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Removal of Zn(II) from aqueous solutions by adsorption using different types of waste bricks

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ABSTRACT

Bricks constitute a significant part of construction and demolition wastes. These bulky materials, if not reused or recycled, are mostly landfilled for disposal. In this study, adsorption of Zn(II) by four different types of waste bricks was systemically investigated through batch experiments. The maximum adsorption capacities were 0.31–3.67 mg/g for the four types of bricks, and the capacities decreased in the order of cinder bricks>lime sand bricks>gray clay bricks>red clay bricks, which is due to the different nature of the bricks such as the elemental composition, surface functional groups, surface area and pore volume. Adsorption of Zn(II) could be well described by the Langmuir model and Pseudo-second-order kinetics model. The removal efficiency of the two types of non-clay based bricks could reach more than 80% with an 8 g/L dosage, which is much larger than that of the two types of clay-based bricks. Solution chemistry such as pH and ionic strength had significant effects on the adsorption of Zn(II). The adsorption of the two types of clay-based bricks was more sensitive to the influence of pH than the two types of non-clay-based bricks, while the effect of ionic strength on the adsorption of Zn(II) by the four types of bricks was similar. The optimal dosage and particle size of bricks were also examined. The results could provide basic data for the application of bricks as adsorbents for remediation.

Keywords: Adsorption; Bricks; Zinc; Heavy metal

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1. Introduction

Zinc is one of the most important heavy metals in the environment and mainly derives from anthropogenic sources, including tire wear, refuse burning, mining, metallurgy, steel making and refining [1,2]. When the Zn(II) content is relatively small, it is a necessary element for life. However, when the Zn(II) content exceeds a certain level, it is harmful to living things and the environment, and has been identified as an important pollutant in surface water and underground water [3]. The United States Environmental Protection Agency maximum acceptable contamination level of Zn(II) for surface or groundwater to be used in the drinking supply is 0.8 mg/L [4]. Beyond the acceptable range, zinc is toxic. Zinc is reported to be one of the primary polluting elements. The main symptoms of zinc poisoning are dizziness, muscular stiffness, lack of appetite, imbalance of electrolytes and so on [4,5]. Therefore, it is necessary to remove Zn(II) ions from liquid wastes to limits accepted by national and international regulatory agencies before their discharge to the environment [4,6].

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Various advanced chemical or physical treatment processes such as irradiation, electrocoagulation, ion exchange, membrane separation, advanced oxidation and adsorption by various of adsorbents are employed for the removal of heavy metal ions, including Zn(II) ions, from aqueous solutions. Different treatment processes have their own advantages and disadvantages. Among them, adsorption has been recognized as one of the most important techniques for Zn(II)-contaminated wastewater treatment because of its operational simplicity, sludge-free operation and there use potential of adsorbents during long-term application. It can be used in a wide variety of settings [4,7].

Every day, thousands of tons of clay-based construction and demolition wastes are disposed [8]. Among them, construction waste bricks are one of the oldest and most demanding building materials in the world [9]. As a result of the rapid development of the economy in China, huge amounts of building wastes are being generated [10]. The waste bricks generated from reconstruction of old cities accounts for about 50–70% of this waste. The output of waste brick is about 52 million ton/y [11–13]. These large amounts of construction waste mainly end up in landfills through a costly process [8,13]. The disposal of such brick wastes has become a challenging problem. Therefore, to turn those waste bricks into resources is of great importance to both economics and promotion of the reuse of wastes.

In recent years, some researchers have realized the potential application of waste bricks in environmental remediation due to their large surface area,unique characteristics and low cost [3]. Waste bricks are more cost-effective than other similar reported materials. On one hand, waste bricks cost less than other materials, about 60 yuan/ton in China. On the other hand, as waste, the bricks need to be treated, and their reuse as adsorbents could add more value to them. Compared with other similar reported materials, waste bricks are much more economical than materials that are applied in water treatment such as zeolite, bentonite and montmorillonite, which cost hundreds of yuan per ton in China. The waste bricks were reported to show good removal efficiency toward various contaminants including fluoride [14,15], phosphate, nitrate [9,10,16–19], chloride, detergents [20] and heavy metals [21] by an adsorption process. For removal of heavy metals, a number of previous studies focused on examining the adsorption capacities of clay-based bricks. The clay-based bricks are usually negatively charged, which allows adsorption of heavy metals such as Pb(II), Cr(VI), Cu(II) and Zn(II) via electrostatic interaction [13,22] . In addition, bricks subjected to different pretreatments were selected as adsorbents to examine their removal efficiency for heavy metals. Namal et al. investigated adsorption of Cr (VI) on clay-based bricks fired at different temperatures (i.e., 100°C, 200°C, 400°C, 600°C and 900°C). They observed that brick fired at 200ºC showed the highest Cr(VI) removal efficiency, and the extent of Cr(VI) removal reached 60% [23]. Boujelben et al. found that iron-oxide-coated bricks had more adsorption capacity (i.e., 5.5 mg/g) for Pb(II) than pristine bricks [24]. Fethi et al. pretreated bricks with basic solutions, which increased their removal capacity toward basic blue 41 by two-fold. Boujelben et al. evaluated copper and nickel adsorption on artificially manganese oxide-coated burned brick, and discovered that the adsorption capacity could reach 2.4 mg Ni/g and 3.7 mg Cu/g [25]. The HCl treatment of Bangui brick favored the formation of new hydroxyl groups at the brick surface, and a subsequent NaOH leaching generated sodic (negatively charged) sites that were found to improve the adsorption capacity of this material in the removal of divalent metals (Pb(II)) from water. The adsorption capacity of Pb(II) can reach 20.72 mg/g [26]. The above studies all show that pretreatment can change the characteristics of bricks, as a consequence, affect their removal efficiency for heavy metals [27].

Besides pretreatment of bricks, the characteristics of bricks could be also varied by selecting different types of bricks. Nevertheless, most studies have focused on investigation of the removal efficiencies of clay-based waste bricks, possibly due to their large production volume. There are few studies on other types of bricks. Sand lime bricks, a type of new building material, are suitable for the construction of multilayer composite structures. Cinder bricks are applied in walls and foundations of industrial and civil buildings. The above new types of bricks also have a wide range of applications and are expected to generate large amounts of wastes. Also, the commonly used clay-based bricks consisting of red and gray bricks have applied to different constructions according to their color. The adsorption behavior of bricks toward heavy metals may be different based on the different nature, surface area, micropore volume and surface charge of each type of brick. Investigation of the adsorption behavior of contaminants onto different types of bricks is of great importance in selecting adsorbents with good removal efficiency as well as to gain knowledge on the adsorption mechanisms.

In this study, four types of waste bricks (i.e., red clay brick, gray clay brick, lime sand brick and cinder brick) that are commonly used in construction were selected as adsorbents to examine their removal efficiency toward the heavy metal Zn(II) in aqueous solutions. The main objectives of the present study are: i) to compare different types of bricks regarding the removal efficiency of Zn(II); ii) to examine the effect of solution chemistry, including pH and ionic strength, on the adsorption behavior of Zn(II) on each type of brick; (iii) to investigate the effect of dosage and particle size for each type of brick on the removal efficiency toward Zn(II).

2. Materials and methods

2.1. Reagents

All the reagents and solvents used were of analytical grade. A heavy metal–Zn(II) standard solution (1000 mg/L in 2% HNO_3) was purchased from theAnalysis Center of the National Non-ferrous Metals and Electronic Materials. Sodium chloride (Beijing Chemical Works) was used to adjust the ionic strength of the solutions after drying in an oven at 120ºC for 2 h. Hydrochloric acid and sodium hydroxide solutions were used to adjust the pH of the metal ion solutions.

2.2. Four types of bricks used as adsorbents

The four types of bricks used in this study were obtained from Hebei province in China. The red clay bricks and gray clay bricks mainly consisted of clay. The red color of red clay bricks is due to the color of ferric oxide, while the gray color of gray clay bricks is due to the generation of ferrous oxide in the firing process. The main constituents of lime sand bricks are lime and quartz sand, and the main constituents of cinder bricks are lime, sand and cinder. The bricks were crushed into coarse grains and ground into fine powders, followed by sieving through 0.15–0.5 mm, 0.5–1 mm,1–2 mm and 2–5 mm sieves to be used as adsorbents. The brick particles were washed thoroughly with deionized water to remove dust and other foreign matter and then oven-dried at 105°C for 12 h. After drying, 200 g portions of brick particles were then soaked in 0.5 L 0.1 M H_2SO_4 overnight and then washed with distilled water. The bricks were dried in the oven to a constant weight. Then the above adsorbents were stored in a vacuum desiccator until use.

2.3. Characterization techniques

The elemental composition of bricks was determined by X-ray fluorescence (XRF, Shimadzu XRF-1800, Japan). The surface micrographs of bricks were measured by scanning electron microscopy (SEM, Hitachi Limited S-4800, Japan-SEM). N_2 adsorption isotherms were collected at 77 K on a Micromeritics ASAP 2020 HD88 (Mike, USA). The specific surface area was calculated by the BET method, while the pore volume and pore size distribution were calculated by the BJH method. The functional groups on each type of brick before and after Zn(II) adsorption were analyzed by Fourier transform infrared spectroscopy (FTIR) (Bruker, Germany). The concentration of Zn(II) after adsorption by bricks was determined by inductively coupled plasma mass spectrometry (ICP-MS) (Elan 5000, Perkin Elmer, USA).

2.4. Adsorption experiments

2.3.1. Batch experiments

The batch experiments were conducted to investigate the adsorption behavior of Zn(II) onto each type of brick. The experiments were conducted at 25°C. The adsorption kinetics was studied and adsorption isotherms were measured. All experiments were performed in duplicate.

In the adsorption kinetics experiments, 500 mL solutions were prepared with Zn(II) concentration of 5 mg/L. Samples of each type of brick with size ranging from 0.5–1 mm were then added into solution with a dosage of 8 g/L. Solution samples were taken at different time intervals (i.e., 1 min, 3 min, 5 min, 10 min, 20 min, 30 min, 45 min, 1 h, 2 h, … 24 h and 48 h), filtered with 0.22 µm pore diameter nylon micropore membranes, and Zn(II) was then analyzed by ICP-MS. For measuring adsorption isotherms, a series of 40 mL solutions with Zn(II) concentrations of 0–30 mg/L were prepared. To study thermodynamic properties, adsorption tests were conducted at three temperatures (i.e., 15, 25 and 45°C).Adsorbents having the same size distribution (i.e., 0.5–1 mm) were then added into the solutions with a dosage of 8 g/L. After reaching equilibrium, the solutions were filtered and the concentration of Zn(II) was analyzed. During the reaction, the solution pH was controlled at 5.

To investigate the effect of pH on the adsorption behavior of Zn(II) onto each type of brick, batch experiments were carried out at pH 2–6. The pH was adjusted with 0.01 M hydrochloric acid or 0.01 M sodium hydroxide solutions. In order to investigate the influence of ionic strength on adsorption, 40 mL Zn(II) solution with initial concentration of 5 mg/L was put into each centrifuge tube. The pH of the solution was controlled at 5. Four kinds of bricks with particle size of 0.5–1 mm were added to the solution, with solid-liquid ratio of 8 g/L. Sodium chloride was used to regulate the ionic strength at different levels (i.e., 0, 0.2, 1, 5, 10, 20, 50,…100 and 200 mg/L). All samples were shaken at 135 rpm. The effect of particle size on the adsorption of Zn(II)for each type of brick was investigated in four different size ranges: 0.15–0.5 mm, 0.5–1 mm, 1–2 mm and 2–5 mm. The initial concentration of Zn(II) was 5 mg/L and the solution pH was controlled at 5.

The main method used to determine the cation-exchange capacity (CEC) was ammonium acetate extraction [28]. 2.5 g of each type of brick with particle size below 2 µm was placed in a 100 mL beaker with 50 mL of 1 M NH_4O Ac at pH 7. In order to completely disperse the brick powder, the solution was sonicated for 40 min and left to stand for 16 hours. The sample was centrifuged until the brick powder was completely separated from the supernatant. Then the supernatant was collected and the concentrations of Ca, K, Na, Mg and Al were determined by ICP-OES after 10-fold dilution with 5% HNO_3. The brick powder remaining at the bottom of the centrifuge tube was separated, and the above steps were repeated. The results in mg/L were converted to mmol/g to determine the individual cation CECs of Ca, K, Na, Mg and Al. The total CEC of the bricks was determined by summing up the above individual values.

2.4.2. Models of adsorption kinetics and isotherms

The data on the adsorption kinetics of Zn(II) were fitted using the Pseudo-first-order [Eq. (1)] and Pseudo-second-order models [Eq. (2)] [29].

$$q_t = q_e(1 - e^{-k_1 t}) \tag{1}$$

$$q_t = \frac{q_e^2 \cdot k_2 \cdot t}{1 + q_e \cdot k_2 \cdot t} \tag{2}$$

where k_1 (h^{-1}), k_2 (g/mg·h) are the rate constants of adsorption, q_e(mg/g) is the adsorption capacity at equilibrium and q_t is the amount of metal adsorbed at time t (h).

Different models were used to correlate and compare the experimental adsorption data in order to provide accurate fits for the adsorption isotherms. In this study, the Freundlich [30] and Langmuir [30,31] isotherm models were used to fit the adsorption isotherms. The Freundlich model takes into account the heterogeneity of binding sites. The Langmuir model describes adsorption on equivalent sites of the adsorbent material, which can be saturated to obtain a monolayer. The Freundlich and Langmuir models are shown in Eqs. (3) and (4), respectively:

$$q_e = K_F \cdot C_e^{1/n} \tag{3}$$

$$q_e = \frac{q_m \cdot K_L \cdot C_e}{1 + K_L \cdot C_e} \tag{4}$$

where q_m is the maximum adsorption capacity of the material, expressed in mg/g; K_F((mg/g)(L/mg)^n) and K_L(L/mg) are the constants for the Freundlich and Langmuir models, respectively, from which information about the binding capacity or affinity of the adsorbent toward the metal ions can be obtained; C_e (mg/L) is the metal concentration in solution at equilibrium; n is a dimensionless empirical parameter, which gives information on the strength of the adsorption.

The Zn(II) ion adsorption capacity at different contact times (q_t, mg/g) in the kinetic study, as well as the q_e at different metal/adsorbent ratios in the equilibrium study, was calculated by Eq. (5) [30]:

$$q_t = \frac{V \cdot (C_0 - C_t)}{m} \tag{5}$$

where V (L) is the volume of the metal solution and m is the mass of dry adsorbent (g); C_0 (mg/L) and C_t (mg/L) are the Zn(II) metal ion concentrations in solution at t = 0 and t = t, respectively. At the equilibrium condition, Eq. (5) was applied by replacing C_t (mg/L) with C_e (mg/L) to calculate q_e(mg/g). All data was analyzed by Origin 8.6.

3. Results and discussion

3.1. Characterization of different types of bricks

The four types of bricks systematically characterized before and after adsorption of Zn(II) showed different characteristics from the aspects of morphology (Fig. 1), elemental composition (Table 1), specific surface area, pore volume (Table 2) and surface functional groups (Fig. 2). The characterization results combined with adsorption data could give a comprehensive understanding of their adsorption behavior.

Fig. 1. shows SEM micrographs of the four types of bricks before and after adsorption. Before adsorption, it was observed that the surfaces of all types of bricks were uneven with large numbers of pore spaces, which perhaps provide sites for adsorption of Zn(II) either physically or chemically [32]. After adsorption, it could be seen that the pores of all types of bricks are substantially covered due to the adsorption of Zn(II) and aggregation of brick powder after shaking. In addition, after adsorption, the surfaces of all types of bricks become smoother than before adsorption, which was probably due to the deposition of heavy metals by physical adsorption or a progressive change in the mineralogy of the brick surface [33].

Table 1 shows the elemental composition of the four types of bricks. All types of bricks mainly consisted of Ca, Mg, Al and Si. The content of Ca was the largest among all elements for lime sand bricks and cinder bricks (i.e., 32.11% and 32.45%, respectively), while the content of Si was largest among all elements for gray clay bricks and red clay bricks (i.e., 28.71% and 27.62%, respectively). The differences in elemental composition may influence the adsorption behavior toward contaminants. Calcium and magnesium are exchangeable cations, and Zn(II) ion exchange occurs. Therefore, those elements have a positive effect on Zn(II) adsorption. In addition, there is some amount of clay in the two kinds of clay bricks (i.e., red clay bricks and gray clay bricks). Isomorphous substitution or formation of tetrahedral structural units often take place in the eight main sticky clay minerals, so that charge imbalance occurs. Together with crystal breakage with exposure of fracture surfaces to oxygen, these characteristics give the clay mineral surface a permanent negative charge. The chemical composition results show that certain samples (cinder bricks and lime sand bricks) contain Cr. Therefore, an experiment on the release of Cr from the bricks was also carried out. When the brick dosage was 1 g/L, 8 g/L and 20 g/L, the concentration of Cr in aqueous solution was extremely low (<0.047 μg/L or under the detection limit). The amount of Cr originally contained in the bricks was not easily released into the solution and should not affect the adsorption behavior of Zn(II).

Table 2 shows the BET and BJH specific surface area (m^2/g) as well as the pore volume obtained by the BJH adsorption method for the four types of bricks with the same particle size of 0.5–1 mm. The BET and BJH specific surface areas of the four types of bricks were between 2.53–3.97 m^2/g and 2.85–5.45 m^2/g, respectively. The specific surface area of the four types of bricks decreased following the order cinder bricks>lime sand bricks>gray clay bricks>red clay bricks. The BJH pore volume of all types of brick was between 0.0034–0.018 cm^3/g and followed the same order as the specific surface area. Both the specific surface area and pore volume of the bricks could affect their adsorption of Zn(II) [34].

The presence of functional groups on the surface of adsorbents was confirmed using FTIR analysis [4]. Fig. 2 shows the FTIR spectra of the four types of bricks before and after Zn(II) adsorption in order to determine changes in the vibration frequencies of functional groups. The FTIR spectra indicate the complex nature of the bricks. The bands at 1444 cm^{-1} before and 1452 cm^{-1} after adsorption were assigned to the O–H bending vibration of lime sand bricks, with the corresponding peaks for cinder bricks at 1437 cm^{-1} before adsorption and at 1436 cm^{-1} after adsorption [35]. The peaks at 2520 cm^{-1} for lime sand bricks and at 2534 cm^{-1} for cinder bricks before adsorption can be assigned to –C=OH-OH [35]. The bands in the range 729–877 cm^{-1} may be assigned to –C=CH$_2$ double bonds. The bands in the range 1010–1092 cm^{-1} can possibly be assigned to –C=O-O-. More functional groups were observed on the cinder bricks and lime sand bricks than on the two clay-based bricks. Therefore, carboxyl and hydroxyl groups may take part in the adsorption of Zn(II) ions [36]. Carboxyl (–COOH), hydroxyl (–OH), phenol (R–OH) groups are generally accepted as the main groups contributing to coordination of heavy metal ions such as Cu(II), Zn(II), Cd(II), Co(II) and Pb(II) etc. [36,37].

3.2. Adsorption kinetics of Zn(II) onto different types of bricks

In the investigation of the adsorption kinetics of Zinc(II) with brick particles, it is important to monitor how fast the interaction takes place [22]. Fig. 3 shows the kinetic behavior of Zn(II) uptake by the four types of bricks. Under the same conditions (i.e., C_0 = 5 mg /L, w = 8 g/L, stirring speed = 135 rpm, pH = 5), adsorption of Zn(II) onto all types of bricks reached equilibrium within 10 hours. It is apparent that the adsorption is fast at the initial stage and then reaches a plateau, indicating that adsorption equilibrium is reached. Initially, the removal is fast due to adsorption of

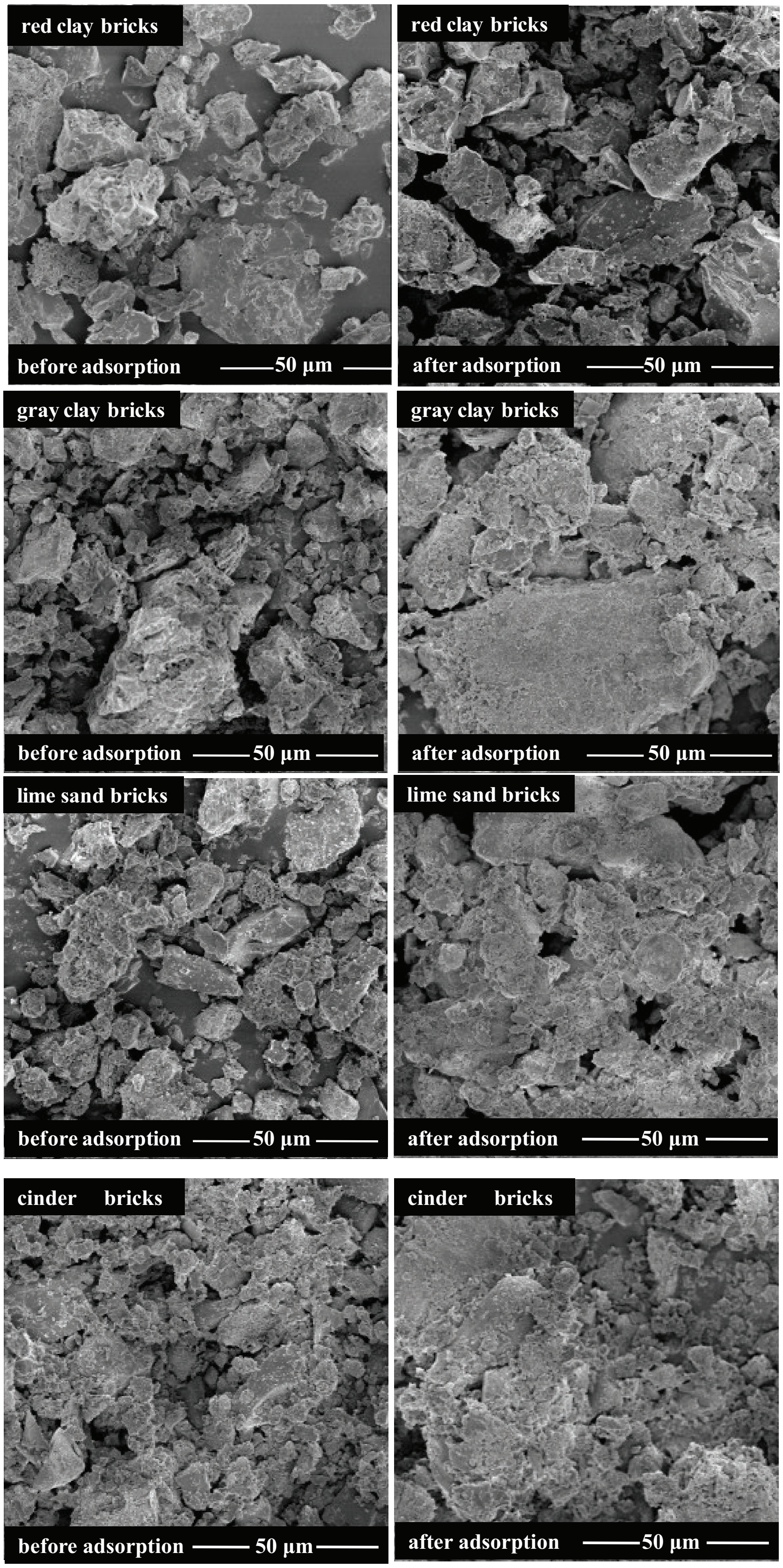

Fig. 1. SEM images of the four types of bricks (i.e., red clay bricks, gray clay bricks, lime sand bricks and cinder bricks) before and after Zn(II) adsorption.

Zn(II) ions onto the vacant surface sites, and thereafter the rate decreases due to saturation of surface sites [38].

The adsorption kinetics data were fitted by Pseudo-first-order and Pseudo-second-order models. Table 3 shows the parameters deduced from the above two models.

Table 1
Elemental composition of the four types of bricks

Element (%)	Cinder bricks	Lime sand bricks	Gray clay bricks	Red clay bricks
Ca	32.45	32.11	4.51	1.30
Al	3.97	4.26	8.96	12.93
Mg	9.64	8.92	1.85	1.02
Si	12.35	12.86	28.71	27.62
K	0.39	0.53	2.19	2.38
Fe	–	–	3.60	4.74
F	0.22	0.33	–	–
Na	0.17	0.13	1.42	0.46
Mn	0.13	0.15	0.09	0.07
S	0.52	0.44	0.48	0.47
Cr	1.06	0.93	0.02	–
Ti	0.26	0.37	0.43	0.56
Cl	0.032	0.038	0.013	0.016

Table 2
Surface characteristic parameters of the four types of bricks

	BET specific surface area (m^2/g)	BJH specific surface area (m^2/g)	BJH Pore volume (cm^3/g)
Red clay bricks	2.53	2.85	0.0064
Gray clay bricks	2.65	3.41	0.011
Lime sand bricks	3.92	4.75	0.017
Cinder bricks	3.97	5.44	0.018

It could be concluded that the adsorption kinetics of Zn(II) onto all types of bricks was better fitted by the Pseudo-second-order model (r^2>0.95) than the Pseudo-first-order model (r^2>0.90) due to the higher r^2 obtained. This indicates that the adsorption followed the Pseudo-second-order model. This model is based on the assumption that the rate-limiting stage can be described by chemical adsorption involving valence forces through sharing or exchange of electrons between the adsorbent and adsorbate [36]. The present result is consistent with Boujelben et al., who observed that the adsorption of Pb (II) onto manganese oxide-coated crushed bricks was fitted well by the Pseudo-second-order model [24]. The rapid metal uptakes was clearly related to the availability of active sites on the brick surfaces.

From Fig. 3 and the calculated parameters in Table 3 it can be seen that both the amount of Zn(II) adsorbed at equilibrium q_e and the sorption rate constant k_2 decrease following the order cinder bricks>lime sand bricks>gray clay bricks>red clay bricks, while the rate constants k_2

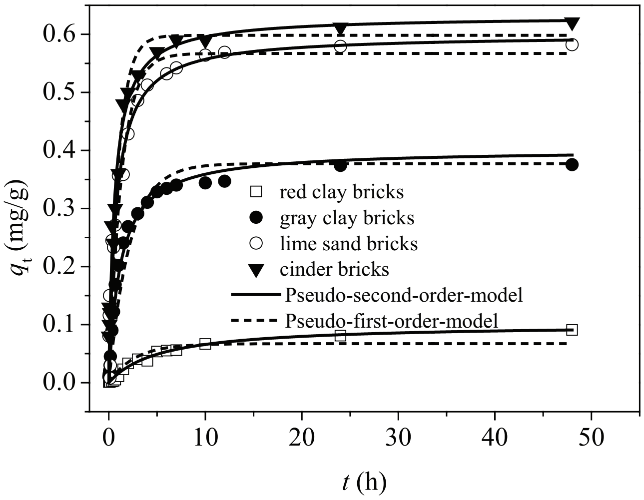

Fig. 3. Kinetics of Zn(II) removal by four types of bricks at 25°C (C_0 = 5mg /L, w = 8 g/L, stirring speed = 135 rpm, pH = 5).

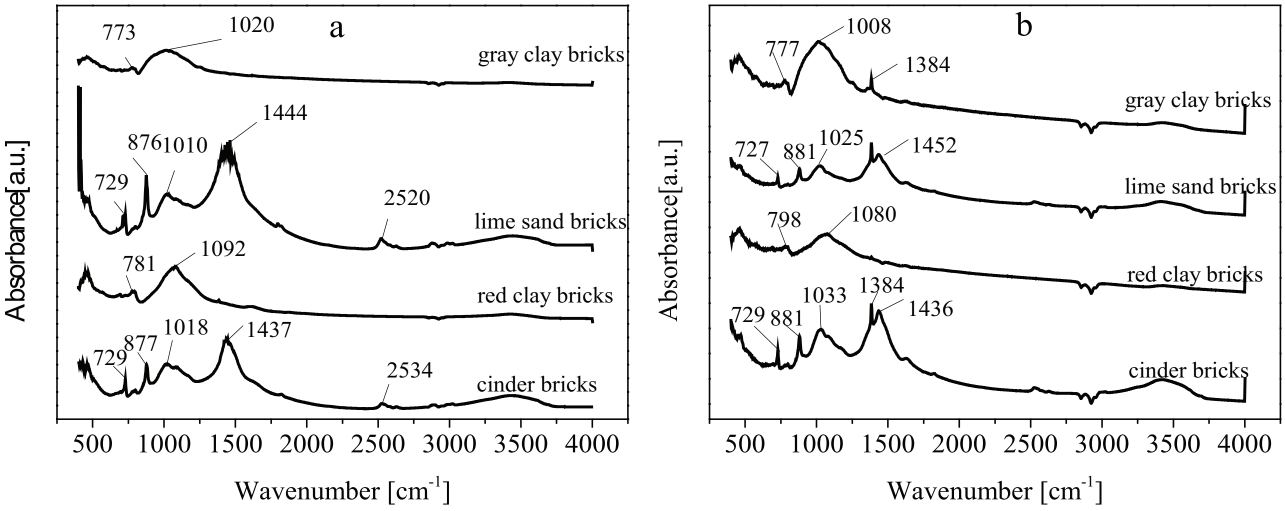

Fig. 2. FTIR spectra of the four types of bricks before (a) and after (b) adsorption to 5 mg/L Zn(II) for 48 h at pH 5.

Table 3
Parameters deduced by pseudo-first and pseudo-second order models for adsorption of Zn(II) by four types of bricks

	Pseudo-first-order			Pseudo-second-order		
	q_e (mg/g)	k_1 (1/min)	r^2	q_e (mg/g)	k_2 (g/mg·min)	r^2
red clay bricks	0.06±0.02	0.37±0.09	0.90	0.10±0.01	1.86±0.25	0.98
gray clay bricks	0.38±0.01	0.46±0.01	0.94	0.40±0.01	2.09±0.21	0.99
lime sand bricks	0.56±0.02	0.75±0.11	0.92	0.59±0.02	2.18±0.42	0.95
cinder bricks	0.59±0.01	1.01±0.10	0.94	0.63±0.01	2.63±0.37	0.97

Table 4
Parameters of Langmuir and Freundlich isotherms for adsorption of Zn(II) by four types of bricks

	Langmuir model			Freundlich model		
	q_m (mg/g)	K_L (L/mg)	r^2	K_F (mg/g)(L/mg)^n	n	r^2
Red clay bricks	0.31±0.02	0.12±0.01	0.99	0.03±0.01	1.54±0.05	0.98
Gray clay bricks	1.07±0.04	0.11±0.01	0.99	0.13±0.01	1.63±0.10	0.97
Lime sand bricks	2.78±0.09	0.22±0.02	0.98	0.69±0.03	2.30±0.19	0.96
Cinder bricks	3.67±0.18	0.19±0.03	0.99	0.68±0.05	1.85±0.10	0.98

decrease following the same order. The faster adsorption by the two types of non-clay-based bricks may be related to their larger pore volume. Table 2 shows that the pore volume of the four types of bricks decreases in the order: cinder bricks>lime sand bricks>gray clay bricks>red clay bricks, which is consistent with their sorption rate. The pore volume of red clay bricks (i.e.,0.0064 cm^3/g) is the smallest among the four types of bricks, thus they have the lowest adsorption rate constant. Boujelben et al. also observed that adsorption of Pb(II) is much faster on sand with larger pore volume [24]. The larger pore volume could allow the Zn(II) access to the sorption sites of the bricks much more easily and faster.

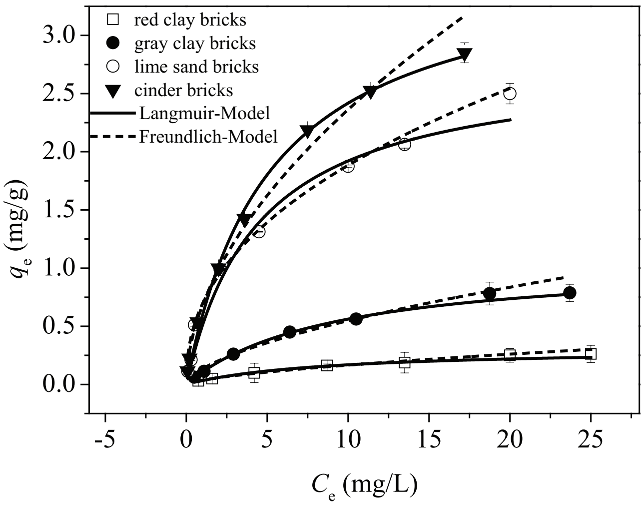

Fig. 4. Adsorption isotherms of Zn(II) by four types of bricks at 25°C (w = 8 g/L, stirring speed = 135 rpm, pH = 5).

3.3. Adsorption isotherms of Zn(II)on different types of bricks

Two of the most commonly used isotherm equations, namely Langmuir and Freundlich [39], were chosen to fit the adsorption isotherms of Zn(II) onto four types of bricks. The values of the deduced isotherm parameters are listed in Table 4 together with the r^2 of the fits.

Both the Langmuir and Freundlich models show good fits for Zn(II) adsorption (Fig. 4). To derive the maximum sorption capacities for the bricks,we chose the Langmuir model for ease of data comparison. Many of the previous studies used this model to describe the adsorption of heavy metals onto heterogeneous adsorbents such as bricks, bentonite [40], calcareous mudstone [6], red claystone [6] and zeolite [41]. The adsorption capacity and affinity of the four types of bricks towards Zn(II) were evaluated by analyzing the parameters q_m and K_L of the Langmuir equation (Table 4). The maximum adsorption capacities of the four types of bricks were between 0.31–3.67 mg/g. The adsorption capacities of the bricks for Zn(II) adsorption are slightly larger than some low-cost adsorbents, such as fly ash [42,43], natural zeolite [43] (1.7 mg/g) and ceramic tiles [3] whose capacities range from 0.4–1.7 mg/g, even though they are smaller than the traditional adsorbents such as activated carbon,with capacities ranging from 1.1–1594.0 mg/g [3,44,45]. As waste materials, bricks could still be applied as adsorbents or filling materials in some filtration devices to remove pollutants due to their large adsorption capacities and low cost. In the process of adsorption of zinc on bricks, both physical adsorption and ion exchange play a role. The thermodynamic results showed that ion exchange was the dominant factor. From Fig. 6 and Table 6 it is observed that the adsorption capacity increases with increasing temperature. This suggests that the adsorption driving force is mainly ion exchange. The driving force of ion exchange is favored by an increase in the temperature, as previously suggested [46].

Both the adsorption capacity and affinity decreased following the order of cinder bricks>lime sand bricks>gray clay bricks>red clay bricks (Table 4), which follows the same

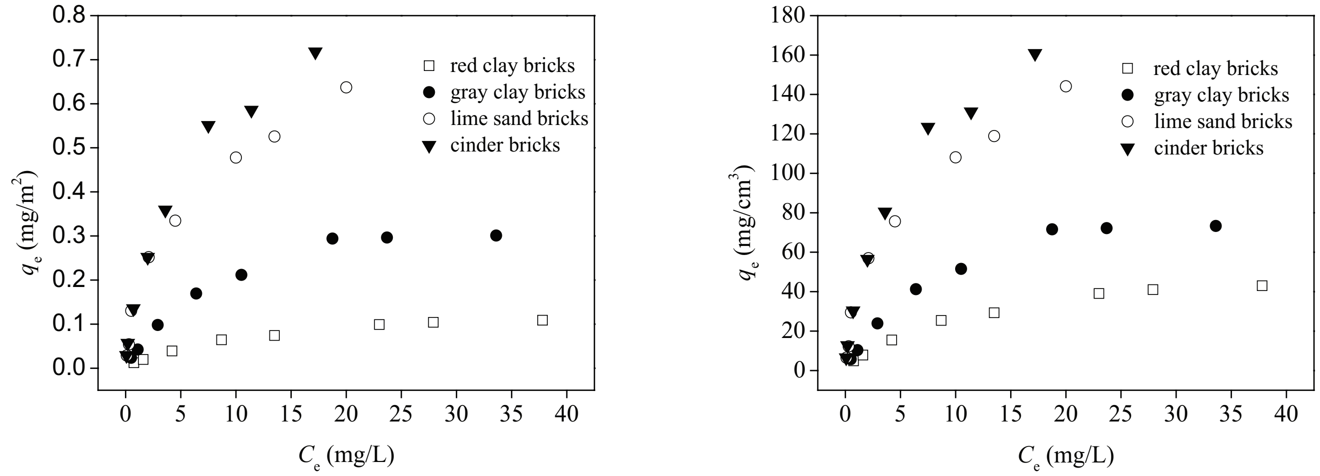

Fig. 5. The surface area-normalized and pore volume-normalized adsorption capacity of three types of bricks.

order as the surface area of the four types of bricks (Table 2). This result indicates that adsorption of Zn(II) is positively correlated with the surface area and micropore volume of the four types of bricks, which play a major role in adsorption. To exclude the role of these factors, the adsorption isotherms were normalized by surface area and micropore volume [31] (Fig. 5). As shown in Fig. 5, the surface area-normalized and micropore volume-normalized adsorption capacities of the four types of bricks still decreased in the following order: cinder bricks>lime sand bricks>gray clay bricks>red clay bricks. The greater adsorption of Zn(II) by cinder bricks and lime sand bricks could be also due to the nature of the bricks. Theoretically, adsorbents consisting of elements such as C, F and N could have strong adsorption toward heavy metals due to the formation of coordination bonds. From the elemental analysis, lime sand bricks and cinder bricks contained F, which could form coordination bonds with Zn(II), and thus have larger adsorption capacities than the two types of clay-based bricks without F content [42]. The higher amounts of exchangeable cations in lime sand bricks and cinder bricks were favorable for ion exchange. In addition, the FTIR spectra show that carboxyl and hydroxyl groups are present on lime sand bricks and cinder bricks. Those groups contribute to coordination of Zn(II) ions [31]. Furthermore, as shown in Table 5, the cation exchange capacity of the bricks follows the order of cinder bricks> lime sand bricks> gray clay brick>red clay bricks, consistent with the order of Zn(II) adsorption, which may also explain the different adsorption capacities of the different types of bricks.

Table 5
Cation exchange capacity of four types of bricks

Bricks	Cation-exchange capacity (mmol/g)
Red clay bricks	26.67
Gray clay bricks	33.33
Lime sand bricks	46.67
Cinder bricks	53.33

Table 6
Predicted isothermal constants for Zn(II) adsorption on bricks

Bricks	T (K)	ΔG (k J/mol)	ΔH (k J/mol)	ΔS (J/(mol ·K)
Red clay bricks	288	12.09	51.71	0.14
	298	9.29		
	318	7.70		
Gray clay bricks	288	7.38	23.72	0.058
	298	5.99		
	318	5.53		
Lime sand bricks	288	5.14	56.04	0.17
	298	0.0063		
	318	-0.0014		
Cinder bricks	288	5.47	57.67	0.18
	298	1.72		
	318	-0.32		

3.4. Effect of adsorbent dosage on removal efficiency

Adsorbent dosage is one of the important parameters in adsorption processes because it determines the capacity of an adsorbent for a given initial concentration of the adsorbate under a given set of operating conditions. Adsorbent loading is also significant in the design of adsorbents for optimization of the process [4]. At Zn(II) initial concentration of 5 mg/L and pH 5, the effect of three brick dosages (i.e., 1 g/L, 8 g/L, and 10 g/L) on Zn(II) adsorption was examined. As shown in Fig. 7, the removal efficiency of Zn(II) increased as brick dosages increased from 1 to 10 g/L. In detail, as brick dosages increased from 1 to 8 g/L, the removal efficiency for Zn(II) increased significantly from 27.98%–85.88%, 21.58%–81.84%, 6.91%–41.67%, 5.09%–15.82% for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. As the brick dosages increased from 8 to 10 g/L, the increase in the removal

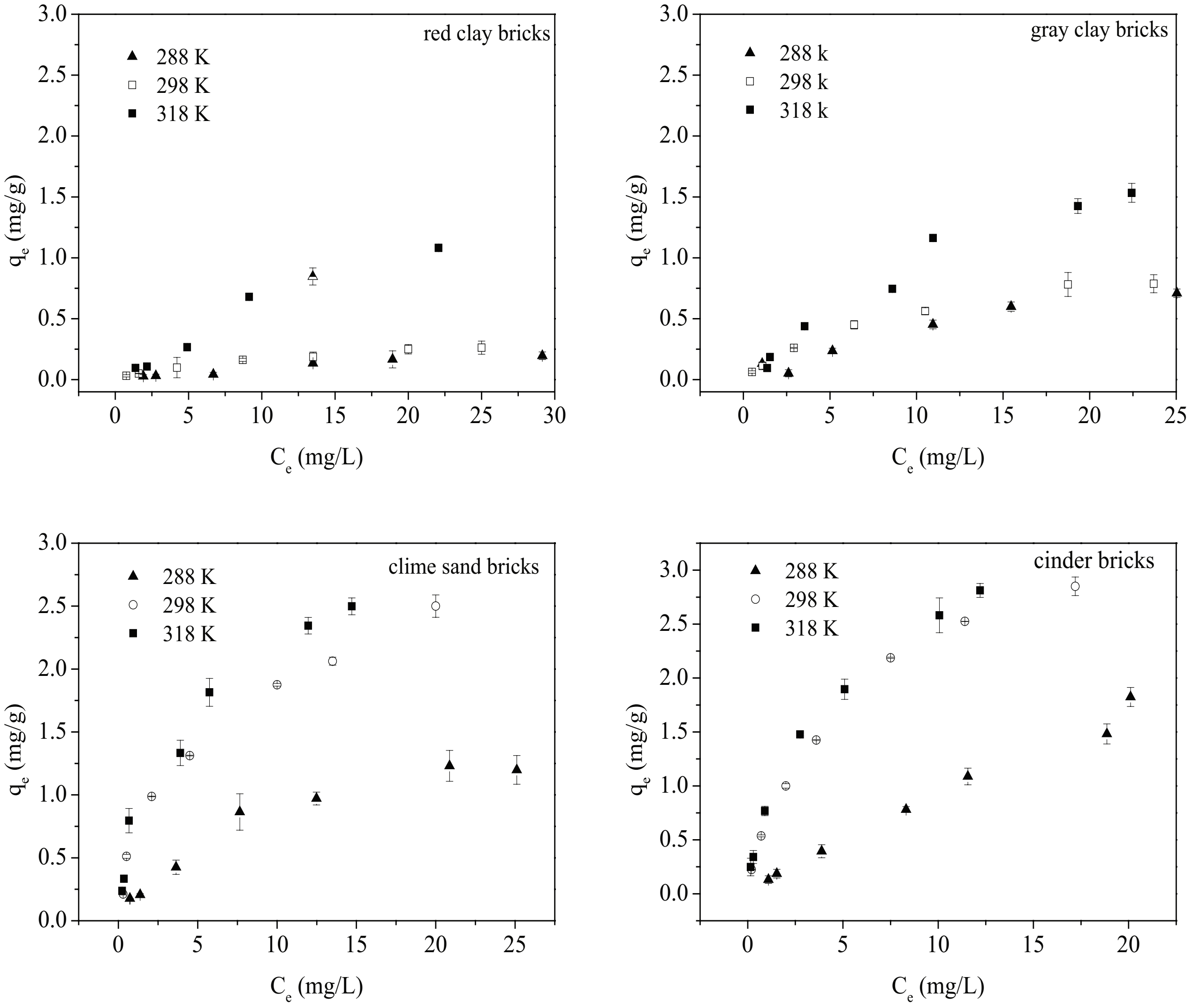

Fig. 6. Isotherms of Zn(II) adsorption on bricks at 288 K, 298 K and 318 K.

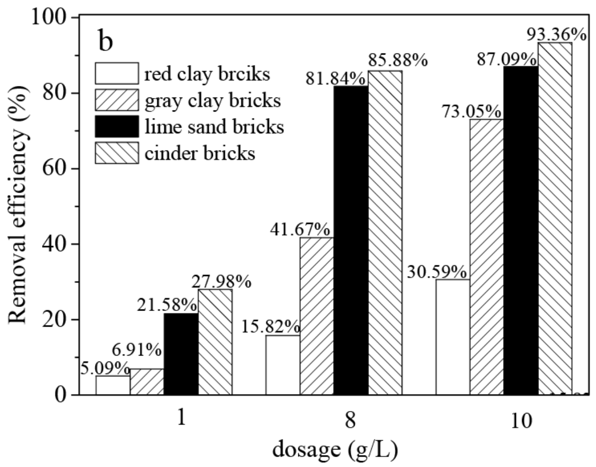

Fig. 7. Effect of brick dosage on the adsorption of Zn(II) from aqueous solution by four types of bricks at 25°C (C_0 = 5 mg /L, stirring speed = 135 rpm, pH = 5).

efficiency of Zn(II) was slight, as shown by the removal efficiency increasing from 81.84%–87.09%, 41.67%–73.05%, and 15.82%–30.59% for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. At one concentration studied (i.e., C_0 = 5 mg/L), the dosage of 8 g/L for cinder bricks and lime sand bricks could remove more than 80% of Zinc (II), so this was selected as the optimum dosage for Zn(II) removal. However, the two types of clay-based bricks showed lower removal efficiency at this dosage (i.e., 8 g/L) and the removal rate was below 41.67%, which indicated that the dosage of the two types of clay-based bricks should be increased in order to reach higher removal efficiency. At the dosage of 10 g/L, the removal rate of red clay bricks and gray clay bricks for Zn(II) reached 30.59% and 73.05%, respectively. Hoda et al. studied sorption of Zn(II) on clay-based bricks, and reported a 60% removal rate at a dosage of 10 g/L [3]. The present study and the study of Hoda et al. indicated that the removal efficiency of bricks for heavy metals could vary significantly even though they are made from similar raw materials, such as clay [3]. The differences observed for different types of bricks could be due to their different sources and production processes as well as their different characteristics, which could be reflected by differences in elemental composition, surface area, surface functionality etc, and as a consequence influence their sorption efficiency toward Zn(II).

3.5. Effect of solution pH on adsorption

The solution pH is one of the most important factors that controls the adsorption of metal ions [47]. The effect of solution pH on the adsorption of Zn(II) by the four types of bricks was determined under fixed conditions (C_0 = 5 mg /L, *w* = 8 g/L). Fig. 8 demonstrates that adsorption of Zn(II) is strongly pH dependent. It can be observed that the adsorption of Zn(II) increases with increasing pH in the range 2–6 for all types of bricks. The adsorption between brick surfaces and Zn(II) takes place through electrostatic attraction forces [47]. At low pH, there is excessive protonation of the adsorbent surface, resulting in a decrease in the adsorption of Zn(II) ions. Additionally, the H^+ ions can be considered as competing with Zn(II) for adsorption sites on the sorbent surface [48]. At relatively higher pH such as pH 6, the maximum adsorption was observed. The results could be explained by the formation of different ionic species and the increasing negative charge on the sorbent surface [3,49]. Generally, various Zn(II) species in aqueous solution are present in the form of Zn^{2+}, $Zn(OH)^+$, $Zn(OH)_2^0$, $Zn(OH)_3^-$ and $Zn(OH)_4^{2-}$as a function of pH [4,50]. At pH < 6.0, the predominant Zn(II) species is Zn^{2+} and the removal of Zn(II) is mainly accomplished by an adsorption reaction. Low removal efficiency is observed in the low pH range, which may be related to the fact that there are more protons in an acidic environment available to protonate the active sites on the brick surface, and competition between zinc ion and H^+ occurs. At higher pH values, the lower concentration of H^+ and greater number of ligands with negative charge leads to greater zinc adsorption [50]. The zeta potential results proved this analysis. As shown in Fig. 9, with the increase of pH, the zeta potential of the four types of bricks gradually decreased. In fact, at pH 2–11, the zeta potential of all the types of bricks is negative. With the increase of pH, the zeta potential decreases, and the surface of the brick has more negative charge, so the electrostatic effect between the brick surface and Zn(II) is enhanced. This is beneficial to the adsorption of Zn(II) on bricks. Higher adsorption of heavy metals onto a heterogeneous sorbent at higher pH was also observed in previous studies. Hoda et al. observed that the Zn(II) removal efficiency by machine-made bricks increases with the increase of pH from 2.5 to 6.5, and the optimum condition for Zn(II) removal is at pH 6–7 [3]. Besides bricks, higher adsorption of heavy metals such as Zn(II), Cu(II), Pb(II) at higher pH was also observed on adsorbents such as raw and base-modified Eucalyptus sheathiana bark, graphene oxide and red paddy soil [4,50,51].

Even though all types of bricks had higher adsorption capacity for Zn(II) at higher pH, the effect of pH on adsorption was slightly different for the different types of bricks. In detail, with the increase of the solution pH from 2–6, the maximum adsorption capacities increased from 0.39–0.59 mg/g, 0.32–0.46 mg/g, 0.17–0.39 mg/g and 0.016–0.31 mg/g for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. The adsorption by the two types of clay-based bricks (adsorption capacity increased by 19.37-fold and 2.29-fold for red and gray clay bricks from pH 2–6, respectively) was more sensitive to the influence of pH than for the two non-clay-based bricks (adsorption increased by 1.43-fold and 1.51-fold for lime sand bricks and cinder bricks from pH 2–6, respectively). The greater pH sensitivity of the two types of clay-based bricks may be due to the faster decrease in zeta potential (more surface negative charges) observed for the clay-based bricks, especially the red clay bricks. As the pH increases, the electrostatic attraction as the driving force between positive zinc ions and the negative surface of clay-based bricks may be enhanced more than for the non-clay-based bricks with less negative charge, leading to more significant changes as a function of pH [50].

3.6. Effect of ionic strength

The effect of ionic strength (0–200 mg/L) on Zn(II) uptake by the four types of bricks was studied (Fig. 10).

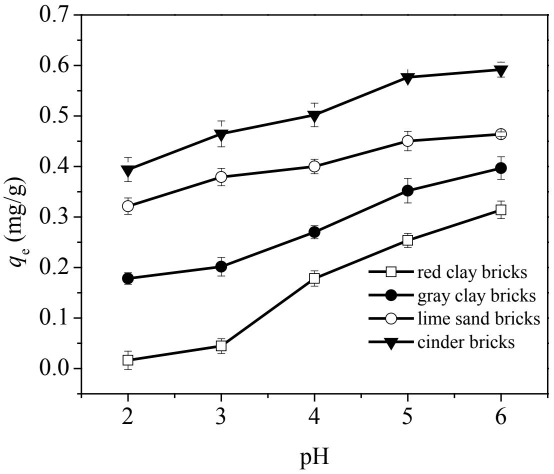

Fig. 8. Effect of pH on the adsorption of Zn(II) from aqueous solution by four types of bricks at 25°C (C_0 =5 mg /L, *w* = 8 g/L, stirring speed = 135 rpm).

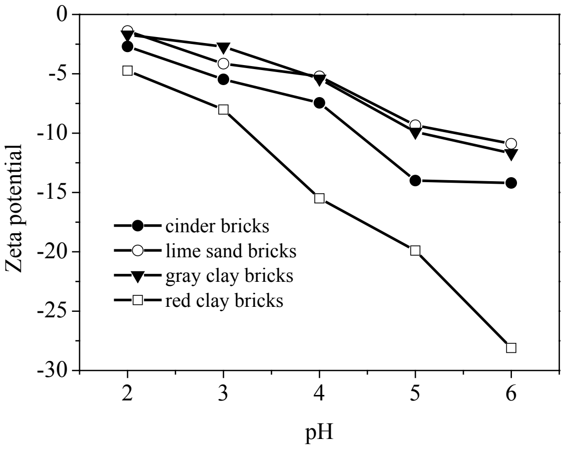

Fig. 9. Zeta potential of four types bricks in different pH.

The effect of ionic strength was similar for all types of bricks. The results indicate that at lower sodium chloride concentrations (i.e., 0–1 mg/L), the adsorption of Zn(II) by all bricks decreased with increasing salt concentration, with adsorption capacity decreased from 0.58–0.51 mg/g, 0.50–0.43 mg/g, 0.28–0.22 mg/g and 0.10–0.08 mg/g for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. With the increase of NaCl concentration from 1–10 mg/L, the adsorption of Zn(II) increased rapidly from 0.51–0.57 mg/g, 0.43–0.51 mg/g, 0.22–0.28 mg/g and 0.08–0.12 mg/g for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. With the further increase of ionic strength beyond 10 mg/L, the adsorption of Zn(II) reached a plateau for all types of bricks. Ion exchange was suggested to be the main mechanism in the adsorption process. The initial depression of Zn(II) adsorption by ionic strength might be due to the competition of Na^+ ions with Zn cations for the same binding sites on the sorbent surface [4,47,52]. The adsorption of Zn(II) was then increased with the increase of salt concentration, which can be explained by the fact that sodium chloride acts as an in situ regenerating agent for the sorbent via removal of oxygenated complexes as soluble chloro complexes, increasing thereby the number of adsorption sites and hence the adsorption of Zn(II) species [47]. Moreover, with continuing increase in the ionic strength, the electrostatic double layer of the adsorbent is more compressed, leading to more adsorption of metal ions [47]. However, further increase of ionic strength from 10–200 mg/L did not significantly influence the adsorption on the adsorbents, which may be due to the limitation of the ability of NaCl to regenerate the active sites. In this range of ionic strength, the increasing and decreasing effects of NaCl on the adsorption capacity reached a balance. Therefore, at higher NaCl concentrations greater than 10 mg/L, ionic strength has little effect on adsorption. Djeribi and Hamdaoui reported a similar effect of ionic strength on the sorption of Cu(II) onto crushed bricks. They reported a critical concentration of 5 g/L NaCl and observed decreasing and increasing of adsorption before and after this critical concentration [47]. In the present study, the critical concentration of NaCl is 1 mg/L which is far less than the value Djeribi and Hamdaoui observed [47]. The difference between their and our study may be due to differences in the types of bricks and heavy metals, so that the regenerating effect of Na^+ occurs at a different concentration.

3.7. Effect of brick particle size

Differences in particle size could lead to different surface areas, and thus may influence the adsorption of Zn(II) due to the occupation of adsorption sites on the sorbent surface. For better application of the bricks in remediation, examination of the effect of the size of brick particles on adsorption could provide information on optimal size selection [3,53]. The effect of four size ranges (i.e., 0.15–0.5 mm, 0.5–1 mm, 1–2 mm and 2–5 mm) on Zn(II) adsorption is shown in Fig. 11. As shown in Fig. 8, with the increase in particle size, adsorption of Zn(II) by all types of bricks decreased sharply with the increase in particle size from 0.15–0.5 mm to 1–2 mm. From then on, adsorption did not further decrease when the brick particle size increased from to 1–2 mm and 2–5 mm. The results show that brick particles provide better metal uptake at finer sizes. This is likely owing to the increased external surface area per unit mass and reduced internal diffusion resistance for smaller particles[27]. Generally, smaller particles present larger surface area and better adsorption performance, so the adsorption efficiency will be higher [27]. With the decrease of brick particle size from 2–5 mm to 0.15–0.5 mm, the removal rate increased 42.74%, 46.91%, 42.99% and 36.75% for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. Smaller particles have higher adsorption capacities; however, to produce finer particles, the cost will be increased due to the crushing procedure. In addition, bricks maybe applied in infiltration devices to treat contaminants in storm water. Smaller particle size with lower hydraulic loading shock resistance ability may lead to flooding. Bricks with larger particle size in filtration

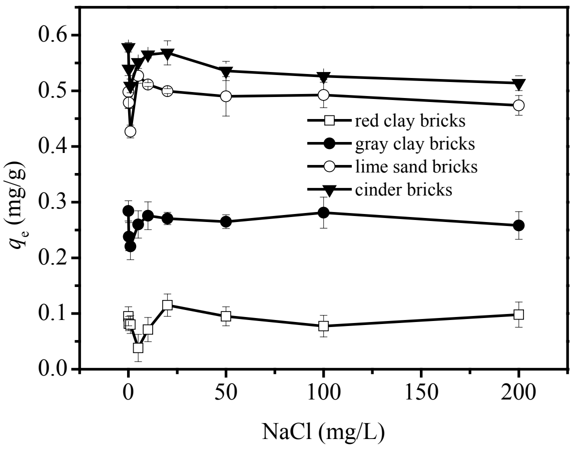

Fig. 10. Effect of ionic strength (NaCl) on the adsorption of zinc from aqueous solution by four types of bricks at 25°C (C_0 =5 mg /L, w = 8 g/L, stirring speed = 135 rpm, pH = 5).

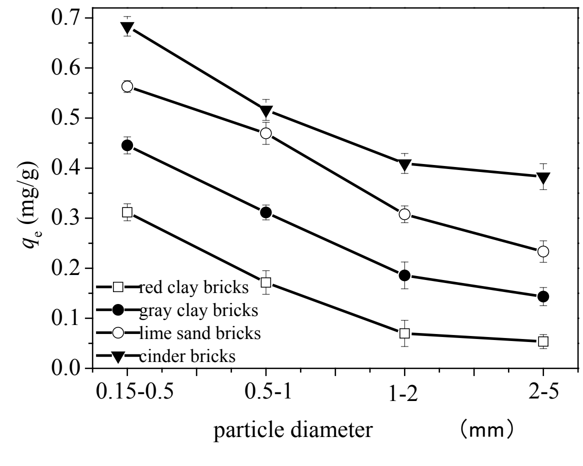

Fig. 11. Effect of particle size on the adsorption of Zn(II) from aqueous solution by four types of bricks at 25°C (C_0 =5 mg /L, w = 8 g/L, stirring speed = 135 rpm, pH = 5).

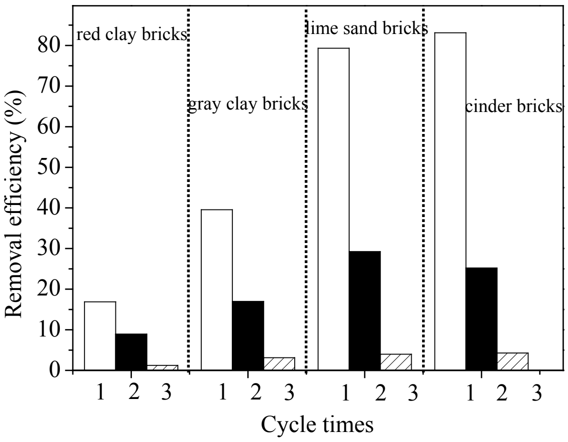

Fig. 12. Cycling experiment of four types of bricks to adsorb Zn(II).

devices will reduce the runoff volume, and the peak flow may be more appropriate [54]. Therefore, once applied in the field of remediation, one could select a proper size distribution of brick particles according to the real situation.

3.8. Regeneration experiment for bricks

It is of great value if an adsorbent is easily regenerated. In order to evaluate the regeneration performance of the four kinds of bricks, we carried out a cycling experiment. In this experiment, 1 mol/L HCl was used to regenerate the adsorbent materials over five cycles. The regeneration time was 12 h and the dosage was 8 g/L. The Zn(II) concentration was 5 mg/L, and the adsorption temperature was 25°C. The results after three cycles are shown in Fig 12. After three cycles, the removal rate of Zn(II) decreased rapidly from 83.12%–4.29%, 79.32%–3.99%, 39.55–3.12%, 16.89%–1.25% for cinder bricks, lime sand bricks, gray clay bricks and red clay bricks, respectively. Although the waste bricks still have a certain removal effect toward Zn(II), considering the cost of regeneration and that the bricks are a kind of waste, recycling of spent brick adsorbent was abandoned.

4. Conclusions

The adsorption of Zn(II)by four types of bricks including red clay bricks, gray clay bricks, lime sand bricks and cinder bricks was systematically investigated and compared under different experimental conditions. The adsorption behavior toward Zn(II) varied for the different types of bricks in terms of solution chemistry parameters such as pH and ionic strength, and the characteristics of bricks including morphology, specific surface area, surface functionality and elemental composition. The main conclusions could be drawn as follows:

1) The adsorption kinetic study showed that the adsorption rate follows the order of cinder bricks>lime sand bricks>gray clay bricks>red clay bricks, and the difference is related to the pore volume of the bricks. The adsorption kinetic data was fit best by the Pseudo-second-order model for the four types of bricks, suggesting that the adsorption of Zn(II) is controlled by a chemical adsorption process.
2) Consistent with the adsorption rate, the maximum adsorption capacity of bricks followed the order of cinder bricks > lime sand bricks > gray clay bricks > red clay bricks, and Langmuir model is most suitable to describe adsorption process. The maximum metal adsorption was strongly influenced by the total specific surface area, as well as the presence of functional groups on the bricks.
3) The removal efficiencies of the two non-clay-based bricks (i.e., cinder bricks, lime sand bricks) were much larger than those of the two clay-based bricks (i.e., red clay bricks, gray clay bricks) and could reach>80% for Zn(II) removal at the dosage of 8 g/L. The removal efficiency increased with increasing dosage of bricks.
4) The adsorption of Zn(II) increased with increasing pH from 2–6 for all types of bricks. The adsorption of the two types of clay-based bricks was more sensitive to the influence of pH than the two types of non-clay based bricks.
5) In the lower NaCl range from 0–10 mg/L, the adsorption decreased and then increased. With further increase of the ionic strength to 200 mg/L of NaCl, the adsorption capacities of all types of bricks showed almost no change. The effect of ionic strength on the adsorption of Zn(II) by the four types of bricks was similar.
6) Adsorption of Zn(II) by all types of bricks decreased with increasing particle size. The decrease was sharp from the size range from 0.15–0.5 mm to 1–2 mm, while the adsorption did not further decrease when the brick particle size increased to 2–5 mm.

Combining adsorption data with the systematic characterization of bricks, the present study shows that both the solution chemistry and the nature of bricks have significant influence on the sorption of Zn(II). The results of this study provide basic data and a reference for the application of waste brick in the remediation field.

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