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Removal of heavy metals using Amine crosslinked Reduced Graphene Oxide

Cadiam Mohan Babu[1,2], Rajangam Vinodh[1,2], Aziz Abidov[1,2], Rramaswamy Ravikumar[1,2], Mei Mei Peng[1,2], Wang Seog Cha[2,3] and Hyun-Tae Jang[1,2]

[1]Department of Chemical Engineering, Hanseo University, Seosan 360-706, South Korea
[2]Korea Carbon Capture and Sequestration R and D Centre, 360 Daegok-ri, Haemi-myun, Seosan-si 356 706, Chungcheongnam-do, South Korea
[3]School of Civil and Environmental Engineering, Kunsan National University, Kunsan 573-701, South Korea
E-mail: htjang@hanseo.ac.kr; Fax: +82-041-688-4143; Tel: +82-041-660-1423

Abstract. Graphene oxide was synthesized by modified Hummers method and then reduced with hydrazine hydrate to obtain reduced graphene oxide. (3-aminopropyl)trimethoxysilane used as a precursor for amine functionalized reduced graphene oxide. The amine functionalized nanoparticles were characterized using FT-IR, TGA, Raman, N_2 sorption, SEM and TEM techniques. The surface area was found to be 346 m^2/g due to functionalization of amine over reduced graphene oxide. The amine functionalized nanoparticles were used as adsorbent for the removal of heavy metals from aqueous solution. The adsorption of heavy metal was high for the reduced graphene oxide nanocomposites when compared to bare amine functionalized nanoparticles. The maximum adsorption reached within fifteen minutes.

Keywords: Reduced Graphene Oxide, Amine Functionalized, Heavy metals and Nanocomposites.

1 Introduction

Water is one of the basic needs required for sustaining healthy life. It is therefore not a surprise that water has occupied an important position in the activities of mankind. With the development of industrialization and urbanization, water utilization has increased double every fifteen years [1]. Due to the industrialization the water is contaminated by various contaminants in that heavy metal plays a vital role. A heavy metal is any metal or metalloid of environmental concern. The word specified with reference to the harmful effects of lead, mercury and cadmium, all of which are denser than iron. It has also been applied to other toxic metal, or metalloid such as arsenic. Heavy metals "can bind to vital cellular components, such as structural proteins, nucleic acids and enzymes, and interfere with their routine functioning". Heavy metals are very dangerous, unlike organic contaminants, they are non-biodegradable and making great impact and threat to the ecological environment and

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human health. The most common heavy metals which present in water are lead, cadmium, arsenic, chromium, mercury, lead, mercury, zinc and nickel. These heavy metal ions can cause adverse effects to human health [2]. Lead is the one of the most abundant natural heavy element. The major form of lead in nature as galena (PbS), cerussite ($PbCO_3$) plattnerite (PbO_2), and anglesite ($PbSO_4$). Lead is more toxic towards human as well as aquatic life. Lead produces structural modifications in chromosome and binds very strongly to mitochondrial membranes. The presence of lead in water is discharged from solder, paint, discarded batteries, pipes and leaded gasoline. Lead is one of the high toxic element. Water containing excess lead was taken by humans was affected by the central nervous system, gastrointestinal system, kidney and liver, and some of the diseases caused by lead is anemia, hepatitis, encephalopathy and nephritic syndrome [3].

Some of the methods used for the removal of heavy metals are coagulation, reverse osmosis, adsorption and ion-exchange are used for the removal of heavy metals from aqueous solution [4]. Adsorption is one of the efficient method for the heavy metal removals [5]. Zeolites were used as adsorbent in heavy metal removal applications due to diffusion, adsorption and ion exchange properties present in the materials. Zeolites channels can adsorb organic or inorganic molecules through internal cavities. Modified zeolites can be used as an ion exchanger for the adsorption of heavy metal ions. The removal of arsenic from water by natural zeolite in the batch process was studied by Payne & Fattah [6]. Magnetism is one of the unique physical property of the metal oxides that can be utilized in water purification process [7]. Iron oxide (Fe_3O_4 and γ-Fe_2O_3 nanoparticles) is used as magnetic material for the heavy metal removals [8]. Natural plant roots used for lead adsorption and the adsorption rate was 88 percentage was achieved [9].Various types of carbon materials was used for water treatment [10-12]. Graphene oxide used as a potential carrier support for the metal oxide nanoparticles in the adsorption studies. Reduced graphene oxide supported polypyrole nanocomposites used as an efficient adsorbent for the removal of mercuric ions [13]. In this work we synthesized amine crosslinked reduced graphene oxide nanoparticles and these nanoparticles used for the removal of heavy metal ions from aqueous solution. The concentration of heavy metal ions before and after adsorption over nanocomposites was studied using ICP-OES.

2 Reagents and Materials

Tetraethylorthosilicate (TEOS), (3-aminopropyl)trimethoxysilane (APTMS), Ferric chloride ($FeCl_3.6H_2O$) and ferrous sulfate ($FeSO_4.7H_2O$) were used for the synthesis of amine functionalized nanoparticles. Graphite flakes from Alfa essar used for the preparation of graphene oxide. Lead nitrate, sodium arsenate dibasic heptahydrate and cadmium nitrate tetrahydrate were used as the source for lead, arsenate and cadmium ions.

2.1 Characterization

The X-ray diffraction (XRD) patterns, textural properties and surface morphology of nanocomposites were recorded on a PANalytical X'Pert Pro X-ray diffractometer, Micromertics ASAP 2020 volumetric adsorption analyzer, scanning electron microscope (SEM) SEM-JEOL JSM-5600 model and High resolution transmission electron microscope JEOL, JEM model. The lead, arsenate and cadmium ion concentration in the aqueous solution before and after adsorption were analysed using a Perkin Elmer (OPTIMA 7000 DV) inductively coupled plasma-optical emission spectrometer (ICP-OES).

2.2 Synthesis of GO-Nanocomposites

Graphene Oxide was synthesized by modified Hummers method [14]. Magnetic Iron oxide Fe_3O_4 nanoparticles were synthesized by coprecipitation method. Amine functionalized nanoparticles was synthesized by using the solution method [5]. Add 10mg of synthesized reduced graphene oxide and 90 mg of crosslinked amine crosslinked reduced graphene oxide nanopaticles in round bottom flask containing 150ml water and stirred the solution for 5 h. After 5 hours, filter the black precipitate by using ethanol. Then dried at 80 °C under vacuum condition to obtained reduced graphene oxide nanocomposites.

2.3 Batch Sorption Experiment

The adsorption experiments were carried out using mechanical shaker at 250 rpm in 200ml polypropylene flask. Various concentration of lead ions (5-100 mg/L) were prepared and used for the adsorption studies. Take 0.2 g of RGO nanocomposites, dispersed in flask containing 100ml of arsenic, cadmium and lead ion solution and shake the flask for 24 h. After the adsorption, the concentration of the lead ions was analyzed by ICP-OES (optima 7000).

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