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Preparation of Graphene Nanoparticle Surface Modified Metal Oxide Doped Soda-Lime Glass Composite for Application in Water Purification

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Abstract

Synthesis of the nano-adsorbent has become one of the crucial areas of research emphasizing on environmental protection and water remediation issue. This work involves the production of Graphene Oxide Soda-Lime Glass Composite(GOGC), with excellent adsorption capacities as filter media, prepared using simple one pot hydrothermal reaction (post activation of the soda lime glass) followed by thermal ageing of the composite from colored glass containing metal oxides and Graphene oxide. Characterization of the novel adsorbent has been done using Scanning Electron Microscopy and Fourier-transform infrared spectroscopy. Batch experiments were performed to quantify the adsorption kinetics and adsorption capacities of the nano- composite Performance of the synthesized composite was evaluated for the removal of organic components and adsorption of As (V). The synthesized nano-composite exhibited higher dye removal efficiency, better adsorption capacities for heavy metals, removal of turbidity and organic matter. This improved performance may be due to better synergistic molecular interactions of metal oxides in the soda lime glass, the functionalized groups in Graphene Oxide and greater surface area of the GOGC. Results have illustrated high potential of the newly synthesized nano adsorbent material for ground water and wastewater treatment purposes.

Keywords

Nano adsorbent, Graphene oxide, Glass composite

Introduction

Remediation of water pollution and environmental protection primarily relies on using technologies such as adsorption, absorption, size exclusion filtration and bioremediation for efficient removal of pollutants from water. Synthesis of nano-adsorbent has been understood to be a crucial area in this regard owing to the high surface area to volume ratio, which often results in higher reactivity, contrasting to traditional approaches nanomaterial-based adsorbents can be functionalized with specific functional groups for remediation of targeted pollutants of interest, further the physical properties(such as porosity, morphology, etc.) of such nano-adsorbents can be tuned which will amplify the remediation of the pollutant [1]. Carbon-based nano-adsorbents has been widely explored in the past, where Graphene, Graphene Oxide and Carbon Nanotubes were found to have excellent applications in water purification [2]. Graphene Oxide consisting of hydroxyl, carboxyl and epoxy functional groups has high negative charge thickness [3] and thus can be used as metal cation exchange, based on the presence of the ionic carboxyl group which results in ion-exchange with organic as well as metal positive species. Despite the high pollutant removal efficiency of Graphene oxide, the separation of the adsorbent from the treated water and its consequent reusability remains to be a challenge [4]. Soda-Lime Glass which is both hydrophilic and has a high surface to volume ratio serves as an excellent option for the substrate of the nano-adsorbent. The ordered porous silica network is suitable for selective adsorption [5]. The modifier network of the Soda-lime glass which contains Na+, Mg^{2+} and Ca^{2+} can readily be replaced by higher electronegative species [6]. Thus, this study is intended to utilize the aforementioned adsorbents, which apart from showing high adsorptive capacities, has great regenerative properties as well as can be easily isolated after the adsorption process.

Materials and Method

Waste brown and green colored Soda-Lime glass bottles were mechanically processed into a fine powder, the Fe-O doping in the glass network, imparted the glass green and brown color and treated in alkaline and acidic media. Based on the aforementioned background Graphene oxide-Soda Lime glass composite adsorbent synthesized using pretreated soda-lime glass and aqueous dispersed Graphene Oxide in a one-pot hydrothermal process, followed by thermal ageing. The composite was washed with ethanol and dried at 60 °C (Figure 1). Further batch adsorption studies were performed with As(V) and Methyl blue dye. The adsorbent was further characterized using FTIR, Scanning Electron Microscopy and Point of Zero charge.

Figure 1: GO-SLS Composite.

Results and Discussion

Characterization of the soda-lime glass composite

Characterization of the synthesized adsorbent was done by its morphology analysis using microscopic imaging. Figure 2a represents the morphology of the adsorbent surface which is a corroded surface with high surface roughness. The rough with fine flaky bulges might be sodium silicate and calcium silicate in crystalline form due to the hydrothermal treatment [7]. In figure 2b, the presence of Graphene Oxide nanoparticles grafted on the surface of Soda Lime glass particles has been manifested.

The iso-electric point of the GO-SLS Composite was observed to be at a pH of 4.65 whereas 1.75 for that of SLS(Figure 3), the nature of the relation between change in pH vs ΔpH can be attributed to the main constituent of the Activated Glass, SiO2 and the functional groups of Graphene Oxide, which demonstrates the importance of surface oxides, the interaction of these species with water results in the development of hydroxyl groups at the surface, thus an amphoteric nature of the surface can be concluded. The development of the surface charges on the oxides and the hydroxides may be explained in terms of the following equations(Equation 1 & Equation 2) as reported elsewhere [8]

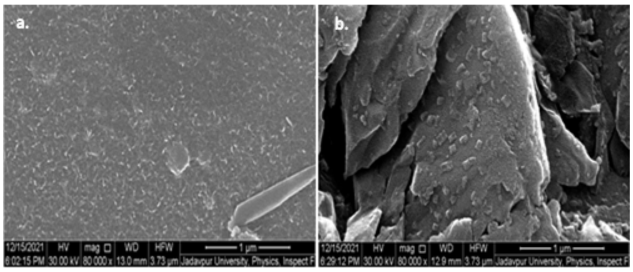

Figure 2: Scanning Electron Microscopy Image of a) Activated Glass b) GO-SLS composite.

$$R - OH_2^+ \leftrightarrow R - OH + H^+ \quad \text{(Equation 1)}$$

$$R - OH \leftrightarrow R - O^- + H^+ \quad \text{(Equation 2)s}$$

Where R is Glass surface

Thus with increasing pH, it can be correlated that the negatively charged species on the glass surface increases, hence it would be safe to conclude that a negative zeta-potential is developed in the pH range of 5-12, and is pH-dependent, which conforms to previous literature as reported for the Zeta-Potential of Soda-Lime glass structures [9]. The change in Iso-electric point can be attributed to the additional negative

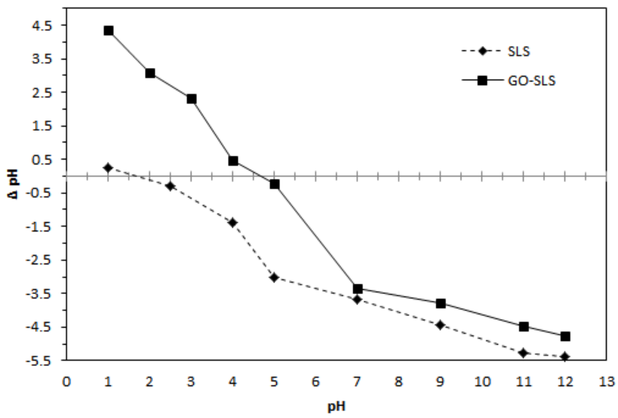

Figure 3: Change in pH vs pH, depicting the isoelectric point of SLS and GOSLS composite.

functional groups of GO in GO-SLS.

FTIR analysis (Figure 4a) of the GO-SLS composite, Soda Lime Glass and Graphene Oxide were done and characteristic broad bands of tetrahedral Si-O-Si bonds were observed in both SLS and GO-SLS. Further, peaks corresponding to FeO were observed in the fingerprint zone both in GO-SLS and SLS. Characteristic Graphene oxide peaks corresponding to the Carboxyl Group and peaks corresponding to C-O-C stretching were also observed in the GO-SLS composite. The small peak (Figure 4b) near 1400 cm^{-1} was evidence of the presence Si-Phenyl bonds, which represents the incorporation of the GO on the Soda Lime Glass.

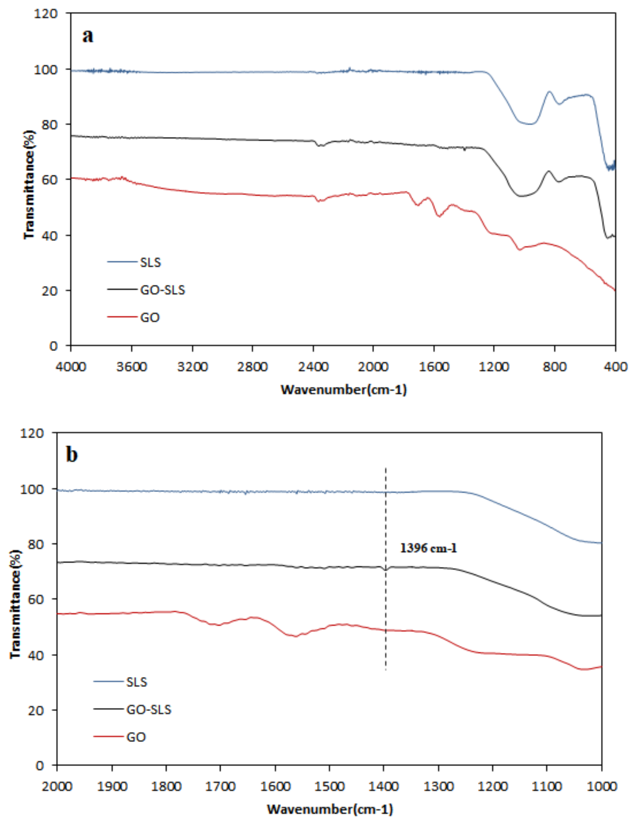

Figure 4: FTIR of SLS, GO-SLS and GO a: whole spectra b: Peak corresponding to Si-Phenyl bond.

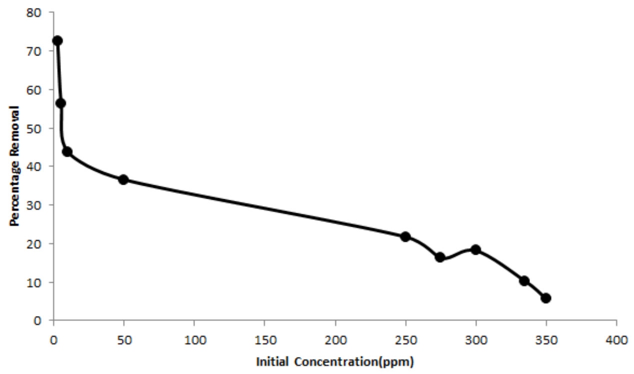

Figure 5: Dye: Effect of contaminant concentration on percentage removal.

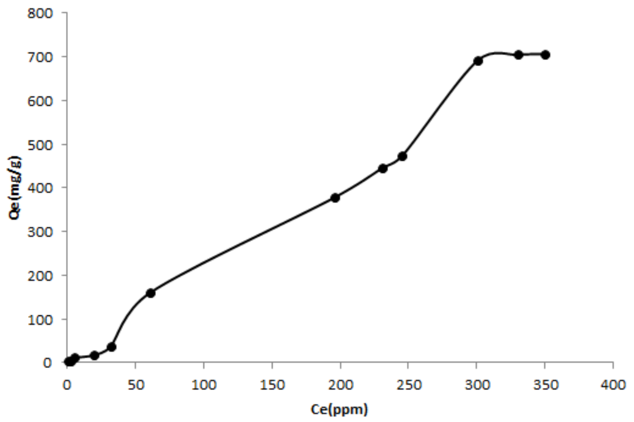

Figure 6: Adsorption Isotherm for Removal of Methylene Blue.

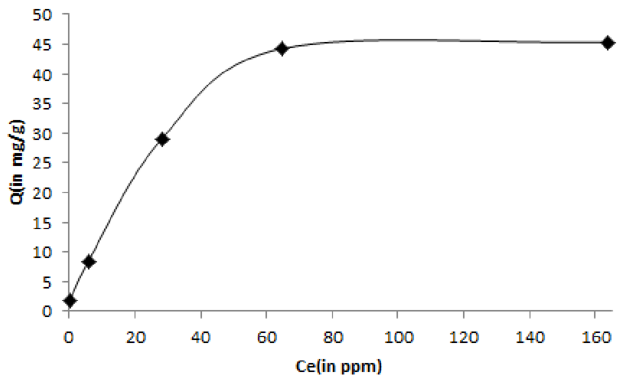

Figure 7: Adsorption Isotherm for Removal of As(V).

Studies on the adsorption characteristics of soda-lime glass composite

The Methyl blue adsorption studies have shown high removal capacity of cationic dye, Methyl blue, the adsorption capacity of the GO-SLS composite was observed to be 708 mg/g (as evident from Figure 5) and is very efficient in the removal of Methyl Blue dye at low contaminant concentrations(removal efficiency being 72%) conditions as evident from the figure 6.

Adsorption phenomenon of As (V) was studied at various As (V) concentrations, an adsorbent dose of 0.5 g/L was used, and a fixed temperature of 32 °C ± 5 °C and a pH of 7 ± 0.5 was maintained. The experimental value of Q_{max} was calculated to be 44.6 mg/g (Figure 7), which was found superior to both GO and Soda Lime glass.

The influence of the initial concentration of As(V) ion on adsorption at the temperature of 32 °C ± 5°C was experimented. It was observed that the adsorption amount of As(V) was enhanced with the initial As(V) concentrations and then gradually reached a plateau, which was due to the saturation adsorption on the active sites of the GO-SLS composite. Langmuir (Figure 8) and Freundlich (Figure 9) equations were used to fitting the isotherm data, the results obtained clearly show that the Langmuir equation was well fitted for the adsorption isotherm due to a higher correlation value than that of the Freundlich equation. In addition to that, the adsorption capacity of Qm acquired by the Langmuir theory was 52.91005 mg/g, which is very closer to the obtained experimental value. These obtained results suggested monolayer adsorption of As(V) by GO-SLS adsorbent.

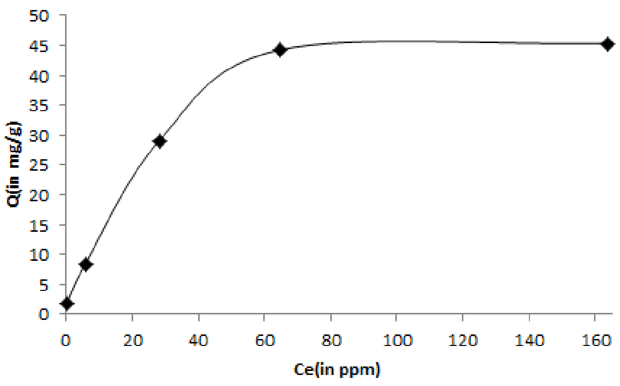

Figure 8: Langmuir Isotherm For the Adsorption of As(V).

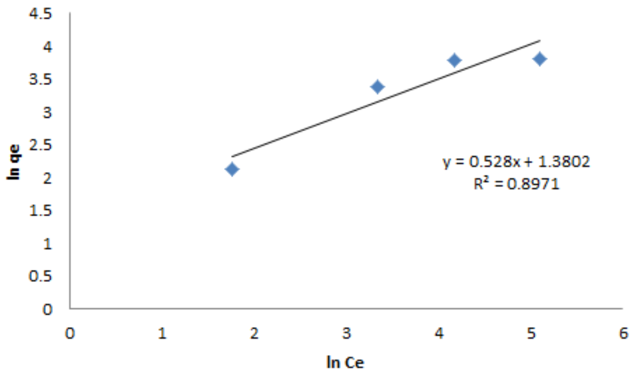

Figure 9: Freundlich Isotherm For the Adsorption of As(V).

On examining the dependence of percentage removal of As(V) on initial concentration(Figure 10), it was found that the GO-SLS composite exhibits maximum removal(89%) of Arsenic at lower concentrations 1ppm gradually decreasing to 18% for 200 ppm. This is a very important observation to note, as high removal at low concentrations makes the GO-SLS optimal for real-time use where As concentrations in underground mostly remains below 1ppm.

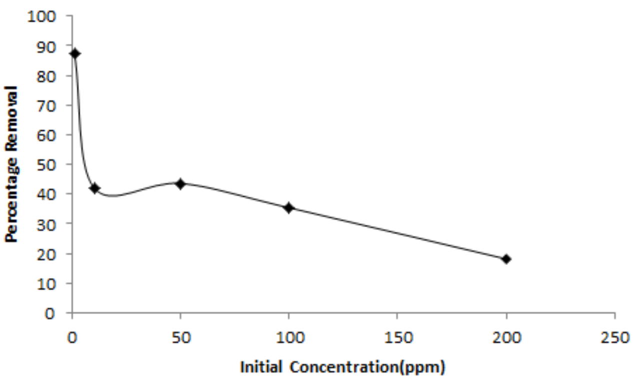

Figure 10: Effect of initial concentration on percentage removal.

Conclusion

The Graphene Oxide-Soda Lime Composite was found to be a suitable nano adsorbent for removal of As (V), at low concentrations, which is novel as most presently available nanoparticles are inefficient in remediation of Arsenic at low concentrations (naturally occurring Arsenic pollution in underground water). The GO-SLS composite shows 89% removal at 1ppm concentrations of As(V) and illustrates to have a maximum adsorption capacity of 44.6 mg/g, for cationic dyes like Methyl Blue GO-SLS composite exhibit a high maximum adsorption capacity of 708 mg/g. The adsorption data for Arsenic removal fits well with Langmuir isotherm models, suggesting the formation of a monolayer. Optical Imaging depicts the surface morphology of the activated glass and the effect of the hydrothermal treatment, the zero point charge of the GO-SLS was noted to be at a pH of 4.65 which suggest applicability in the wide range of pH. The high performance is attributed to better synergistic molecular interactions of metal oxides in the soda-lime glass, the functionalized groups in Graphene Oxide and the greater surface area of the GO-SLS Composite. These results have illustrated the high potential of the newly synthesized nano adsorbent material for arsenic remediation in groundwater.

Conflict of Interest

There is no known conflict of interest between the content of the research and any other available work on the subject.

Acknowledgements

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