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Adsorption characteristics of arsenic from micro-polluted water by an innovative coal-based mesoporous activated carbon

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HIGHLIGHTS

- A coal-based mesoporous AC (NCPAC) was prepared by a novel multi-step procedure.
- The novel method lead to high mesoporosity, ash content and adsorption ability.
- The Langmuir model fitted the experimental data well for both As(III) and As(V).
- NCPAC was a promising adsorbent for arsenic removal (<0.5 mg/L).
- Regenerated carbon preserved high adsorption capacities of arsenic ions.

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ABSTRACT

An innovative coal-based mesoporous activated carbon (NCPAC) was prepared by re-agglomeration, oxidation and two-step activation using coal-blending as precursor. Adsorption capacities of As(III) and As(V) ions (<0.5 mg/L) onto NCPAC as a function of pH, adsorbent dose, initial arsenic concentrations, contact time, and adsorption isotherms at 7 °C was investigated. The innovative methods promoted total pore volume (1.087 cm³/g), mesoporosity (64.31%), iodine numbers (1104 mg/g), methylene blue (251.8 mg/g) and ash contents (15.26%). The adsorption capacities of NCPAC for As(III) and As(V) were found to be strongly dependent on pH and contact time. The optimal pH value was 6. The equilibrium time was 60 min for adsorption of As(III) and As(V) by NCPAC. The Langmuir model fitted the experimental data well for both As(III) ($R^2 = 0.9980$) and As(V) ($R^2 = 0.9988$). Maximum adsorption capacities of As(III) and As(V) ($C_0 = 0.50$ mg/L) by NCPAC were 1.491 and 1.760 mg/g, respectively.

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1. Introduction

Urban water supply is threatened by hazardous chemicals both in domestic and international cities (Li et al., 2012). The presence of arsenic ions in natural waters has become an important issue around the world. Arsenic was identified as a highly toxic metalloid that causes severe environmental and public health problems. Researches focusing on effects of arsenic concentration on human health were conducted by US Environmental Protection Agency (USEPA). Results suggested that long-term drinking water exposure even in a very low arsenic concentration (0.01–0.05 mg/L) can cause lethal diseases like cancers of skin, lung, bladder and

kidney (Neumann et al., 2013). Therefore, arsenic is now regulated at 0.01 mg/L for drinking water by USEPA, China and the World Health Organization (WHO), downward from the old standard of 0.05 mg/L.

Arsenic removal from water is an important subject and it has recently attracted great attention. Arsenic is usually encountered in two forms: arsenite (AsO_3^{3-}) and arsenate AsO_4^{3-} , referred to as arsenic(III) (As(III)) and arsenic(V) (As(V)) (Zhang et al., 2013). Various treatment technologies are available for the removal of As(III) and As(V) ions from water, such as oxidation/precipitation, coagulation/co-precipitation, ion-exchange, reverse osmosis and adsorption. But none of them is found to be completely applicable, all suffer from one or more drawbacks, limitations and scope of application. Chemical precipitation and co-precipitation are limited by effects of added reagent, the production of chemical sludge and low efficiency at low temperature (Allende et al., 2012). Moreover, arsenic precipitates are unstable under some definite conditions and arsenic could be released in the environment. Ion exchange

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Nomenclature

C_0	Initial concentration of arsenic in solution (mg/L)	q_t	The amounts of arsenic ions adsorbed at time t (mg/g)
C_e	Concentration at equilibrium (mg/L)	S_{BET}	BET Surface Area (m^2/g)
C_t	Experimental concentrations of arsenic in solution at time t (mg/L)	S_{Micro}	t -plot micropore area (m^2/g)
K_F	Freundlich constant indicative of the relative adsorption capacity of the adsorbent ($(\text{mg})^{1-n} \text{L}^n/\text{g}$)	S_{Ext}	t -plot external surface area (m^2/g)
K_L	Langmuir equilibrium constant (L/mg)	$S_{(\text{BJH-Adsorption})}$	BJH Adsorption cumulative surface area of pores (17–3000 Å),
M	The mass of the adsorbent added (g)	$S_{(\text{BJH-Desorption})}$	BJH Desorption cumulative surface area of pores (17–3000 Å),
n	Freundlich constant indicative of the intensity of the adsorption	t	Time of reaction (min)
k_1	The pseudo-first-order equation rate constants (1/min)	V	The volume of sample used (L)
k_2	The pseudo-second-order equation rate constants (g/mg min)	V_{total}	Single point adsorption total pore volume of pores less than 793.112 Å diameter at $P/P_0 = 0.974971777$ (cm^3/g)
P	The equilibrated pressure (Pa)	V_{micro}	t -plot micropore volume (cm^3/g)
P_0	The saturation pressure (Pa)	V_{meso}	Calculated mesopore volume (cm^3/g)
q_m	Maximum adsorption capacity from Langmuir (mg/g)	$V_{(\text{BJH-Adsorption})}$	BJH Adsorption cumulative volume of pores (17–3000 Å)
q_e	PAC surface complex concentration at equilibrium (mg/g)	$V_{(\text{BJH-Desorption})}$	BJH Desorption cumulative volume of pores (17–3000 Å)

has the disadvantage of releasing harmful chemicals into the environment when the resin is regenerated. Reverse osmosis is commonly employed, but it is expensive, mainly because of the high energy requirements. Adsorption is based on materials having a high affinity for dissolved arsenic. Although this process also has some drawbacks, it still offers significant advantages for controlling the extent of aqueous arsenic pollution, such as high efficiency, mainly an exothermal reaction and the possibility of metal(loid) recovery (Foo and Hameed, 2012; Kadirvelu et al., 2003).

As the most widely used adsorbents for water purification, activated carbon (AC) adsorption was studied extensively for arsenic removal. Studies suggested that metal(loid) ions adsorption on AC is more complex than uptake of organic compounds, mainly because ionic charges affect the removal kinetics from water. Adsorption capacity of commercial AC for metal ions in high concentrations from industrial polluted water is relatively high. The removal rate gap under low temperature and room temperature is slight. However, commercial AC has low selectivity and effectiveness for metal(loid) species in low concentration (<0.5 mg/L) from low temperature water due to the competitive adsorption of natural organic matters (NOM) and other organic contaminations. That is why many ACs are available commercially, but few are selective for adsorption of As(III) and As(V) in low concentration (<0.5 mg/L) from water. Although the adsorption capacities of arsenic with virgin ACs were poor, fortunately, it can be improved by modifications. Surface impregnation has been identified as one of the key methods for promoting adsorption affinity, selectivity and effectiveness for certain metal(loid) species. Different methods have been developed to impregnate AC with more iron, silver and copper, so as to increase arsenic adsorption capacity (Awual et al., 2013; Babu et al., 2013; Daud and Ali, 2004). The impregnation process offers high affinity for arsenate and arsenite (Chen et al., 2007), but it impairs surface area, pore structure and uptake of organics from water (Rivera-Utrilla et al., 2011). Therefore, improved or tailor-made carbon materials are imperative to balance inorganic ion adsorption with organic matter adsorption. The ideal carbon should have high adsorption efficiency for metal(loid) ions and organic matters without impregnation process. Therefore, it is significantly important to study and establish an innovative method to prepare ACs with proper adsorption capacities for metal(loid) ions and organic matters.

In the current study, an innovative coal-based mesoporous activated carbon (NCPAC) was originally prepared to enhance

adsorption capacity for NOM and performance in bio-enhanced AC filter (BEAC). High adsorption affinity for arsenic and $\text{NH}_4\text{-N}$ was found by accident. Consequently, a series of improvements was conducted in the production process of this innovative carbon, aiming at the efficient removal of arsenic ions in low concentrations from low-temperature source water. Therefore, objectives of this work are the following: (i) to prepare and characterize the physical and chemical surface properties of NCPAC; (ii) to investigate the effects of pH, adsorbent dose and initial concentrations on arsenic adsorption onto NCPAC; (iii) evaluate the adsorption kinetics and equilibrium isotherms of As(III) and As(V).

2. Methods

2.1. Adsorbent

NCPAC (200 meshes) used in present work was prepared using coal blending of Shenfu coal (SFC) and Datong coal (DTC). SFC and DTC were provided by Shanxi Xinhua Co., Ltd., Proximate and ultimate analyses of precursor substance were shown in Table 1. The preparation process of NCPAC mainly consists of following steps. Firstly, coal-blending of SFC(60%) and DTC(40%) was pulverized to powder approximately 325 microns in size and evenly mixed. The mixing product was re-agglomerated into briquettes under 200 Mpa. Briquettes were crushed to the desired size (2–10 mm) and carbonized consequently in a rotating carbonization furnace. The carbonization was conducted in N_2 flow at a heating rate of $5^\circ\text{C}/\text{min}$ to $630 \pm 20^\circ\text{C}$, then held at this temperature for 30 min. Before activation, the carbonized product was oxidized in CO_2 flow at $600 \pm 20^\circ\text{C}$ for 20 min. Then, two-step activation including regular and depth activation was conducted. Regular activation was conducted at $900 \pm 20^\circ\text{C}$ for 120 min with the amount of 0.8 mL $\text{H}_2\text{O}/(\text{h g char})$ in Stepple furnace. In this process, the generation

Table 1
Proximate/ultimate analysis of precursors.

Coal	Proximate analysis (%)				Ultimate analysis (%)				
	V_{daf}	A_{d}	FC_{daf}	M_{ad}	C	H	O	N	St
SFC	34.98	2.64	60.83	1.55	90.57	6.85	2.71	0.76	0.11
DTC	33.23	3.71	57.58	5.48	89.42	3.92	5.24	1.21	0.21

Here, SFC: Shen Fu Coal; DTC: Da Tong Coal; V_{daf} : volatiles, A_{d} : ash, FC_{daf} : fixed carbon, M_{ad} : moisture in air-dried sample.

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