



Adsorption kinetics of magnetic biochar derived from peanut hull on removal of Cr (VI) from aqueous solution: Effects of production conditions and particle size



Yitong Han^a, Xi Cao^a, Xin Ouyang^a, Saran P. Sohi^b, Jiawei Chen^{a,*}

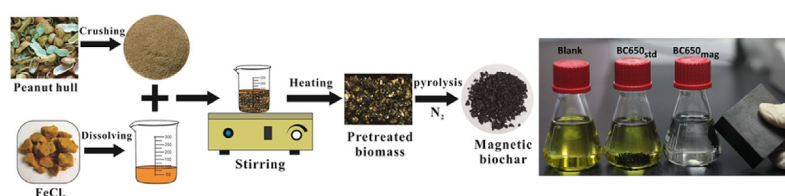
^a State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Beijing 100083, PR China

^b UK Biochar Research Centre, School of GeoSciences, University of Edinburgh, The King's Buildings, Edinburgh EH9 3JN, UK

HIGHLIGHTS

- Magnetic biochar derived peanut hull was simplified created with higher adsorption for Cr(VI) in aqueous.
- γ -Fe₂O₃ loaded on biochar played a key role in Cr(VI) adsorption.
- Higher pyrolysis temperature and smaller particle size of magnetic biochar could result in higher adsorption of Cr(VI).

GRAPHICAL ABSTRACT



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ABSTRACT

Magnetic biochar was made from peanut hull biomass using iron chloride in a simplified aqueous phase approach and pyrolysis at alternative peak temperatures (450–650 °C). Magnetic biochar showed an extreme capacity for adsorption of hexavalent chromium Cr (VI) from aqueous solution, which was 1–2 orders of magnitude higher compared to standard (non-magnetic) biochar from the same feedstock. Adsorption increased with pyrolysis temperature peaking at 77,542 mg kg⁻¹ in the sample pyrolysed at 650 °C. In contrast to magnetic biochar, the low adsorption capacity of standard biochar decreased with increasing pyrolysis temperature. The fine particle size of magnetic biochar and low aqueous pH were also important for adsorption. Surfaces of products from batch adsorption experiments were characterized by scanning electron microscopy, energy-dispersive X-ray analysis, X-ray diffraction, X-ray photoelectron spectroscopy and vibrating sample magnetometer. This revealed that γ -Fe₂O₃ was crucial to the properties (adsorbance and magnetism) of magnetic biochar. The removal mechanism was the Cr (VI) electrostatic attracted on protonated –OH on γ -Fe₂O₃ surface and it could be desorbed by alkaline solution. Findings suggest that pyrolysis has potential to create effective, magnetically recoverable adsorbents relevant to environmental application.

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1. Introduction

Biochar is a carbon rich solid created by high temperature pyrolysis of biomass – heating in the absence of oxygen. Since the

* Corresponding author.

E-mail address: chenjiawei@cugb.edu.cn (J. Chen).

term 'biochar' was established in 2006 (Lehmann et al., 2006) it has been the subject of intensive investigation, both its direct effects on soil and relevant studies (Conte, 2014; Lehmann, 2007; Sohi, 2013). Biochar is a robust way to store plant-derived carbon and thus limits the natural recycling of CO₂ back to the atmosphere (Renner, 2007). Moreover, biochar has great potential for remediation of water contamination. It has been noted that diverse functional groups inhabiting the formation of the stable porous structure of

high specific surface area might benefit aqueous contaminant removal from solution (Ahmad et al., 2014; Cao and Harris, 2010; Mohan et al., 2014; Zhang et al., 2013a, 2013b).

The general capacity of biochar to adsorb hexavalent chromium Cr(VI) has been previously reported. Chromium is a common heavy metal pollutant in water and the potential utility of biochar in Cr(VI) removal is attracted. For instance, results have ranged from 123 mg g⁻¹ for biochar from sugar beet tailings (Dong et al., 2011) to 64.1 mg g⁻¹ for sewage sludge biochar (Agrafioti et al., 2014a, 2014b) and 23.1 mg g⁻¹ for biochar from the residues of major food crops, for example rice-husk (Ma et al., 2014). Such feedstock has a low value and price as its abundance exceeds the size of established markets for alternative use. This improves the economic case for biochar manufacture relative to dedicated biomass resources, which has been more widely tested. However, much higher adsorption capacities are still required to accelerate the adoption of biochar for land and water remediation purposes. Simple modification of residue-derived biochar is therefore both important and urgent.

Modifications that have been discussed to date are: (1) carriage of metals, oxides or ions (Agrafioti et al., 2014a, 2014b; Zhang et al., 2012), (2) mixing biochar with reducing or oxidizing salts (Pan et al., 2014), (3) modifying biochar by organic compounds that display specific functional groups (Ma et al., 2014). Magnetic biochar has been described among these approaches and successfully produced by chemical co-precipitation of iron oxides. Magnetic biochar has been found to provide effective adsorptivity, ready separation and easy recycling (Chen et al., 2011; Reddy and Lee, 2014; Zhang et al., 2013a, 2013b).

Meanwhile, China is the largest producer of peanut (*Arachis hypogaea*) globally and generates more than 5 million metric tons of peanut hulls from processing and utilization every year. Although a small proportion of peanut hull is used in animal feed, most is burned or discarded, causing environmental problems (Wang et al., 2013). Prior work has shown that biochar from peanut hulls has rough surface, high porosity, capillary structure and various chemical functional groups such as hydroxyl –OH, ester C=O and aromatic C=C (Ahmad et al., 2012).

The recoverable magnetic biochar derived from peanut hull is great potential for water remediation. In the present study the main objectives include: (1) to create magnetic biochar with much higher capacity to adsorb Cr(VI) in aqueous; (2) to study the effects of production conditions, particle size on Cr(VI) adsorption on magnetic biochar.

2. Materials and methods

2.1. Chemicals and materials

The starting material for the manufacture of biochar in this study was peanut hulls from the farmlands of Fengnan county, Hebei province, China. The peanut hulls were thoroughly washed using deionized water (18.3 MΩ cm) and stored in an oven at room temperature (25 ± 0.5 °C) before pyrolysis. FeCl₃•6H₂O (Sinopharm Chemical Reagent Co., Beijing, China) were used for biochar magnetization. 1,5-diphenylcarbazine (Alfa Aesar, Shanghai, China) was used as chromogenic reagent in Cr(VI) determination. Cr(VI) stock solution of 4.0 g L⁻¹ was prepared by dissolving K₂CrO₇ (Sinopharm Chemical Reagent Co., Beijing, China) in deionized water. All chemicals were analytical grade and used without further purification.

2.2. Preparation of magnetic biochar

We manufactured magnetic peanut hull biochar by modifying

the approach of Zhang et al. (2013a, 2013b). Our objective was to achieve a more even distribution of iron in biochar as well as to use less time and energy. Briefly, as shown in Fig. 1, 10 g of dried peanut hulls were ground lightly using micro plant grinding machine (FZ102, Zhongxing Co., Beijing). Biomass particles were then soaked in 80 ml of FeCl₃ solution of 2 M. After 0.5 h mixing under magnetic stirring, hydrolysis and Fe³⁺ precipitation was accelerated by 'ageing' at 70 °C for 0.5 h. The pre-treated biomass was then separated from the solution and pyrolyzed in an oxygen-free furnace under 200 ml min⁻¹ N₂ atmosphere. Five alternative peak temperatures were used: 450, 500, 550, 600 and 650 °C. Each was reached using the same heating rate of 5 °C min⁻¹ and each held at peak temperature for 1 h. After cooling biochar particles were rinsed by deionized water for several times, then dried at 70 °C for 1 h. To assess the effect of particle size on adsorption capacity, we isolated two different size fractions for comparable experiments by dry sieving: 0.15–0.50 mm and 0.50–1.00 mm. Non-magnetic (standard) biochar was made in the same way and pyrolysed directly under the same set of temperature conditions. Biochar samples are identified as 'BC', with a suffix indicating peak temperature in pyrolysis and subscript denoting prior immersion in FeCl₃ (mag) or non-immersion (std).

2.3. Characterization of biochar

All biochar samples (BC_{mag} and BC_{std}) were characterized for their physical and chemical micro-scale features. Field emission scanning electron microscopy (FE-SEM) was applied at ultra-high resolution using a Merlin VP Compact instrument (JSM-6460LV, Zeiss, Germany) equipped with an energy-dispersive X-ray analyzer. X-ray photoelectron spectra (XPS) showing elemental valence states were obtained using ESCALAB 250Xi (Thermo Fisher, USA). The crystallographic structures of the synthesized biochar was examined by X-ray diffraction (XRD) analysis, using a D/Max 2500 (Rigaku, Japan) with a Cu Kα radiation in the range of 10–90° (2θ). Inductively coupled plasma optical emission spectrometry (ICP-OES) was used to measure the concentration of iron oxides in biochar (Spectroblue, Spectro Analytical Instruments, Kleve, Germany) via dry ashing and acid digestion. BET surface area was quantified based on N₂ multilayer adsorption at room temperature using a Quadrasorb Station 1 analyzer (Quantachrome Instruments, Boynton Beach, USA). The magnetic properties of BC_{mag} were assessed using a vibrating sample magnetometer (VSM-7307, Lake Shore Cryotronics, Westerville, USA).

2.4. Batch experiments

The BC samples used in this study were used in adsorption experiments to ascertain the extent and kinetics of Cr(VI) removal from water. 0.10 g BC was added into conical vitreous vessels with 50 ml solution of K₂CrO₇ (200 mg L⁻¹). The initial pH of Cr(VI) solutions with no buffer were measured to know pH = 5.13 ± 0.02 using pH meter (PB-10, Sartorius, Goettingen, Germany). 0.1 M HCl and 0.1 M KOH solution were used for initial pH adjustment in Cr(VI) solution. The closed vessels were placed on a reciprocating shaker at 160 rpm at room temperature (25 ± 0.5 °C). At intervals 0.30 ml of suspension were withdrawn and filtered at 0.22 μm (substitute Syringe filter with Whatman Nylon 66 membrane). 0.20 ml of the filtrate was used for colorimetric measurement under UV–visible spectrophotometry (UV1750, Shimadzu, Kyoto, Japan) at 540 nm by 1,5-diphenylcarbazine method (Cao and Zhang, 2006), while the solid and remaining solution were returned to the vessels. As a blank, aliquots of the initial 200 mg L⁻¹ Cr(VI) solution with no biochar was also measured on each occasion by colorimetric analysis. No change in absorbance intensity indicated that

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