



Sorptive removal of ionizable antibiotic sulfamethazine from aqueous solution by graphene oxide-coated biochar nanocomposites: Influencing factors and mechanism

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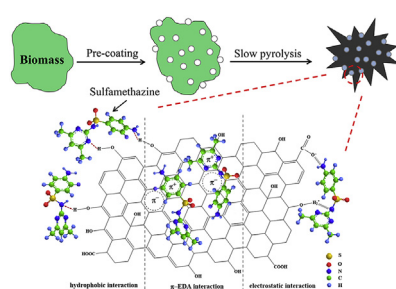
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HIGHLIGHTS

- Graphene oxide-coated biochar nanocomposites (GO-BC) were synthesized.
- Sorption capacity of SMT onto GO-BC increased by 1.14 times compared to biochar (BC).
- Chemical aging can significantly improve sorption performance of BC and GO-BC.
- The main sorption mechanism for SMT was π - π electron-donor-acceptor interaction.

GRAPHICAL ABSTRACT



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ABSTRACT

Significant concerns have been raised over antibiotics pollution in aquatic environments in recent years. In this study, sorption of sulfamethazine (SMT) by novel graphene oxide-coated biochar nanocomposites (GO-BC) based on graphene oxide (GO) with bamboo sawdust biochar (BC) was investigated. In comparison with the original BC, the sorption capacity of GO-BC for SMT increased by 1.14 times. Sorption of SMT onto GO-BC was proved to be dominantly by chemisorption, and Freundlich isotherm described the sorption adequately. It was found that variation of pH and ionic strength obviously affected the sorption of SMT, and GO-BC had a good sorption effect on SMT at pH 3.0–6.0 and lower ionic strength. Obvious enhancement (by 30%) in sorption of SMT on GO-BC was observed, which might be attributed to the increase of functional groups on the surface of GO-BC. Moreover, the main sorption mechanism for SMT was π - π electron-donor-acceptor interaction, while auxiliary sorption mechanisms were inferred as pore-filling, cation exchange, hydrogen bonding interaction and electrostatic interaction. The results indicated that GO-BC sorption was an efficient way for the removal of SMT.

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1. Introduction

With the advent of penicillin, pharmaceutical antibiotics are becoming more and more extensively used in farming industry and human therapy all around the world.

To improve the growth rate of animal, animal feed was usually incorporate with veterinary antibiotics in some countries (Sarmah et al., 2006; Zhou et al., 2013). Most antibiotics given to livestock cannot be metabolized and excreted via urine and faeces, and these residues eventually end up in soil, in which they may play a positive role in the proliferation of antimicrobial-resistant pathogens, thus affect humans potentially (Zhou et al., 2013). The effluent and sludge from domestic wastewater, hospitals or livestock farms with antibiotics lead to their release to the environments (surface and groundwater, soil and sediment) (Zhang et al., 2015). It cannot be denied that antibiotics are gradually becoming prevalent contaminants in our environment, and the augment in its concentrations are being observed whether in soil or in water. There has been found that the content of antibiotics in manure reached up to 900 mg kg⁻¹, even in surface water and sediment, the concentration ranged from ng L⁻¹ to µg L⁻¹ (Teixidó et al., 2013). In 1982, antibiotics were first detected in a river located in England, the concentration of sulphonamides, tetracyclines and macrolides were 1 µg/L. Thirty years later, the occurrence of antibiotic residues in aquatic ecosystems have been reported, and concentration of them 100 ng/L to 6 mg/L (Wang et al., 2016a,b). In the long run, antibiotic contamination is becoming more and more serious, and its pollution remediation is also imminent.

The object we studied is sulfamethazine (SMT; 4-amino-*N*-[4, 6-dimethyl-2-pyrimidinyl] benzenesulfonamide), which is commonly used among many veterinary medicine and has been continually reported in high concentration (García-Galán et al., 2010; Li et al., 2012). As a major sulfonamide drug, SMT is extensively used to control infectious diseases as well as treatment. Neutral form of SMT predominates between pH 3.0 and 7.0 and its high hydrophilicity (octanol water partition coefficient log $K_{ow} = 0.27$) still remained. SMT exist in water via several forms including uncharged molecule (SMT⁰), zwitterion (SMT[±]), cation (SMT⁺) and anion (SMT⁻), owing to proton exchange of aromatic amine and sulfonamide groups on molecule (pK_{a1}, 2.28; pK_{a2}, 7.42). In the aqueous system, the complexity of the SMT speciation indicates the complexity of the sorption behavior during its removal. Moreover, researchers have conducted lots of experiments and verified that sulfonamides are not easy to be adsorbed and not readily biodegradable, but leachable (Kim et al., 2010). Sorption behaviors of SMT have been investigated on materials such as multi-wall carbon (Ji et al., 2009; Yang et al., 2015) and graphene oxide (Gao et al., 2012; Nam et al., 2015), biochar (Zheng et al., 2013; Jing et al., 2014; Lian et al., 2014; Huang et al., 2016b), activated biochar (Rajapaksha et al., 2015), clay minerals (Gao and Pedersen, 2005), organophilic zeolite (Braschi et al., 2010), soils (Thiele-Bruhn et al., 2004; Rajapaksha et al., 2014), and even molecularly imprinted polymers (Xu et al., 2012; Huang et al., 2015, 2017; Wang et al., 2016a,b). A decrease in SMT sorption was observed with the pH increasing in both loamy sand and loam sediments, for the change of pH values affected the speciation of SMT (Vithanage et al., 2014). Such changes in environmental conditions are likely to have a great influence on the removal efficiency. Among numerous different parameters which may exist, ionic strength, pH, and ionic speciation were regarded as the most significant factors affecting the sorption of SMT to the majority of adsorbing materials (Lertpaitoonpan et al., 2009; Braschi et al., 2010). Varieties of mechanisms of SMT sorption including hydrophobic partitioning, cation exchange, and complex surface reactions (hydrogen bonding and other polar interactions) between the functional groups (carboxyl, phenol, and amino) of the adsorbate molecules have been proposed (Chen et al., 2014a,b).

Biochar (BC) is a kind of porous carbon material, and favored in environmental remediation with the characteristic of alternative and low-cost (Tan et al., 2015). It has been applied to remove

varieties of organic and inorganic pollutants including PAHs, PCBs, phenolic compounds, aldehydes, organic pesticides, and heavy metals (Zhang et al., 2014; Tran et al., 2015; Huang et al., 2016a,c; Liang et al., 2017), especially the removal of SMT, which has aroused wide public concern gradually (Zheng et al., 2013; Lian et al., 2014). Nevertheless, the sorption capacity of BC cannot meet the needs of the contaminants removing, no matter in water or in soil. Therefore, researchers have made efforts to prepare BC-based composite, considering that combine BC with organic or inorganic materials via physicochemical methods can improve its properties, create or incorporate new structure, and enhance the sorption capacity of BC (Inyang et al., 2014). There has been reported that synthesized BC-based nanomaterials exhibited strong capability of sorbing phosphate and nitrate (Zhang et al., 2012; Tang et al., 2014), methylene blue (Inyang et al., 2014), phenanthrene (Tang et al., 2015), lead, cadmium (Liu et al., 2016) and mercury (Tang et al., 2015), respectively. Graphene oxide (GO), exfoliated from graphite, has been considered as an emerging sorbent for various functional groups including hydroxyl, carboxylic, and epoxy groups on the surface (Wang et al., 2013a,b,c). Though this emerging nanomaterial possess excellent sorption ability, it is not appropriate for applying on a large scale because of the huge cost and potential risk to the environment (Wang et al., 2013a,b,c). If only incorporate this emerging nanotechnology with the available biochar technology to create novel nanocomposites, the performance of biochar can be improved. Besides, the carbonaceous nanocomposites still maintain low-cost (Zhang et al., 2016b; Wang et al., 2013a,b,c).

Consequently, the overall objectives of this study were to prepare hybrid GO-BC nanocomposites, and further test the SMT sorption behavior on the novel carbonaceous nanocomposites. Sorption kinetics, sorption isotherms, influence factors and the related mechanisms were investigated to obtain a deep understanding of the sorption behavior. Finally, the aging of GO-BC (primarily chemical oxidation) was also explored to simulate the effects of actual conditions in environment.

2. Materials and methods

2.1. Materials

Deionized water (resistivity of 18.2 MΩ cm) was used to prepare various solutions in this experiment. Sulfamethazine (99%, w/w) was purchased from Sigma-Aldrich, and some primary properties of which were shown in Table S1. Graphite powder was obtained from Sinopharm Chemical Reagent Co. Ltd. (China). Standard solution (365.23 mmol L⁻¹) was prepared via sulfamethazine dissolved in methanol and was used for preparing the starting concentrations of SMT in the following experiment. Biochar samples were produced from bamboo sawdust and ground in a knife mill to a size of 0.5–1.0 mm before production. Synthetic graphite powder (<20 µm particle size) and 1, 3, 6, 8-pyrenetetrasulfonic acid tetrasodium salt hydrate (PySA) were purchased from Energy Chemical (Shanghai, China). All the other chemicals used were analytical grade or better.

2.2. Preparation of biochar and graphene oxide/biochar nanocomposites

A modified Hummers method (Hummers and Offeman, 1958) was used to prepare graphene oxide. Briefly, 2 g of graphite and 1 g of NaNO₃ were mixed and placed in a 500 mL beaker. Under an ice bath condition, 46 mL of sulfuric acid (98%) was added. When the temperature was below 10 °C, slowly added 6 g of KMnO₄ and 1 g of NaNO₃ to the suspension and stirred vigorously. After keeping

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