



# The modification of carbon materials with carbon disulfide for the removal of $Pb^{2+}$



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## ABSTRACT

Carbon spheres (CSs) were prepared with hydrothermal method, and multiwall carbon nanotubes (MWCNTs) and graphites were oxidized with Hummer's method. The obtained CSs, oxidized CNTs (OCNTs) and graphene oxides (GOs) included the oxygen-containing functional groups on these zero, one and two dimensional carbon materials. Carbon disulfide ( $CS_2$ ) was used to react with oxygen-containing groups for the formation of xanthate groups, which could produce the chelation structure with  $Pb^{2+}$ . The effect of  $CS_2$  dosage on the  $Pb^{2+}$  adsorption kinetic and isotherm of the modified CSs, OCNTs and GOs was investigated. The kinetic adsorption fit the pseudo-second-order model and the isotherm data followed the Langmuir model well. The maximum adsorption capacities for  $Pb^{2+}$  were 250.0, 357.1 and 500.0 mg/g for the modified CSs, OCNTs and GOs, respectively. These  $CS_2$ -modified carbon materials were competitive with other adsorbents for the heavy metal ions. Although the  $Pb^{2+}$  adsorption decreased much with the increasing of adsorption-desorption cycle times, these carbon materials still exhibited the good  $Pb^{2+}$  adsorption capacity.

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## 1. Introduction

The presences of heavy metals (such as arsenic, chromium, lead, mercury, copper) in the aquatic environment have been paid considerable attention [1–5]. These heavy metals are harmful to human beings and toxic to microorganisms. Carbon materials are well-known “universal” adsorbents and present unique advantages because of high adsorption capacity and easy disposal. The performance of carbon materials depends greatly on their texture and surface chemistry [6].

The different dimensional carbon materials have been researched. The hydrothermal carbonization is a well-known process to produce zero dimensional carbon spheres (CSs) [7,8]. In this process, glucose is converted into 5-hydroxymethyl-2-furaldehyde, which was subsequently condensed and polymerized to form carbon materials [9]. CSs exhibit a core-shell chemical structure composing of the aromatic nucleus (hydrophobic) with oxygen forming stable groups (i.e. ether, quinone, pyrone, etc.) and a hydrophilic shell, which includes the reactive and hydrophilic oxygen functional groups (i.e. hydroxyl, phenolic, carbonyl, carboxylic, ester, etc.) [10]. The products from glucose were more reactive than other mono-saccharides [9].

Recently, carbon nanotube (CNT) and graphene have attracted interest as new adsorbents due to their large specific surface areas and layered structures [11–14]. Two dimensional graphene oxide (GO) has

many chemically reactive oxygen functional groups: carboxylic acid groups at the edges of GO, and epoxy and hydroxyl groups on the basal planes [15]. As the one dimensional carbon material, CNT is regarded as an array of graphene sheets rolled up into a hollow cylinder. Despite the many desirable properties, the hydrophobicity and chemical inertness of CNT hinders their application [16]. The oxygen containing groups could be introduced into CNT with Hummer's oxidation, which were often used to prepare GO [17]. As the ubiquitous material, carbon materials with these oxygen containing groups could further chemically modified for the different applications [18].

Many functional groups have contributed to good removal of heavy metal ions from water. The thiol and hydroxyl groups were responsible for the adsorption of Hg ions from water [19], amino alcohol groups for Cu and Pb ions [20], N and S containing groups for Cu ions [21], epoxy, hydroxyl, and carboxyl groups for Cu ions [22], and hydroxyl and carboxyl groups for selenate [23]. Xanthate groups were also the effective functional groups to remove heavy metal ions due to the chelation between xanthate groups and heavy metal ions [24]. The hydroxyl groups of natural polysaccharides could easily react with carbon disulfide to produce xanthate groups [25]. The modification of carbon materials involving xanthate groups was rarely reported. This work attempted to confirm that the functional groups from the modification of natural polysaccharides could be produced on carbon materials containing the reactive groups. CSs from hydrothermal carbonization, oxidized CNT (OCNTs) and GOs from Hummer methods were reactive to carbon disulfide to form xanthate groups on zero, one and two dimensional carbon

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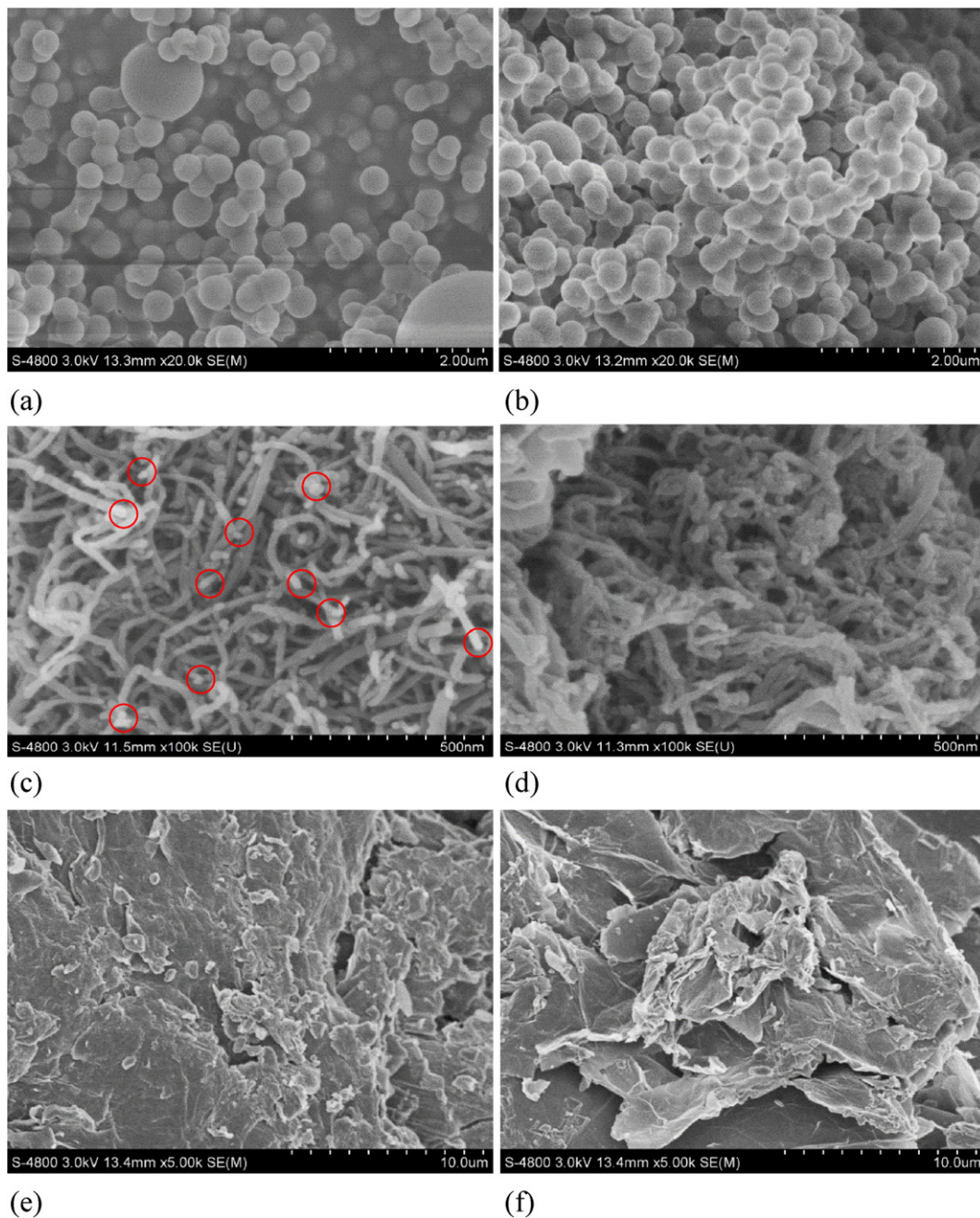


Fig. 1. SEM images of CS (a), CCS-0.1(b), OCNT (c), COCNT-0.2 (d), GO (e) and CGO-0.05 (f).

materials. Lead was chosen as the template. The main sources of lead in water are the effluents of processing industries, and lead poisoning in humans can cause severe damage to the kidney, nervous system, reproductive system, liver, and brain [1]. Subsequently, the adsorption patterns of  $Pb^{2+}$  ions by these different carbon materials with xanthate groups were characterized.

## 2. Experimental

### 2.1. Materials

MWCNT (with an average diameter of about 50 nm) were purchased from Chengdu Organic Chemicals Co. Ltd., Chinese Academy of Sciences, China. Natural graphite flakes were provided by Qingdao Tianhe Graphite Co., Ltd., China. All other reagents were commercially available and of analytical grade.

### 2.2. Preparation

#### 2.2.1. Preparation of carbon spheres (CSs) with hydrothermal method

5.4 g glucose was dissolved into 20 mL water in a Teflon-lined autoclave. The reactor was then placed in an oven fixed at 180 °C for 7 h. After hydrothermal carbonization, the product was collected by centrifugation, and then repeatedly washed with water and ethanol. CSs were obtained after being dried under vacuum at 80 °C for 5 h.

#### 2.2.2. Preparation of oxidized CNT (OCNT) and graphene oxides (GOs)

OCNTs and GOs were prepared using a modified Hummer's method [17]. Concentrated  $H_2SO_4$  (46 mL) and  $NaNO_3$  (1 g) were added into a flask and cooled to below 5 °C, followed by the addition of 2 g CNT (or graphite). Solid  $KMnO_4$  (6 g) was added gradually under stirring and stirred for 1 h at lower than 20 °C. The temperature was then increased

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