were also confirmed in the adsorption kinetics. The conclusion also showed that the waste bricks could be used as a promising adsorbent in decontamination of cimetidine in the water environment.

3.4. The Effect of pH on the Adsorption of Cimetidine

The effect of pH on the adsorption of cimetidine onto WRB, GWRB, and HWRB are shown in Figure 6. The results show that the solution pH values affect the adsorption of cimetidine remarkably. For all three bricks, the adsorption capacity of cimetidine increased to a maximum with pH values increased from 4.0 to 6.0 and then decreased with the solution pH values increased further. This result is in agreement with previous investigations on the adsorption of other pharmaceuticals, which showed a similar structure [35]. The effect of solution pH could be attributed to the speciation of cimetidine in aqueous solution and the main surface chemistry of the three bricks.
As shown in Table 1, the pKa value of cimetidine was about 6.8. When the pH < 6.8, cimetidine in the solution was positively charged, while cimetidine showed negative charged with the pH > 6.8. As shown in Figure 7, in the experiment pH values, the electrical property of WRB, GWRB, and HWRB was almost negative. With the pH values increase from 4.0 to 7.0, the percentage of the neutral form of cimetidine increased, which caused the cimetidine more easily complexed with bricks. The adsorption capacity of cimetidine onto bricks increased due to the increasing electrostatic attractions. With the pH values increasing further, the percentage of anionic form of cimetidine increased. Due to the increasing electrostatic repulsion, the adsorption of cimetidine onto three bricks was inhibited. The maximum adsorption capacity of cimetidine onto three bricks was almost at about 6.0, which is close to the pKa values of cimetidine suggesting a significant fraction of the N–H groups. Such adsorption behavior, which the maximum adsorption occurs at the pKa values has also been observed for the adsorption of some other pharmaceuticals onto iron species [32,35]. Furthermore, the adsorption capacity also followed the order GWRB > HWRB > WRB when the pH values below 7.0, which is also confirmed in the adsorption kinetics and isotherms.
3.5. Effect of Ionic Strength on the Adsorption of Cimetidine onto Bricks

The effect of ionic strength on the adsorption of cimetidine onto WRB, GWRB, and HWRB is presented in Figure 8. The results showed that the ionic strength affected the adsorption greatly and the increasing ionic strength restrained the adsorption of cimetidine onto all three bricks. Compared with the inner-sphere complexes, the outer-sphere complexes adsorption is expected to be more vulnerable by the ionic strength. Therefore, the adsorption of cimetidine onto WRB, GWRB, and HWRB conjectured to be outer-sphere surface complexes.

![Figure 8. The effect of ionic strength on the adsorption of cimetidine onto WRB, GWRB, and HWRB.](image)

As shown in Figure 8, the effect of ionic strength on the adsorption could be divided into two regions. With the ionic strength increase from 0 to 0.02 mol/L, the adsorption capacity of cimetidine decreased sharply from 92.7, 119.4, 108.6 to 14.2, 17.5, and 4.6 μg/g for WRB, GWRB, and HWRB, respectively. The competition between cimetidine and Na⁺ may induce the decrease in cimetidine adsorption. Resemble results could be reported by the adsorption of antibiotics on adsorbents [36]. With the ionic strength further, the adsorption capacity increased slightly for all bricks. This phenomenon was possible derived from the charge screening effect that can reduce the repulsion between cimetidine and bricks, which was also observed by the adsorption of triclosan onto activated carbon, kaolinite, and montmorillonite [37].

4. Conclusions and Recommendations

In this study, WRB with and without iron species were shown as potential adsorbents for the removal of cimetidine from aqueous solutions. The pH values affect the adsorption properties greatly and the maximum adsorption capacity was around the pH value 6.0. The adsorption isotherms fitted the Langmuir modes well and the \( q_{\text{max}} \) of three bricks followed the order GWRB > HWRB > WRB. After the incorporation of goethite and hematite, the \( q_{\text{max}} \) of cimetidine onto WRB increased about 39.6% and 24.4% respectively. The adsorption mechanism mainly attributed to the outer-sphere complexes between cimetidine and adsorbents. This investigation confirmed the possibility of using WRB as an efficient adsorbent for the removal of micropollutants from aqueous solution. This result could show new guidelines and methods for the elimination of micropollutant and reuse of construction waste.

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