PREPARATION OF INVERSE SPINEL MAGNETITE GRAPHENE OXIDE FROM SUGAR FOR THE REMOVAL OF PB (II) FROM AQUEOUS SOLUTIONS

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Abstract- The present study has been described about synthesis of Inverse Spinel Magnetite graphene oxide from sugar using an eco-friendly method. It is a simple and efficient method for magnetite graphene oxide preparation. FT-IR, XPS and XRD confirmed that the material is graphenic in nature. It effectively removes contaminants from water. Here, we used Pb(II) for this application. The batch adsorption studies conclude that the adsorption of Pb(II) was monolayer chemisorptions on the homogenous surface of Magnetite graphene oxide (MGO), subsequently follows pseudo-second-order kinetics. Finally the results concluded that the prepared MGO was easily recovery and re-usable for removal of heavy metals. The adsorbent was eco-friendly, cost-effective technique for the commercial water tenement.

Keywords- Sugar based graphene oxide; Magnetic graphene oxide; Pb(II) removal; Adsorption.

I. INTRODUCTION

Graphene is a single two-dimensional (2D) layer of carbon atoms and its oxide form, graphene oxide (GO), have large specific surface areas and good biocompatibility. The amorphous nature of graphene oxide generated by the chemical fabrication method, understanding the atomic structure and its effects on the oxidation process is still a big problem. Recently, many studies have dealt with the oxidation of graphene by ozone and O ions [1-5]. However, the recovery of loaded graphene or graphen oxide has been challenged. graphene based nanocomposites has become a hot topic of research due to their enhanced functionalities that cannot be achieved by either component alone, and therefore holds a great promise for a wide variety of applications in catalysis, surface enhanced Raman scattering, biomedical fields, drug delivery and removal of contaminants from wastewater [6-11]. In the present study, synthesized magnetic graphene oxide (MGO) by chemical precipitation and was well characterized with suitable spectroscopy techniques in order to use for adsorption removal of Pb(II) from aqueous solutions.

II. PREPARATION OF SUGAR BASED GRAPHENE OXIDE AND MAGNETITE GRAPHENE OXIDE

As shown in the Fig.1, GO was prepared by programmed thermal heating of sugar at N_2 atmosphere for 5 h. At first the sugar was kept in silica crucible and heats it up to 200^{o}C with in 30min and then kept for 1h, after that increased temperature from 200 to 750^{o}C within 30 min and held that temperature for 3h to get sugar based graphene. The prepared graphene at pH 1.0 was exposed to ozone gas which is generated by ozone generator as shown in Fig.1.

After ozonation, the product, GO was collected by filtration and dried in hot oven at 65^{o}C.

First,0.1g GO was dispersed in 100 mL water by sonication for 1 h to transform the carboxylic acid groups to carboxylate anions. Then add 0.5g $Fe(III)Cl_3{\cdot}6H_2O$ and 0.183g $Fe(II)Cl_2{\cdot}4H_2O$ (Fe^{3+}: Fe^{2+} was 2:1) were dissolved in 25 mL of water solution at room temperature with vigorous stirring. Then, adjust the solution pH (> 12) and heat the solution at 85 ^{o}C for 45min results a black precipitate in the solution and was separated by use external magnetic field. The obtained product was washed several times with de-ionized water, dried at 60 ^{o}C under vacuum for 12h.

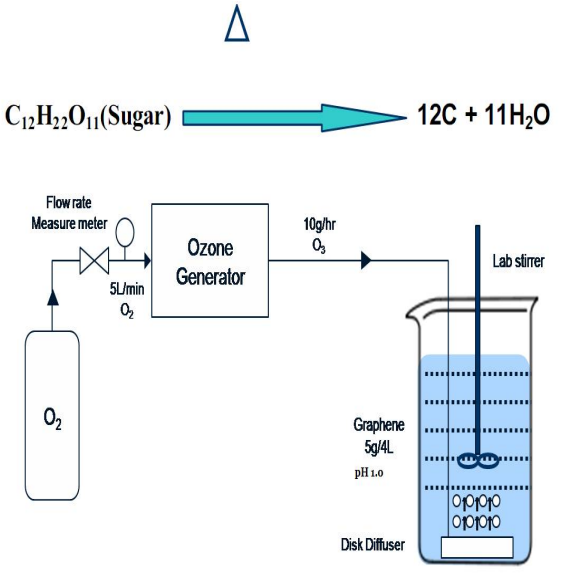


Fig.1. Schematic representations of preparation GO from Sugar.

III. RESULTS AND DISCUSSION

The prepared sugar based MGO confirmed by characterizing using FT-IR XRD, and XPS spectroscopy techniques. From FT-IR spectrum of MGO it was clearly observed the presence of -OH, C-O , C=O ,C=C-H along with M-O functions were confirmed the formation of MGO.

The BET results were indicated the high surface area MGO ($622m^2/g$) which is more favorable for adsorption. The morphology and surface composition was evaluated by SEM and SEM-EDS where clearly observed the pours surface morphology.

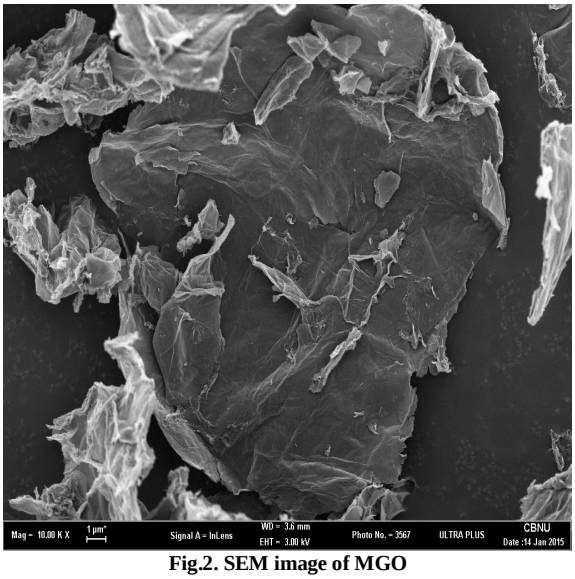


Fig.2. SEM image of MGO

The prepared Magnetite Graphene oxide(MGO) surface properties was tested by batch adsorption of Pb(II) from aqueous solution. In this order to evaluate sorption characteristics of sugar based MGO, the factors affected on the sorption of Pb(II) was tested. The sorption of Pb(II) is plotted in as a function of contact time ranging from 5 min to 24 h at pH 5.0, initial concentration of Pb(II)10mg/L and 0.4g/L dosage . It can be seen that the sorption was rapid and a contact time of 120min is enough to reach equilibrium. The adsorption kinetics results concluded that 99% removal of Pb(II) and it addressing the rate limiting pseudo second order on heterogeneous surface of Magnetite Graphene oxide.

In order to evaluate sorption capacities of sugar based MGO for Pb(II) applied Langmuir and Freundlich isotherm models .the result concluded that the correlation coefficients value of 0.99 for uranium that the Langmuir equation fits the experimental data better than the Freundlich model. The maximum adsorption capacities (Q_{max}) of Pb(II) ions on MGO at 298K are 65.69 mg g-1 was achieved in the present study .The data also indicate a monolayer adsorption occurred at binding sites on the surface of the adsorbent.

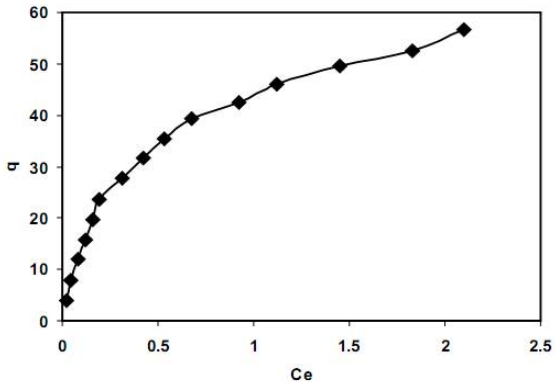


Fig.3. Adsorption isotherm of Pb(II)) on to sugar based MGO.

TABLE 1. Pb(II) isotherms with sugar based MGO.

Langmuir			Freundlich		
q_{max}, $mg\,g^{-1}$	K_L, Lmg^{-1}	R^2	K_F, mgg^{-1} $(Lmg^{-1})^{1/n}$	n	R^2
65.69	5.45	0.993	32.65	2.05	0.941

ACKNOWLEDGMENT

This work was supported by the Korean Ministry of the Environment as part of the "GAIA project (2014000550003)" with additional supported from the "Research Grant–2015" of Kwangwoon University, Seoul, Korea.

REFERENCES

[1] K. Geim and K. S. Novoselov, "The rise of graphene," Nat. Mater., 2007, vol. 6, pp. 183−191.

[2] C. Gomez-Navarro, J. C. Meyer, R. S. Sundaram, A. Chuvilin, S. Kurasch, M. Burghard, K. Kern and U. Kaiser, "Atomic structure of reduced graphene oxide," Nano Lett., 2010, vol. 10, pp.1144-1148.

[3] A. Sinitskii and J. M. Tour, "Patterning Graphene through the Self-Assembled Templates: Toward Periodic Two-Dimensional Graphene Nanostructures with Semiconductor Properties," J. Am. Chem. Soc., 2010, vol.132, pp.14730-14732.

[4] S. Huh, J. Park, Y. S. Kim, K. S. Kim, B. H. Hong, and J.-M. Nam, "UV/ozone-oxidized large-scale graphene platform with large chemical enhancement in surface-enhanced Raman scattering" ACS Nano, 2011, vol.5 (12), pp. 9799–9806.

[5] L. P. Lingamdinne, J. R. Koduru, Y. L. Choi, Y. Y. Chang, J. K. Yang Studies on removal of Pb(II) and Cr(III) using graphene oxide based inverse spinel nickel ferrite nano-composite as sorbent, Hydrometallurgy, 2015: http://dx.doi.org/10.1016/j.hydromet.2015.11.005

[6] J. A. Yan and M. Chou, "Oxidation functional groups on graphene: Structural and electronic properties," Phys. Rev. B, 2010, vol. 82, p. 125403.

[7] F. Belloni, C. Kütahyali, V.V. Rondinella and A. Mangione, Can carbon nanotubes play a role in the field of nuclear waste management, Environ. Sci. Technol., 2009, 43, 1250-1255.

[8] S. Chen, J. Hong, H. Yang, and J. Yang, Adsorption of uranium(VI) from aqueous solution using a novel graphene oxide-activated carbon felt composite, J. Environ. Radioactivity, 2013, 126, 253-258.

[9] P. J. Lebed, J.-D. Savoie, J. Florek, F. Bilodeau, D. Larivière, and F. Kleitz, Large Pore Mesostructured Organosilica-

Phosphonate Hybrids as Highly Efficient and Regenerable Sorbents for Uranium Sequestration, Chem. Mater., 2012, 24, 4166−4176.

[10] Y. Yao, S. Miao, S. Liu, L.P. Ma, H. Sun and S. Wang, Synthesis, characterization, and adsorption properties of magnetic Fe_3O_4@graphene nanocomposite, Chem Eng. J., 2012, 184, 326-332.

[11] Y. Zhang, B. Chen, L. Zhang, J. Huang, F. Chen, Z. Yang, J. Yao, Z. Zhang, Controlled assembly of Fe_3O_4 magnetic nanoparticles on Graphene oxide, Nanoscale, 2011. 3, 1446-1450.

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