



Removal of Lead from Wastewater by Graphene Coated Sand Composite

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Abstract

In this research a process is used to treat contaminated water for its most recent use into an effluent that can be either returned to the water cycle with minimal environmental issues or reused. Lead pollution impacts all the systems of the human body. Many of the methods utilized for treatment. An easy and low cost method was employed to remove lead from wastewater. Graphene was prepared from sugar and normal sand as a media of adhesion in order to produce graphene coated sand media which is well known as graphene coated sand composite (GSC). Physical and chemical tests of the obtained composite were carried out and the functional groups on the surface were identified. SEM and EDX for the composite were also analyzed. Batch process was employed for the study the effect of pH, temperature, contact time and dose, for lead removal. Langmuir and frendlich models were applied. Isotherm data showed that the maximum adsorption capacity of lead was 59.17 mg/g. This fast, low-cost technology can be used to manufacture commercial filters to treat contaminated water using appropriate engineering designs.

Key words: GSC; wastewater; lead removal.

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1. Introduction

Pollution of freshwater systems by industrial and agricultural waste has become one of the key environmental problems [1]. Among all water contaminations, heavy metals ions can cause severe health problem in human and animals [2]. The sorption technique has proven to be an effective process for the removal of heavy metal ions from wastewaters [3-7]. Currently, different types of adsorbents used for the removal of heavy metals ions from water include zeolites, metallic oxides such as manganese oxides, ion exchange resins, limestone, chitosan, bagasse fly ash and activated carbon, lignite, modified carbon nanotubes [8-15]. The developments of nanoscience have received remarkable attention due to their special features [16]. Following the discoveries of fullerene and carbon nanotubes (CNTs) in earlier decades, the emergence of graphene with its combination of extraordinary physical properties has opened up an exciting new field in the science and technology of two dimensional (2D) nanomaterials [17]. Several aspects, such as high thermal and electrical conductivity, electronic properties, quantum hall effect, and application in drug delivery and DNA sensing, have been investigated in the recent past [18-20]. Several studies have been conducted on chemically synthesized graphene, as well as graphene oxide, can be anchored onto the surfaces of river sand to make effective adsorbents that remove heavy metal ions, pesticides, and natural dyes [21-26].

In the present study, we report the facile green synthesis of graphene, a versatile carbon source from common sugar and capped with sand grains and used the composite for adsorption of heavy metal.

2. Material and theory

2.1. Preparation of graphene coated sand composite (GSC)

Graphene sand composite was made through followed three steps by using sugar as a source of carbon [27]. The first step was dissolving different ratios of sugar in water and mixing it with fine sand. The mixture was heated by hot air oven for 90°C until the sugar hardens on the grains of sand. The second step was burning the sample in furnace with N₂ gas at 750°C for three hours. When the temperature reached (186 °C) which is the melting point of sugar, the sugar was converted to dark brown colored and then converted to carbon. This process was accompanied by releasing of CO gas in large quantities. The last step is activation process by strong acid (H₂SO₄). Then, the sample was filtered and washed with distilled water and dried in oven at 120°C for 2 hours.

2.2. Preparation of pb²⁺ solution

1000 mg/L stocks solution were prepared for pb²⁺ by dissolving 1.5985 of pb(NO₃)₂ in one liter of distilled water then diluted to (50 mg/L) concentration to be used in subsequent experiments. The specific concentration of pb²⁺ ions after adsorption processes were measured by using Atomic Absorption Spectrophotometer (model Novaa400).

2.3. Adsorption experiments

Adsorption of pb²⁺ was studied by GSC using different variables (pH, Temperature, Contact time and dose) to

find the optimum parameter for adsorption. A certain amount of GSC was added to 50 ml solution with a constant concentration (50 mg/L). The mixtures were shaken by a shaker (model: VRN-480). Then the Samples were put in Centrifuge (80-1 table top low, china) at 4000 rpm for 10 min to separate adsorbent particles.

The percentage of pollutant removal was calculated using the following equation:

$$\% \text{Removal efficiency} = \frac{C_0 - C_e}{C_0} * 100 \quad (1)$$

Where C_0 and C_e is the initial and final pollutant concentration (mg/L), respectively.

An adsorption isotherm is a graphical representation showing the relationship between the amount adsorbed by a unit weight of adsorbent and the amount of adsorbate remaining in a test medium at equilibrium. It maps the distribution of adsorbable solute between the liquid and solid phases at various equilibrium concentrations. Adsorption isotherms are important to describe the interaction of adsorbate molecules with adsorbent surface. In this study, Langmuir and Freundlich models were employed for the treatment of the equilibrium adsorption data for Pb^{2+} ions. The adsorption capacity of GSC was investigated in a batch reactor. The adsorbent dose and volume of solution were 3000mg and 50 ml, respectively. The adsorption capacity was calculated through the following equation:

$$\left[q_e = \frac{V_L (C_o - C_e)}{W_o} \right] \quad (2)$$

Where q_e : amount of adsorbate per mass of adsorbent, mg/g

V_L : volume of solution, L

W_o : mass of medium, g

The adsorption isotherms were obtained by plotting the weight of the solute adsorbed per unit weight of each medium (q_e) against the equilibrium concentration of the solute in the solution (C_e) (Crittenden, 1987).

3. Results and discussion

3.1. Scanning electron microscopes (SEM)

The physical morphologies and surface properties of GSC samples were examined by using scanning electron microscopy (SEM AIS2300C). EDAX was also carried out for the elemental analysis chemical or characterization of a sample nergy-dispersive X-ray spectroscopy. **Figure 1** represents SEM images of GSC and showed the formation of carbon on the surface of sand. Elementary mapping by EDAX measurement were also carried out for compositional studies it showed that the percentage of carbon (73.15%) ,oxygen (22.98%) and silicon Si (1.07%) and traces of other elements like aluminum and sulfur .

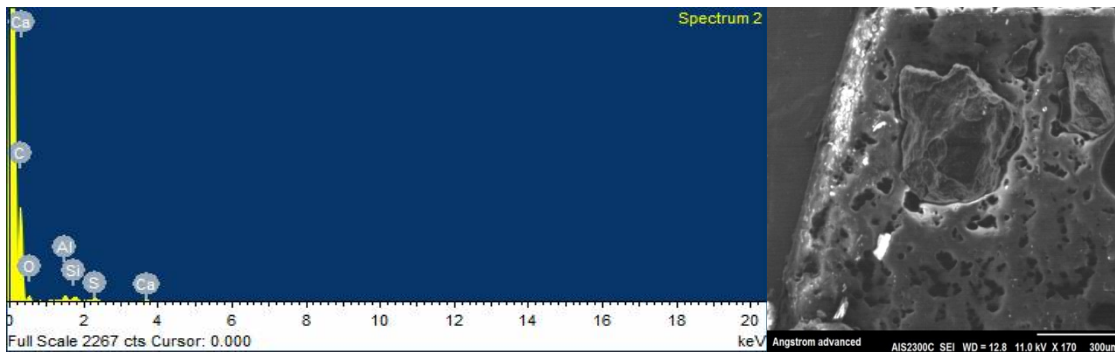


Figure 1: SEM images of GSC.

Figure 2: EDAX of GSC

3.2. Fourier Transformation Infrared Spectroscopy (FTIR)

The FTIR studies were performed to determine the various functional groups on the surface of the material (GSC) [28]. The instrument used to record the FTIR spectra by FTIR instrument with KBr as reference. The spectrum was recorded by using FTIR (WQF-510A FTIR) spectrophotometric in a spectral range of 400-4000 cm^{-1} . **Figure 3** shows the FT-IR of GSC. The spectrum of GSC shows an absorption band at 1620 cm^{-1} corresponding to the C=C stretching illustrating the structure of graphene [29]. This confirms the formation of the graphenic material from common sugar. The peaks at 1093 cm^{-1} , 3593 cm^{-1} and 795 cm^{-1} were corresponded to the C-O, O-H and C-O-C stretching that are representing the oxygen functionality present. The peak at 777 cm^{-1} was due to C-H stretching.

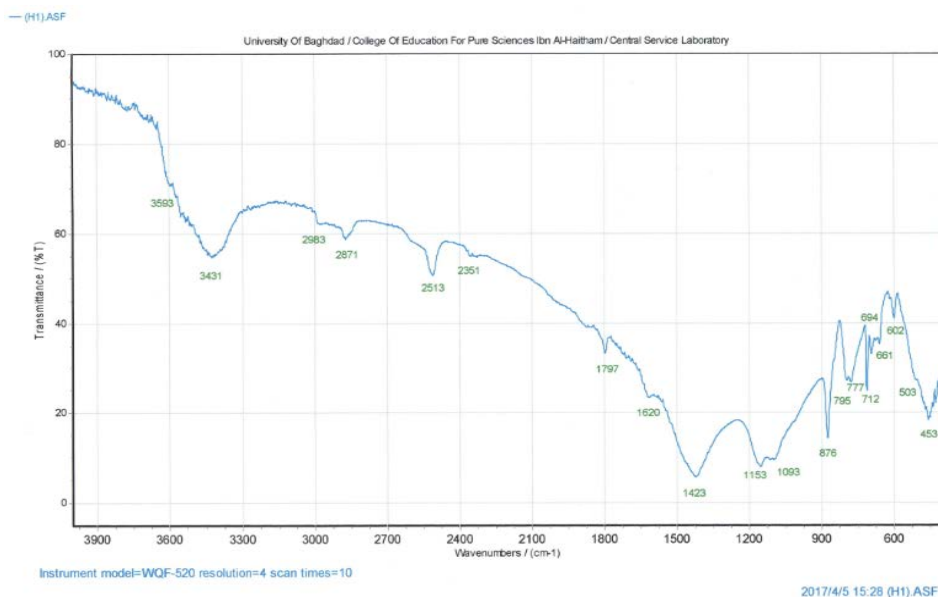


Figure 3: FT-IR analysis of GSC

3.3. Adsorption study on pb^{2+} ions by GSC

3.3.1. Effect of pH

Figure 4 shows increasing pH of solution from 2 to 4 make The removal efficiency of adsorbed $Pb^{2+}(II)$ increase from 94.26% to 97.96. The fact that the amount of Pb^{2+} removal at low pH is considered lower that may be accounted for the competition between Pb^{2+} and H^+ ions, the active sites on sorbent surface [30]. The decreasing of the amount of adsorbed Pb^{2+} above pH = 4 due to the formation of soluble hydroxyl complexes. It is assumed that OH^- in the alkaline medium effects at first on hydrolysis products of $Pb(OH)_2$, that effects on hydrolysis complexes, causes decrease the adsorption. Optimal pH = 4 which was used in further subsequent experiments

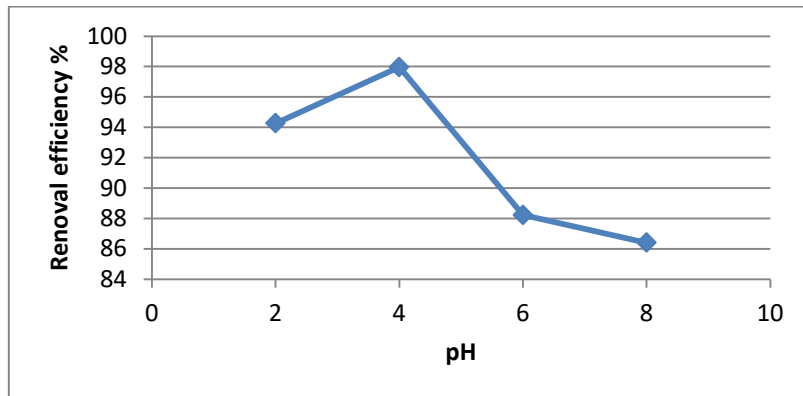


Figure 4: Effect of pH on lead removal, $C_0=50$ mg/L , $m=3$ g.

3.3.2. Effect of temperature

The observed decrease in the adsorption capacity with the increase of temperature indicated that low temperature is favorable for Pb^{2+} ions removal by adsorption using GSC. This may be due to a tendency of the Pb^{2+} ions to escape from the solid phase to the bulk phase With increasing temperature. This effect involves a physical process, which is usually associated with low adsorption heat. This means that the adsorption process is exothermic [31].

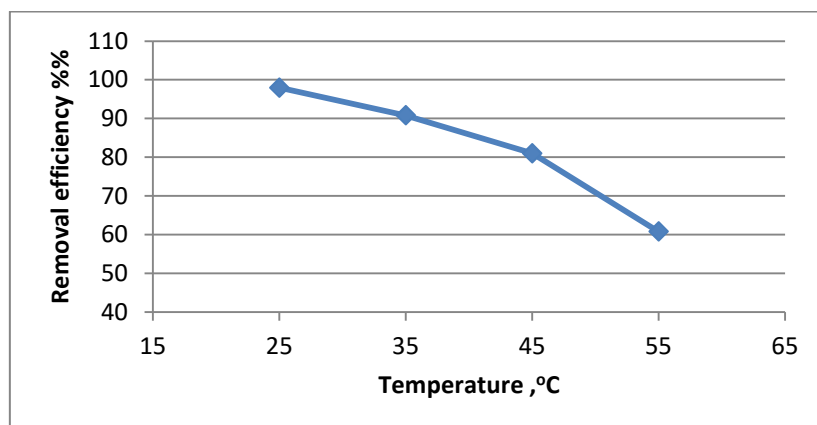


Figure 5: Effect of temperature on lead removal, $C_0=50$ mg/L, $m=3$ g, pH =4.

3.3.3. Effect of contact time

Figure 6: illustrates the effect of contact time on the adsorption of Pb^{2+} ions by GSC. It is shown that uptake is rapid at the beginning (at first 30 and 60 min). The adsorption curve is continuous until saturation and indicates the possible monolayer coverage on the surface [32]. The equilibrium time was 150 min and there are no significant change in removal after this time because of saturation of absorption sites on adsorbent.

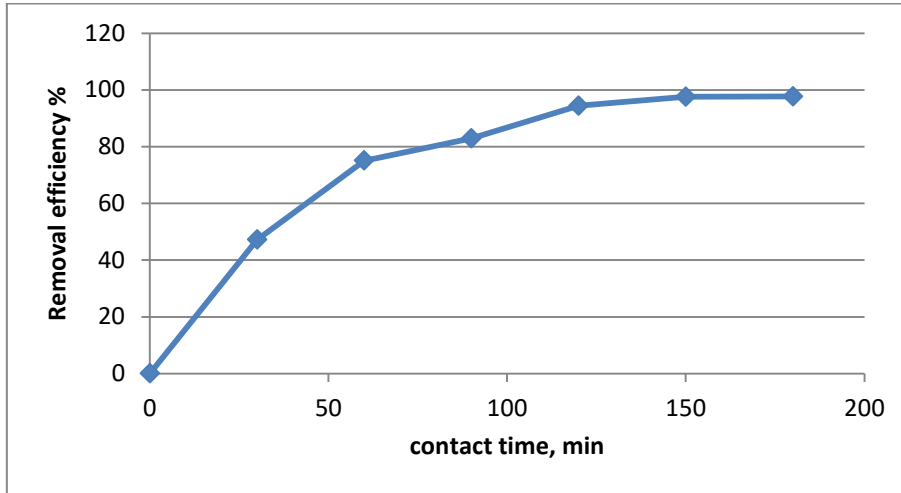


Figure 6: Effect of contact time on lead removal, $C_0=50$ mg/L, $m=3$ g, $pH=4$.

3.3.4. Effect of Dose

The effect of GSC dose on the removal percentage of Pb^{2+} was shown in **Figure 7**. It could be clearly seen that, increase in sorbent dose caused best removal of Pb^{2+} . When the sorbent dose increased from 0.5 to 4 g, the percentage of Pb^{2+} removal increased from 46.18% to 98.5%. Increase in adsorption with adsorbent dosage can be attributed to increased adsorbent surface area and availability of more adsorption sites [33].

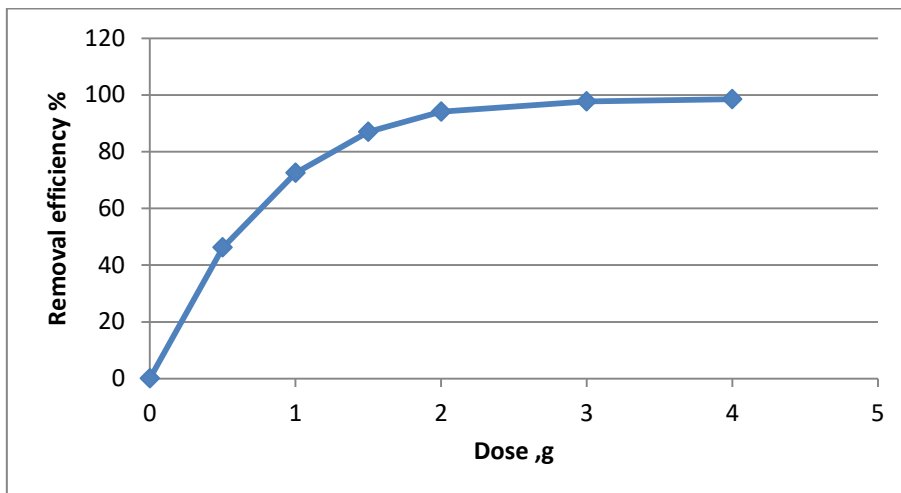


Figure 7: effect of dose at $pH=4$, temperature = $25^{\circ}C$, initial concentration = 50mg/l, contact time = 150 min.

3.4. Adsorption isotherm

Figure 8 shows the adsorption isotherm, which is considered irreversible because of its convex upward shape which is favorable. **Figure 9** and **Figure 10** showed the linear equation of Langmuir and Freundlich isotherms for adsorption of Pb^{2+} by GSC. The results showed that The maximum adsorption capacity was 59.17 mg/g of carbon .Also, RL value obtained was 0.062.

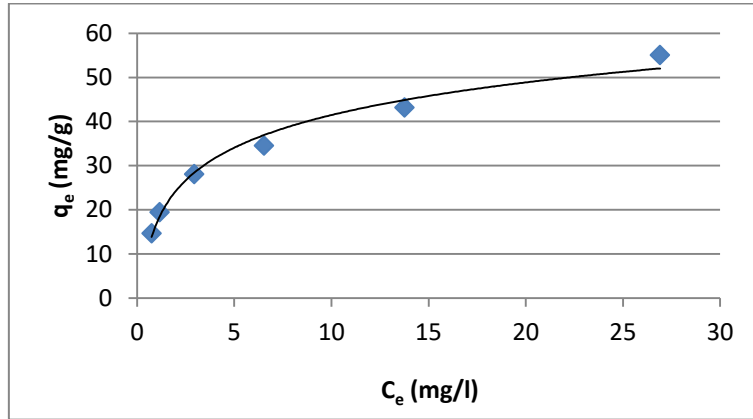


Figure 8: Relation between C_e and q_e

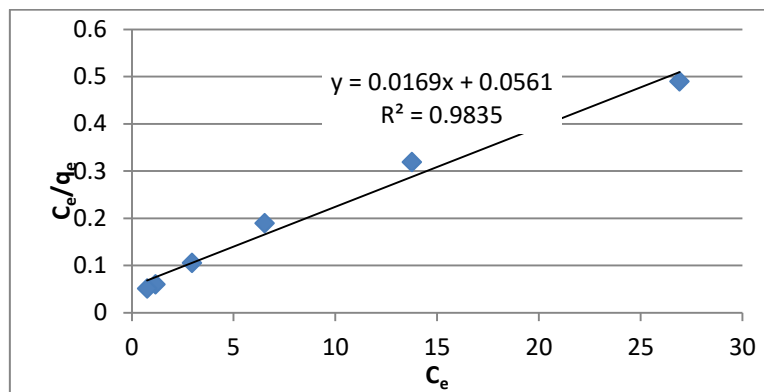


Figure 9: Langmuir plot for lead adsorption

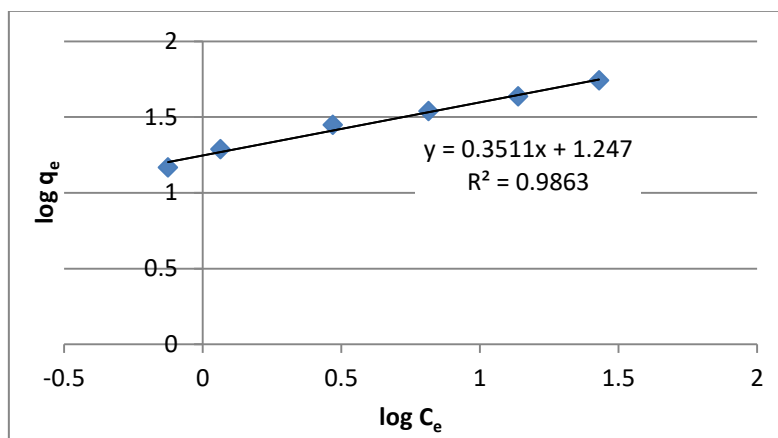


Figure 10: Freundlich plot for lead adsorption

4. Conclusion

This study explained that GSC as a low cost filter was an effective adsorbent for the removal of Pb^{2+} ions in batch and fixed-bed column. The experimental parameter were discussed in details including variation of Ph , temperature ,contact time and dose . GSC shows higher removal percent at lower pH values and temperature. At equilibrium study , The maximum adsorption capacity of lead was found 59.17 mg/g. The experimental data of breakthrough curves for Pb^{2+} were described by BDST model for finding the adsorption capacity No' ($mg\ I^{-1}$) of GSC bed.

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