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Biochar prepared by fish feces residue: properties and its application for removing antibiotics in aquaculture tail water

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ABSTRACT

Fish feces and residues are a kind of common waste in aquaculture system to require timely handling while aquaculture wastewater generally contains antibiotics to require efficient treatment. This study prepared biochar by using fish feces and residues to investigate the removal of typical fluoroquinolone enrofloxacin by biochar. Biochar showed the sheet-dot mixed structure with multiple elements on surface. The content of carbon/nitrogen/hydrogen of biochar made of fish feces and residues reached 46.5%/3.1%/2.5%. The removal efficiency of enrofloxacin reached 94.7% with biochar dosage of 1 g/L. The adsorption capacity of biochar at pH of 2 or 10 was higher than that at the other pH values. Adsorption capacity of biochar increased by 10.6% from 15°C to 25°C to reach 9.47 mg/g at 25°C while that slightly increased from 25°C to 35°C. Pseudo-first-order model was the best model with $R^2 = 0.9982$ for describing adsorption kinetics of enrofloxacin by biochar. Langmuir model could better fit results of isotherm for enrofloxacin by biochar with Langmuir constant of 0.38–1.65 L/mg. Biochar used 3–4 times still kept good removal efficiency for enrofloxacin. The prepared biochar exhibited good application potential. The findings of this study provide new idea for treating aquaculture solid wastes and wastewater.

Keywords: Aquaculture; Antibiotics; Biochar; Adsorptive removal; Fish feces and residues

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1. Introduction

Aquaculture has served as the important industry in coastal zone to provide plenty of proteins for human beings. However, aquaculture tail water is also important source of various pollutants such as antibiotics and antibiotic resistance genes (ARGs) in coastal water [1–3]. Antibiotics have been widely used as medicine for human beings and growth promoters for animal husbandry. Overuse or abuse antibiotics could induce antibiotic pollution and ARG pollution [4]. Antibiotics generally exist in the natural environment with trace concentrations while they could exist in the tail water with higher concentrations. Therefore, removal of antibiotics in aquatic environment needs deep investigation.

Many techniques have been used for removal of antibiotics in different matrices. Anammox process was used to remove antibiotics in wastewater [5]. Other materials such as carbon nanomaterials and metal oxides have been used to remove antibiotics [6]. Moreover, fungi and microalgae have also been used to remove antibiotics in wastewater [7]. Oxidization methods including catalytic ozonation and

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Fenton process have been regarded as the good techniques for removing antibiotics in water [8–10]. Nanoparticles have been widely used as catalyst for oxidation of different pollutants such as benzyl alcohol [11,12]. S and N co-doped carbon nanotubes were synthesized for effective desulfurization [13]. Some other nanoparticles possessed the ability for treating groundwater or soil [14,15]. Some nanoparticles showed good photocatalytic performance or good feature for monitoring [16–18].

Although these methods possess good performance for removing target antibiotics, relatively high cost and low removal efficiency might still hinder the wide application of them. Preparation of some materials often take long time and high cost so that it is not feasible to use these materials for removing antibiotics from the aquatic environment. Some other materials might possess low removal capacity. Therefore, it is necessary to find suitable material with low cost and good capacity to remove antibiotics. Biochar which is prepared by bio-wastes under anoxia or anaerobic conditions has been widely used for wastewater treatment including antibiotics-polluted water treatment [7–20]. Hydrothermal and pyrolysis are two frequently-used techniques for preparing biochar [19–21]. Hydrothermal method has shown the advantages of simple process and relatively low cost in comparison with pyrolysis technique to be often used to prepare biochar in recent years [19–22].

Enrofloxacin is a common fluoroquinolone used in aquaculture especially in fish-farming [23]. Enrofloxacin has been frequently detected in different matrices and occurred in wastewater with relatively high concentrations [24]. It is important to look for feasible, efficient, and cheap technique to get rid of enrofloxacin in wastewater including aquaculture tail water. Moreover, fish feces and residues are a kind of common solid waste for aquaculture. The recycle and reuse of fish feces and residue is a challenge for aquaculture. Therefore, this study synthesized new biochar using fish feces and residues in an aquaculture system. Effect of different factors including biochar dosage, pH, temperature, and initial concentration on removal of enrofloxacin by biochar was investigated while reuse of biochar was also explored. The kinetics and thermodynamics were discussed. The final goal is to obtain the important information on disposal of fish feces and residue as well as antibiotic pollution control.

2. Materials and methods

2.1. Chemicals and reagents

Enrofloxacin with purity of 98% was purchased from Shanghai Aladdin Biochemical Technology Co., Ltd., China. Enrofloxacin was solved in methanol to get stock solution with concentration of 100 mg/L which was stored at –20°C while working solutions with different concentrations were prepared by diluting the stock solution using methanol before the experiment. Methanol and acetonitrile were HPLC grade and obtained from MREDA Company (Beijing, China).

Fish feces and residues were collected from the fish tank of tilapia cultivation system. Kimwipes were used to get rid of surface water of fish feces and residues. Hydrothermal method was used to make biochar using fish feces and residues. The source materials (fish feces and residues) with wet weight of 10 g were placed into hydrothermal reactor with volume of 25 mL under 190°C for 6 h. The prepared biochar was dried at 105°C for 12 h before use.

2.2. Biochar characterization by instrument

C, H, and N content of biochar was measured by a CHN analyzer (Elementar Vario Micro Cube, Germany). The pH of biochar was regarded as the pH of supernatant of biochar vs. water with 2.5:1. Surface morphology of biochar was measured by scanning electron microscope (SEM; S-4800, Hitachi, Japan). Energy-dispersive X-ray spectroscopy (EDS) was used to determine the elements on surface of biochar. X-ray diffraction (XRD) was used for possible phase identification. Fourier-transform infrared (FTIR) spectrometer (Thermo Nicolet iS50, Thermo Fisher, USA) was used to investigate the FTIR spectrum of biochar.

2.3. Adsorption of enrofloxacin by biochar

All assay were performed in triplicate. The effect of biochar dosage on adsorption of enrofloxacin was investigated under 25°C, initial concentration of 10 mg/L, pH of 7, shaking rate of 150 rpm for 24 h, and dosages of 0, 0.05, 0.1, 0.2, 0.5, and 1.0 g/L. The effect of pH on adsorption of enrofloxacin was investigated under 25°C, initial concentration of 10 mg/L, dosage of 1.0 g/L, shaking rate of 150 rpm for 24 h, and pH of 2, 4, 6, 7, 8, and 10. The effect of initial concentration on adsorption of enrofloxacin was investigated under 25°C, pH of 7, dosage of 1.0 g/L, shaking rate of 150 rpm for 24 h, and initial concentration of 1, 5, 10, 20, and 50 mg/L. The effect of temperature on adsorption of enrofloxacin was investigated under pH of 7, biochar dosage of 1.0 g/L, initial concentration of 10 mg/L, shaking rate of 150 rpm for 24 h, and temperature of 15°C, 25°C, and 35°C.

The biochar adsorbing the enrofloxacin at the equilibrium with weight of 50 mg was placed in the beaker containing the de-ionized water with volume of 50 mL which was place in the ultrasonic cleaner (50–60 Hz) at 25°C for 30 min to desorb the pollutant. The used biochar was placed in the beaker for adsorption test. Three replicates were used to make sure the data quality.

Adsorptive kinetics were explored by using pseudo-first-order, pseudo-second-order, and Elovich models while adsorptive isotherm was discussed by using Langmuir and Freundlich models [11].

The pseudo-first-order model is as follows:

$$\ln\left(q_e - q_t\right) = \ln q_e - k_1 t \tag{1}$$

where q_e is the saturated adsorption capacity; t is the time; q_t is the adsorption capacity at t; k_1 is the rate constant of pseudo-first-order model.

The pseudo-second-order model is as follows:

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

where q_e is the saturated adsorption capacity; t is the time; q_t is the adsorption capacity at t; k^2 is the rate constant of pseudo-second-order model.

The Elovich model is as follows:

$$q_t = a + b\ln t \tag{3}$$

where t is the time; q_t is the adsorption capacity at t; a and b are the rate constant of Elovich model.

The Langmuir model is as follows:

$$\frac{1}{q_{\text{eq}}} = \frac{1}{b_S Q_m C_{\text{eq}}} + \frac{1}{Q_m} \tag{4}$$

where q_{eq} is the adsorption capacity of 1 g adsorbent; C_{eq} is the equilibrium concentration; Q_m is the adsorption capacity of mono-layer; b_S is surface adsorption constant.

The Freundlich model is as follows:

$$\ln q_{\text{eq}} = \ln k_F + \frac{\ln C_{\text{eq}}}{n} \tag{5}$$

where q_{eq} is the adsorption capacity of 1 g adsorbent; C_{eq} is the equilibrium concentration; k_F is the Freundlich parameter; n is Freundlich experimental constant.

After adsorptive experiment, biochar was collected by 5 min centrifugation at 12,000 rpm while the supernatant was collected for UPLC (ultra-performance liquid chromatography) analysis. The collected biochar was used for the adsorption experiment again to conduct the recycle experiment. The biochar was recycled 6 times to investigate its sustainability of treating wastewater.

ACQUITY UPLC H-Class (Waters Corporation, Milford, USA) was used to determine the concentrations of enrofloxacin. A reversed-phase Waters C-18 column (2.1 mm × 50 mm, 1.7 μm) was used for chromatography separation with column temperature of 40°C. Mobile phase consisted of mixture made by 0.1 M oxalic acid aqueous solution: acetonitrile: methanol with volume ratio of 80:10:10. The injection volume was 10 μL and flow rate was controlled at 0.2 mL/min. Enrofloxacin was detected at 278 nm.

3. Results and discussion

3.1. Characteristics of biochar made of fish feces and residues

The content of carbon, nitrogen, and hydrogen of biochar made of fish feces and residues reached 46.5%, 3.1%, and 2.5%, illustrating that the synthesized biochar was a kind of nitrogen doped material. The pH of synthesized biochar was 5.8, which was similar to that of hydrothermal biochar. The SEM analysis showed that the biochar made of fish feces and residues possessed the amorphous appearance with mixture of dot and sheet structure (Fig. 1a). Aggregated structure of biochar could often occur during the hydrothermal preparation [20]. EDS analysis on the synthesized biochar showed that multiple elements such as C, O, Mg, Al, Si, P, and S were the major elements on surface of biochar (Fig. 1b). Carbon and oxygen accounted for 94% of surface element weigh. XRD spectrum showed that the synthesized biochar was amorphous (Fig. 1c). FTIR analysis exhibited that C–O–C (1,060 cm^{-1}), C–H (2,922 cm^{-1}), C=C or N–H (1,652 cm^{-1}), and other functional groups might exist in the synthesized biochar (Fig. 1d). Previous study [13] also reported that C–H group had the peak at 2,919 cm^{-1} which was identical to that this study observed.

3.2. Effect of different factors on adsorption of enrofloxacin on biochar

Adsorptive removal of enrofloxacin by biochar made with fish feces and residues was influenced by different factors (Fig. 2). Biochar dosage could significantly affect the removal of pollutant because more biochar would provide more adsorption sites for the pollutant [25]. Removal efficiency significantly increased with higher biochar dosage and 1 g/L of biochar addition could made 94.7% of enrofloxacin be removed (Fig. 2a). Wastewater treatment cost is in close relationship with adsorbent dosage so that more addition amount of adsorbent will significantly increase the treatment expenditure. Therefore, reasonable dosage of this study should be carefully evaluated to be 1 g/L. The dosage used by this study was much less than that of other studies [26,27], illustrating that the synthesized biochar with fish feces and residues was a prospective adsorbent for treating antibiotics.

The effect of pH on removal of enrofloxacin by biochar made with fish feces and residues showed different stages (Fig. 2b). The adsorption capacity of biochar with pH of 2 or 10 was higher than that with the other pHs, which was completely different from the previous study [13]. The adsorption capacity of biochar decreased with increasing pH when pH was lower than 4 while that increased with higher pH when pH was higher than 4. The lowest adsorption capacity of biochar at pH of 4 was only 37.1% of that at pH of 10. Enrofloxacin is a kind of fluoroquinolone antibiotics with different speciation forms at variable pH [28]. Therefore, adsorption of enrofloxacin was dependent on pH. The cationic enrofloxacin mainly exists when pH is lower than pKa_1 which is 6.19 while the anionic enrofloxacin mainly exists in solution when pH is higher than pKa_2 which is 7.75 [28]. Enrofloxacin mainly exists with both cationic and anionic form with pH ranges between pKa_1 and pKa_2. Enrofloxacin only exists with cationic form when pH is lower than 4.0 while that only exists with anionic form when pH is higher than 9.5 [28]. Adsorption of enrofloxacin on biochar was the lowest at pH of 4 and the optimal pH could be 7–10.

Temperature could affect the reaction rate for different processes including adsorption [29,30]. Adsorption capacity of biochar increased by 10.6% from 15°C to 25°C to reach 9.47 mg/g at 25°C while that slightly increased from 25°C to 35°C (Fig. 2c). The optimal temperature for removal of enrofloxacin was 35°C. Aquaculture wastewater could retain 25°C–35°C, exhibiting that biochar made with fish feces and residues might be useful for removing antibiotics in wastewater.

Initial concentrations of pollutants could significantly affect the removal efficiency of adsorption [28–30]. Initial

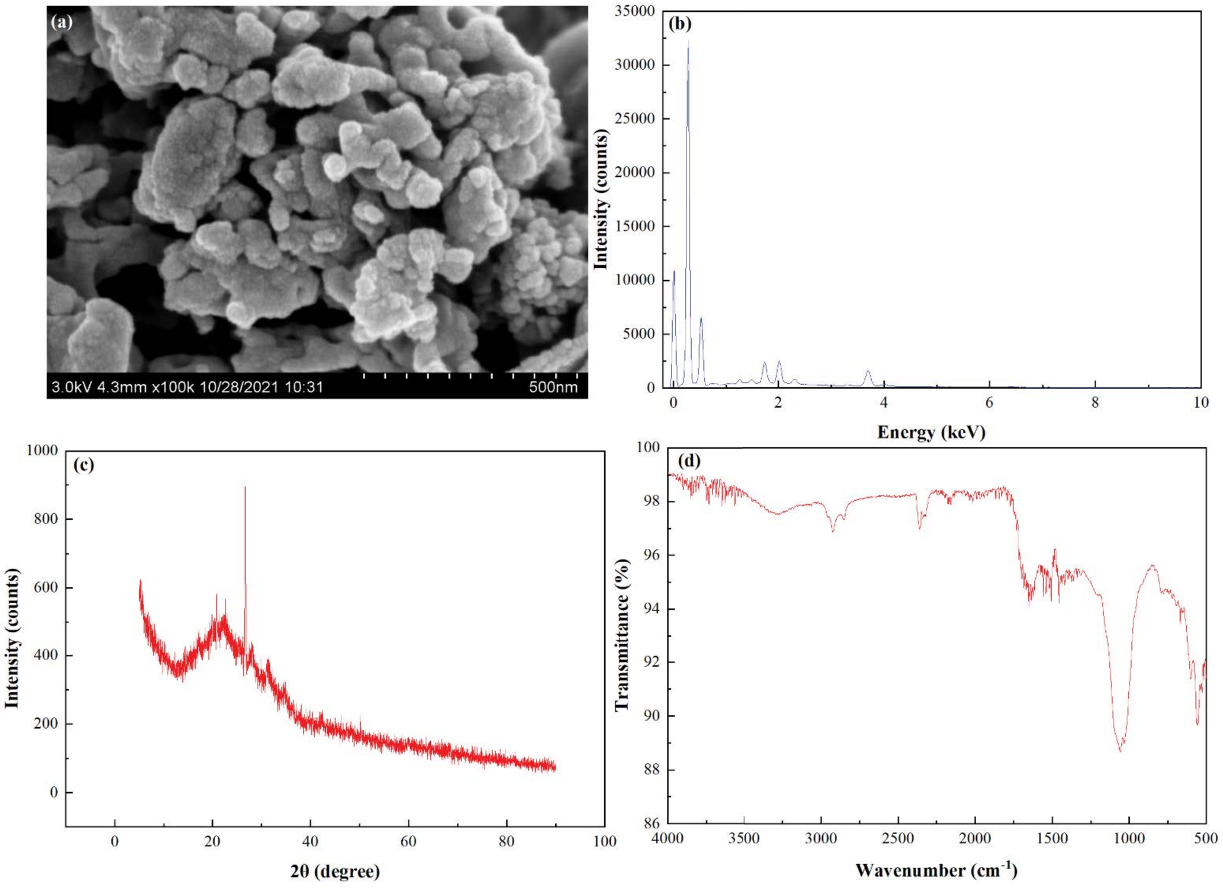

Fig. 1. SEM (a), EDS (b), XRD (c), and FTIR (d) analysis of synthesized biochar.

enrofloxacin concentrations also influenced the adsorption capacity of biochar (Fig. 2d). Adsorption capacity increased with the initial concentration of enrofloxacin. The removal rate could reach 96.7% after 24-h treatment with initial concentration of 1 mg/L while that was only 60% with initial concentration of 50 mg/L. Higher initial enrofloxacin concentration meant removal of pollutant needed more adsorption sites. Therefore, the removal efficiency would decrease with higher concentration due to limited adsorbent amount. More addition of adsorbent suggested higher treatment cost so that reasonable addition amount of adsorbent should be carefully evaluated. The concentration of antibiotics in wastewater was generally ng/L, μg/L, even mg/L so that 10 mg/L of initial concentration might be the extreme for wastewater. Therefore, the current biochar usage was reasonable.

3.3. Reuse of biochar made with fish feces and residues

Wastewater treatment cost is both dependent on the adsorbent amount and adsorbent reuse. Adsorbent which could be reused for more times will significantly decrease the treatment cost and enhance the application of adsorbent. The biochar was recycled 6 times for adsorption of enrofloxacin. The results showed that the prepared biochar could keep good adsorption capacity after at least 3–4 times (Fig. 3). The removal rate of enrofloxacin by biochar used 2/3/4 times was 95%/86%/77% of that of virgin biochar. It was interesting that removal rate of enrofloxacin by biochar used for 5 or 6 times significantly decreased. Some adsorption sites might be occupied after multiple usage so that the removal efficiency would continuously decrease with more reuse. Moreover, the surface area of biochar quickly decreased after it was used for 5–6 times so that the adsorption efficiency of used biochar significantly reduced. It was satisfactory that the biochar prepared with fish feces and residues possessed comparable reusability with those previously reported to exhibit good application potential [31,32]. Biochar preparation also provided new pathway for treating fish feces and residue of aquaculture. Heavy metals in aquaculture might exert potential risks [33]. Therefore, biochar made of fish feces and residues might also have good removal performance for removing heavy metals, which needs further investigation in the future.

3.4. Adsorption kinetics of enrofloxacin on biochar made with fish feces and residues

Three approaches including pseudo-first-order, pseudo-second-order, and Elovich models were used to explore adsorption kinetics of enrofloxacin on biochar made with fish feces and residues (Fig. 4). It was interesting that fitting result of pseudo-first-order model was the best among 3 models with R^2 = 0.9982. The fitting

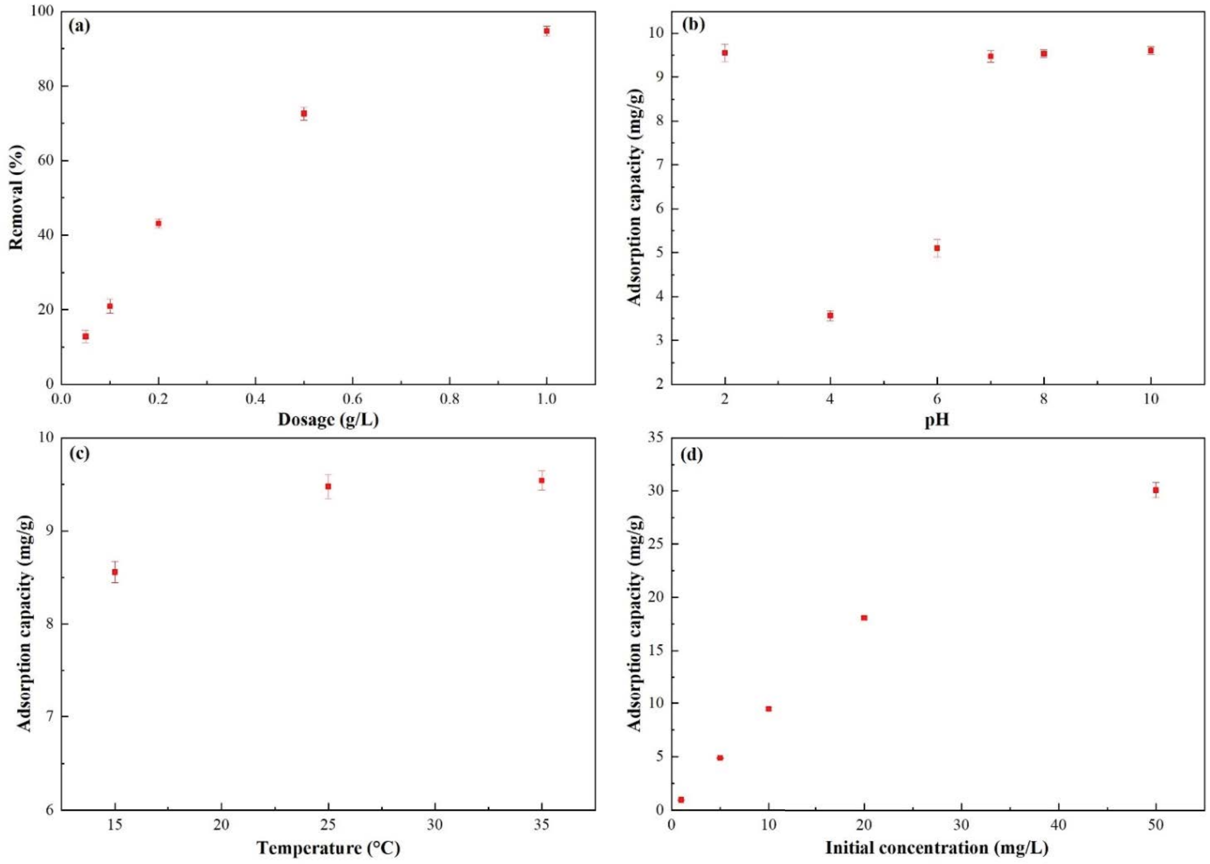

Fig. 2. Effect of different factors including biochar dosage (a), pH (b), temperature (c), and initial concentration (d) on removal of enrofloxacin by biochar prepared with fish feces and residues.

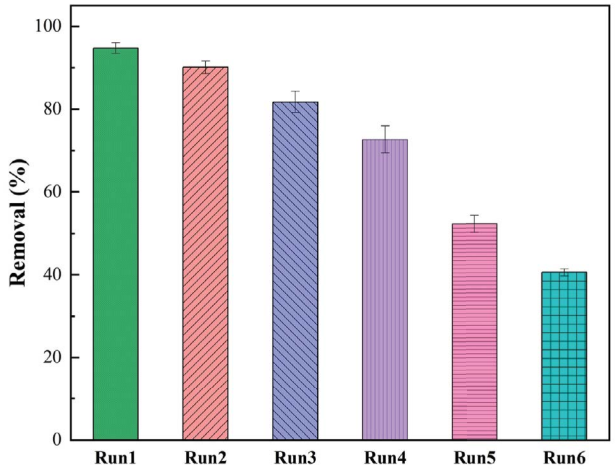

Fig. 3. Removal efficiency of biochar recycled for 6 times.

result of pseudo-second-order model for enrofloxacin on biochar with R^2 = 0.9308 was slightly worse than that of pseudo-first-order but much better than that of Elovich model (R^2 = 0.7971). The Elovich rate constants including a and b were calculated as 7.53 and 0.71, respectively. The Elovich model could generally describe the effect of chemical reactions on pollutant adsorption behaviors on adsorbent [34]. Low regression coefficient of Elovich fitting exhibited that chemical reaction did not occur during adsorption of enrofloxacin on biochar. The rate constant (k_1) for pseudo-first-order model was calculated as 1.85 while rate constant (k^2) for pseudo-second-order model was 0.41. The equilibrium adsorption capacity calculated by pseudo-first-order and pseudo-second-order models reached 9.26 and 9.52 mg/g, respectively. The better fitting result of pseudo-first-order model illustrated that the adsorption of enrofloxacin on biochar might majorly controlled by diffusion process. Most of biochar surface might possess one binding site. Moreover, pseudo-second-order model could also well describe the adsorption of enrofloxacin on biochar, illustrating that adsorption process might also be partially affected by chemical sorption on biochar surface tosome extent.

3.5. Adsorption isotherms of enrofloxacin by biochar made with fish feces and residues

Langmuir and Freundlich models have been frequently used to investigate the thermal kinetics of pollutants [35,36]. Therefore, isotherms of enrofloxacin on biochar made with fish feces and residues were also discussed (Fig. 5). Langmuir model could describe the homogenous adsorption

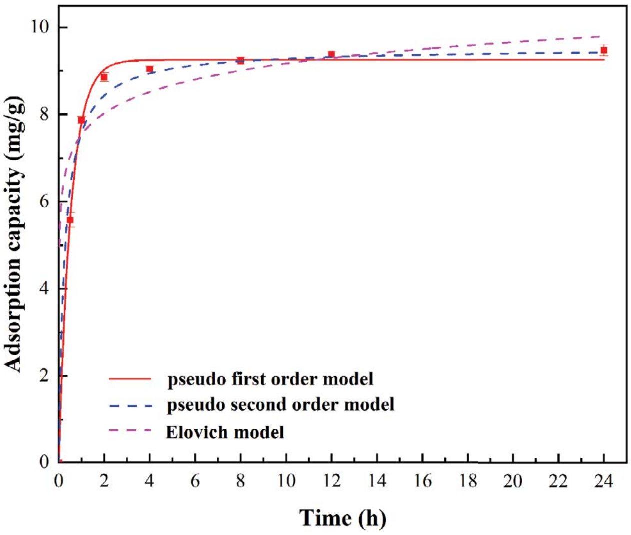

Fig. 4. Adsorption kinetics of enrofloxacin on biochar made by fish feces and residues.

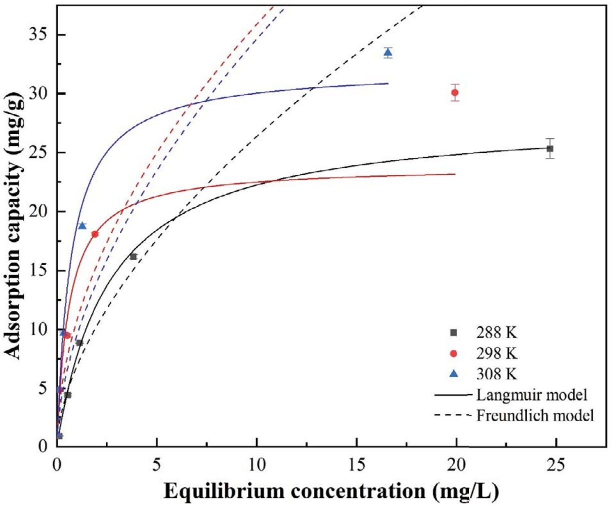

Fig. 5. Adsorption isotherms of enrofloxacin on biochar made by fish feces and residues.

with monolayer sorption process while Freundlich model could describe the heterogeneous adsorption with multiple-layer sorption process [36,37]. The maximal adsorption capacity at 15°C, 25°C, and 35°C reached 28.07, 23.85, and 32.16 mg/g by fitted with Langmuir model while Langmuir adsorption constant was 0.38, 1.65, and 1.41 L/mg, respectively. The Freundlich constant for 15°C/25°C/35°C was calculated as 6.90/10.77/9.51 L/mg while constant n was 1.72/1.91/1.78 for 15°C/25°C/35°C according to Freundlich model fitting. Langmuir model (R^2 = 0.9717–0.9934) could better describe the isotherm of enrofloxacin on biochar than Freundlich model with R^2 = 0.7242–0.8919 (Fig. 4), illustrating that monolayer adsorption might be the major thermal kinetic process. The isotherm mechanism of this study was different from that of previous study [13].

4. Conclusions

Biochar was prepared by fish feces and residues in a typical aquaculture system using hydrothermal method. Biochar showed amorphous structure with multiple elements on its surface. Biochar showed good adsorption capacity for enrofloxacin under 25°C and dosage of 1 g/L with the adsorption capacity of 9.47 mg/g. Pseudo-first-order model could well describe the adsorption kinetics of enrofloxacin by biochar while Langmuir model obtained the best fitting results for adsorption isotherm of enrofloxacin by biochar. Biochar could be reused 3–4 times to keep good removal efficiency higher than 77% so that it could be a prospective material of removing antibiotics in wastewater. The findings of this study will provide new insight on handling fish feces and residue of aquaculture as well as antibiotic pollution control. More application and disposal of aquaculture wastes should be discussed in the future.

Data availability

Data available on request.

Conflicts of interest

No potential conflict of interest was reported by the authors.

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