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Adsorption dynamics and mechanism of aqueous sulfachloropyridazine and analogues using the root powder of recyclable long-root *Eichhornia crassipes*

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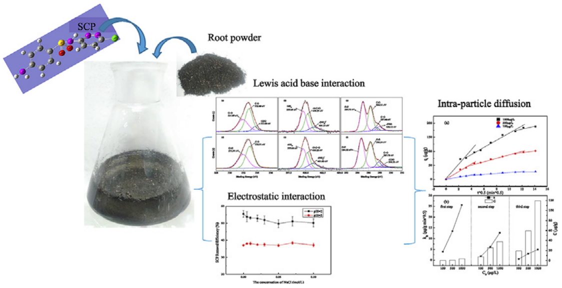
HIGHLIGHTS

- Root powder of long-root *Eichhornia crassipes* could well adsorb sulfonamides.
- Solution pH resulted in a significant effect on SCP and other sulfonamides removal.
- Sorption was generally governed by Lewis acid base interaction and electrostatic interaction.
- Both k_{ip} and the thickness of boundary layer had a linear negative relationship.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, we reclaimed the root powder of long-root *Eichhornia crassipes* (*L.R.E.C.*) as a biosorbent to remove aqueous sulfachloropyridazine (SCP) and other sulfonamides. The adsorption processes were investigated dependent on multiple measurements, including FT-IR and XPS analysis. The results confirmed that the basic amine group of neutral SCP molecules and the carboxyl hydroxyl on the surface of the root powder played the leading role in adsorption processes. Additionally, the experiments of ionic strength effect validated the involvement of electrostatic interaction in adsorption. Meanwhile, the adsorption data were fitted by various models and the results indicated that the Pseudo-second-order model and Freundlich model could well describe the adsorption processes, indicating the existence of physisorption and chemisorption as multi-layer adsorption. The maximum capacities of root powder for SCP were calculated to be $226.757\ \mu g\ g^{-1}$ (288.15 K), $182.815\ \mu g\ g^{-1}$ (303.15 K) and $163.132\ \mu g\ g^{-1}$ (318.15 K) at pH of 3.0. The thermodynamic results revealed that the adsorption was a spontaneous and exothermic process. Moreover, the accordance with intra-particle diffusion presented that the adsorption processes could be divided into three steps and the reaction constant had a negatively linear relationship with the thickness of the boundary layer. The results proved that root powder of *L.R.E.C.* has great potential to remediate sulfonamides at practical level.

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1. Introduction

Antibiotics are probably the most successful medicines for treating diseases and protecting the health of humans and animals. The global consumption of antibiotics keeps rising and increased by 35% from 2000 to 2010 (Van Boeckel et al., 2014). It had been reported that most of the consumed human and veterinary antibiotics are excreted unchanged in urine and faeces (Sarmah et al., 2006). The continuous exposure to antibiotics not only has a detrimental effect on aquatic ecology (Yang et al., 2010) and a stimulative effect on bacterial resistance (Wilson, 2014), but also it consistently has a systematic effect on human protective immunity and tissue immunity (Belkaid and Hand, 2014). Therefore, antibiotics have been increasingly worldwide thought as the 'pseudo-persistent' pollutants (Khetan and Collins, 2007).

Fortunately, a lot of technologies have been developed to remove the antibiotics from aqueous matrices, such as biological processes (Li and Zhang, 2010; Hijosa-Valsero et al., 2011), sedimentation/flocculation/coagulation (Niina M. Vieno et al., 2007), membrane processes (Košutić et al., 2007), advanced oxidation processes (Haidar et al., 2013; Fan et al., 2015), adsorption (Ahmed and Theydan, 2013; Liao et al., 2013) and combination techniques (Klavarioti et al., 2009; Homem and Santos, 2011). Among these remediation methods, adsorption process has been extensively utilized owing to its cheapness (Ahmaruzzaman, 2008), effectiveness (Zhou et al., 2012), and conveniences for subsequent operation. Biomass materials such as algae, yeasts, fungi, chitin/chitosan, activated sludge and bacteria have been attached tremendous attention as adsorbents due to their high abundance and biodegradability characteristics. These extensive characteristics of biomass materials make them suitable for effective removal of contaminants by suitably adjusting the functional groups such as hydroxyl (−OH), carboxyl (−COOH) and silanol (Si−OH) (Tran et al., 2015). The changes in inner structure of natural materials such as surface area, porosity and surface morphology to enhance removal efficiency have also been investigated (Wang and Chen, 2009; Tran et al., 2015). For instance, tendu leaf refuse is considered as an effective biosorbent to remove phenol from aqueous solution and the maximum adsorption capacities of the raw and modified materials were 7.69 and 31.35 mg g^{-1}, respectively (Nagda et al., 2007). Moreover, residue materials such as wood chips, ryegrass roots, orange peels, bamboo leaves and pine needles were also utilized to abate different kinds of PAHs (Chen et al., 2011). Boussahel et al. used wood sawdust and cork wastes to remove 4,4-DDT from water and found maximum adsorption capacities were greatly influenced by the size of the biosorbents (Boussahel et al., 2009). Additionally, these biosorbents could be used as the sustainable energy carriers in anaerobic fermentation (Lakaniemi et al., 2013) at saturated conditions. To date, remarkable efforts have been made to explore the adsorption performances of various biomaterials for antibiotics but scientific evidences to explore the concrete mechanisms is still ambiguous and inadequate. Therefore, understanding the fundamental adsorption processes plays crucial role in improving the adsorption capacity of biosorbents, indispensable for their practical application in future.

L.R.E.C. whose root occupied more than 80% of its biomass, has been planted in Dianchi Lake in China to treat eutrophication but recyclability of these ripe plants is still challenging. Due to massive availability of these plants, it would be much profitable to prepare a biodegradable adsorbent for contaminant removal using these reaped plants. In previous studies, researchers (Colleen Kelley et al., 1999; Zheng et al., 2009, 2016; Módenes et al., 2011) have focused on the adsorption of metals such as Eu(III), Pb(II), Zn(II), Cu(II), Cr(III) and Cd(II) onto root powder of traditional *E.C.* in single or binary systems and concluded that the existence of carboxyl (−COOH), amido (−NH_2) and hydroxyl (−OH) on the surface of root powder via chelation or coordination highly contributed to its adsorption capacity. El-Khaiary (2007) used nitric-acid treated water-hyacinth (N-WH) to remove methylene blue from aqueous solution and results showed that N-WH can remove MB effectively. Zawahry et al (El Zawahry and Kamel, 2004) discussed the adsorption of azo and anthraquinone dyes by raw and three aminated *E.C.*. It was reported that a higher nitrogen percent of aminated *E.C.* showed a higher adsorption capacity than other derivatives. And the comparison of *L.R.E.C.* and traditional *E.C.* in eliminating metals was conducted and results showed that *L.R.E.C.* had higher removal efficiency (Li et al., 2016), indicating that *L.R.E.C.* might had higher removal potential for pollutants than traditional *E.C.*. In this study, the root powder of *L.R.E.C.* was firstly developed to adsorb typical antibiotic sulfachloropyridazine (SCP) and other sulfonamides from the water. XPS and FT-IR were used to inspect the interaction of SCP and the root powder surface as well as explore their transformation in chemical features before and after the adsorption. The influences of different factors on the adsorption were also explored to enhance the adsorption performance, including initial pH, ionic strength and initial SCP concentration, *etc*. In addition, the adsorption kinetics and thermodynamics were also described by multifarious models to analyze adsorption mechanism that was vital for the further improvement of adsorption capacity of the root powder for the sulfonamides.

2. Experimental section

2.1. Materials and reagents

L.R.E.C. was collected from Dianchi Lake at Yunnan Province. Before the experiment, the roots were exhaustively washed, air dried and then grinded for further analysis. SCP and other sulfonamides were analytical grade, purchased from Aladdin Chemistry Co. The methanol and acetonitrile were both chromatographically pure grade and purchased from MACKLIN. All other chemicals used in the experiment were all analytically pure. The target antibiotics stock solution (1 g L^{-1}) was prepared by dissolving 0.1 g target compounds in 100 mL methanol and stored in the darkness at 4 °C. Deionized water was used to prepare all solutions.

2.2. Batch adsorption experiment

During the batch tests of the SCP adsorption processes, the effect of initial pH (1.5–11.0), SCP initial concentration (50–1000 $\mu g\ L^{-1}$), temperature (288.15, 303.15 and 318.15 K), reaction time (0–200 min) and ion strength (I_{NaCl} 0–0.1 M) were evaluated. 0.2 g adsorbent was added into a 250 mL flask containing 100 mL SCP aqueous solution. Solution pH was adjusted by 1.0 M H_2SO_4/NaOH to the particular value. Then the flask was put into a temperature-controlled lucifugal oscillator at about 195 rpm at a given temperature. The supernatant (0.5 mL) was sampled at a given interval and filtrated through a 0.45 μm filter (SCAA-113, ANPEL, China). Subsequently, the filtrate and methanol were mixed in the volume ratio of 3:7 and the mixture was again filtrated through a 0.25 μm filter (SCAA-114, ANPEL, China) for detection. All the experiments were performed in duplicates and average result was reported.

The removal efficiency and the adsorption capacity of SCP and substructural analogues by the root powder were calculated according to the equations as follows:

$$Removal\ efficiency\ (\%) = \frac{C_0 - C_e}{C_0} * 100\% \, . \tag{1}$$

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