

Synthesis and Characterization of Graphene Oxide-Melamine (GO-M) Nano Composite and Poly (Styrene –Alternative – Maleic Anhydride) -Melamine (SMA-M) Using for Removing Lead (II) Ions from Aqueous Solutions.

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Abstract

In this study a new method by using graphene oxide (GO) Nano sheets-melamine composites and derivation of Poly (Styrene –alternative- Maleic Anhydride) (SMA), (SMA+Melamine) (SMA-M) were presented as Sorbents for the elimination of Lead ions from aqueous solutions. The adsorbents have the sufficiency to adsorb the Lead ions. Through the immobilization of melamine onto GO Nano sheets, the desired composite was synthesized and identified by field emission scanning electron microscopy (FE-SEM), Fourier transform infrared (FT-IR) spectroscopy and absorption atomic spectroscopy (AAS) techniques. The various experimental parameters such as pH and concentration of the aqueous solution of Lead (II) ion, the content of the Lead (II) ion the grapheme oxide-Melamine and SMA-M have been optimized. It was shown that the uptake efficiency of Lead (II) ion considerably increased after immobilization of Melamine on the GO Nano sheets and SMA.

Keywords

Graphene Oxide; Elimination; Nano Sheets; Melamine; Adsorbents; Nano Composites.

1. INTRODUCTION

Elimination of heavy metal pollutants from the environment has been one of the basic explores in the recent few years. An Aqueous environment for many industries such as metallurgy, mining, chemical manufacturing and battery manufacturing industries include many kinds of toxic heavy metal ions [1]. Lead impresses the physical conditions of steel, alloys and high innocence metals. It is highly toxic to human and animals and reasons environmental problems. Very low amounts of lead are attendant in different matrices, such as plants, soils, food and water. It is, so, very significant to improve sensitive, selective, speedy and economical manners for quantitative measuring of its trace values. Spectrophotometric styles are simple, inexpensive and economical but are not suitable sensitive and selective. Graphite furnace atomic absorption spectroscopy, neutron activation analysis, , direct current plasma atomic emission spectrometry, inductively coupled plasma-atomic emission spectroscopy and mass spectrometry probably applied, but the needed tools are expensive, day-to-day maintenance cost is high and different types of inward interferences emerge. Lead is a highly toxic heavy metal that

causes human health problems and environmental pollutions, and is also more toxic even after from Mercury and arsenic [2]. Adsorption is purposed to be one of the most operative and economical remedial procedures for the Lead elimination of wastewaters, rivers and other aqueous solutions. Nano science and nanotechnology were the first treatment with the synthesis, characterization, exploration and exploitation of nanomaterial. Carbon, one of the most common atoms in the world, occurs naturally in many forms and as a component in countless substances which are called allotropes of carbon. Graphene, a “wonder material” is the world’s thinnest, strongest, and stiffest material, as well as being a great conductor of heat and electricity. It is the basic building block of other significant allotropes [3]. GO is of the excellent interest due to its cheap, comfort access, and splay might convert to graphite. Scalability is also a much considered trait. GO is the graphite that has been oxidized to intersperse the carbon layers with oxygen molecules, and then reduced, to separate the carbon layers completely into individual or few layer graphene. GO is effectively a byproduct of this oxidation as when the oxidizing agents react with graphite, the inter planar spacing between

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the layers of graphite is increased [4]. The completely oxidized compound can then be dispersed in a base solution such as water, and GO is then produced. A large number of oxygen-containing functional groups have been introduced onto both sides of a single graphite sheet (namely, graphene). The implantation of functional groups overcomes the inter sheet van der Waals force and enlarges the interlayer spacing [5]. The sheets in such an expanded structure are then easily pulled open using an external force such as sanitation [6]. The expanded graphite is exfoliated into multi-layered or even single-layered sheets. Generally, the oxidized graphene sheets, namely, GO, acquire multiple defects and the degree of the defects is subject to the additive amount of oxidant and the oxidizing time [7]. GO is produced by the oxidative treatment of graphite by one of the principle methods developed by Brodie, Hummers or Staudenmeier. In this study adsorption of SMA-M was compared with GO-M that SMA and GO was modified with Melamine [9].

2. EXPERIMENTAL

2. 1. Chemicals

All reagents such as Melamine, Styrene, Maleic Anhydride, H_2SO_4 , NaNO_3 , H_2O_2 , KMnO_4 , other organic and inorganic compounds were prepared from analytical reagent grade chemicals supplied from Merck (Darmstadt, Germany), except $\text{Pb}(\text{NO}_3)_2$ which was purchased from Aldrich. The 1×10^{-3} M Lead (II) salt standard stock solution was prepared by dissolving 0.0962 g of $\text{Pb}(\text{NO}_3)_2$ in a 250 ml volumetric flask and diluting to the mark with distilled water. Lower concentrations were prepared by serial dilution of the stock solution with phosphate buffer; pH 8 Universal buffer solutions (0.1 M) were prepared from phosphoric acid solutions and sodium hydroxide solutions and adjusting pH to the desired value (0.1M). The final pH was adjusted with the addition of a sodium hydroxide.

2. 2. Synthesis of graphen oxide Nanosheets(GO)

GO was provided using a modification of Hummers and Offeman's method [10]. Briefly in a typical reaction, 8 g graphite, 8 g NaNO_3 , and 200 mL H_2SO_4 were stirred together in an ice bath. KMnO_4 (64 g) was slowly added while stirring, and the rate of addition was controlled to prevent the mixture temperature from exceeding 15°C . The mixture was then transferred to a 35°C water bath and stirred for about 80 min, forming a thick paste. Subsequently, 400 mL de-ionized water was added little by little and the temperature was increased to 90°C . The mixture

was further remedied with 80 mL 30% H_2O_2 solution. The solution was then filtered and washed with deionized water until the pH was 6 and dried at 62°C under vacuum [11].

2. 3. Synthesis of covalently attached Melamine GO hybrids (GO-M)

Briefly in a typical reaction, a mixture of 20 mg GO and 50 mg Melamine was taken in a 100 mL round bottom flask and 3 mL of triethylamine and 10 mL of DMF were added and heated to 80°C for 64 h under a nitrogen atmosphere. After the reaction, the solution was cooled to room temperature, and then poured into 250 mL diethyl ether to precipitate the product. The precipitate was collected by centrifuging at 6000 rpm for 20 min. The supernatant which contained dissolved Melamine was discarded and the precipitate was washed thoroughly. After adding another 100 mL of diethyl ether, the mixture was sonicated for 4 min and then centrifuged at 6000 rpm for 30 min to collect the GO-M. Finally, the precipitate was washed with (CHCl_3) three times following the above procedure. The schematic illustration of GO-M was shown in Fig. 1 and Fig. 2.

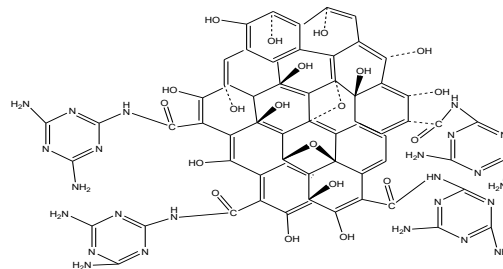


Fig. 1. The structural formula of Grapheme Oxide - Melamine (GO-M).

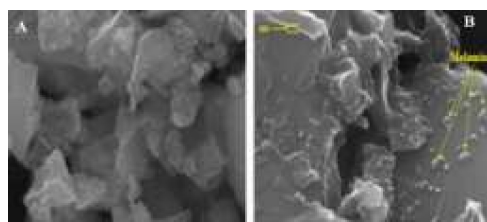
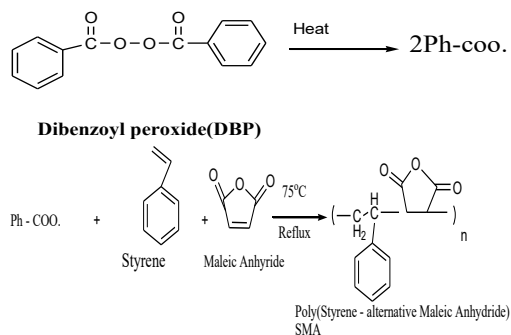


Fig. 2. SEM images of A: GO Nano sheets and B: GO-Melamine composite.

2. 4. Synthesis of poly (Styrene – alternative-Maleic Anhydride) (SMA)

The SMA polymer was prepared by the free-radical polymerization of Maleic Anhydride and Styrene at 71°C in the presence of benzoyl Peroxide as an initiator. In this procedure, 2.1 g (19.2 mmol) of pure Styrene and 1.88 g (19.2 mmol) of Maleic Anhydride at a molar ratio of 1:1 in were dissolved 50 mL THF. Then 0.018 g

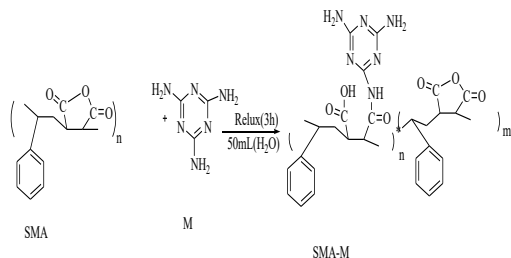
(0.768 mmol) of benzoyl peroxide was added to mixture as an initiator. The reaction mixture was refluxed for 7 h at 71 °C under inert gas in the presence of ultrasonic irradiation along with vigorous stirring bar in THF. The precipitations were completed by the addition of n-hexane as a non-solvent and were separated by filtration and washed by n-hexane several times. The product was dried in a vacuum oven for 23 h at 62 °C (yield = 97.5%). The SMA polymer was prepared as reported previously. The synthesis reactions of poly (SMA) are presented in Scheme (1).



Scheme 1. Structure of polymer (SMA).

2. 5. Preparation of SMA modified with melamine (SMA-M)

For a synthesis of the grafted SMA polymer, 1.0 g (5.0 mmol) of SMA polymer and 0.63 g (5.0 mmol) of Melamine at a ratio of 1:1 were poured into a flask. Then, 0.5 ml (4.0 mile) of triethylamine (TEA) as a catalyst and 50 mL of water as a solvent were mixed in a three necked, round-bottom flask equipped with a condenser, magnetic stirring bar, inlet and outlet for inert gas, and ultrasonic irradiation probe [12]. The reaction mixture was refluxed under these conditions for 3 h. The precipitations were completed by the addition of n-hexane as a non-solvent and were separated by filtration and washed by n - hexane several times. The product was dried in a vacuum oven for 24 h at 60 °C (yield = 98%). The synthesis process of the first chelating resin is shown in Scheme (2) and FT -IR Spectrum of the SMA-M polymer is shown in Fig. 3.



Scheme 2. Reaction for the formation of modified SMA with Melamine (M).

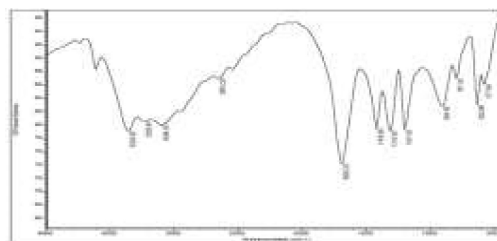


Fig. 3. FTIR Spectrum of the GO -Melamine.

2.6. Structural and spectroscopic characterization of GO-M and SMA-M

The FTIR spectra were recorded using KBr plates in the range 500–4000 cm^{-1} using a Nicolet 6700 FT-IR spectrometer. Fig. 2 and show the typical FTIR spectrum obtained for GO, Go-M and SMA-M. The most characteristic features are the broad, intense band at 3434 cm^{-1} (O–H stretching vibrations) and the bands at 1731 cm^{-1} (C=O stretching vibrations from carbonyl and carbamide groups), 1672 cm^{-1} (skeletal vibrations from oxidized graphitic domains), 1386 cm^{-1} (C–OH stretching vibrations), and 1031 cm^{-1} (C–O is stretching vibrations). The appearance of the peaks at around 2914 cm^{-1} is ascribed to the aromatic stretching vibrations of C–H bonds of GO. After covalent functionalization with Melamine, a new peak appears at 1588 cm^{-1} corresponding to the C=C vibrations of Melamine and the peak of the C–O stretches vibration shifts to 1114 cm^{-1} , and broadens. The peak present at 1719 cm^{-1} is assigned to the bending vibration of the C=N of the Melamine ring. The disappearance of the peak at 2370.78 cm^{-1} (C=O from CO_2 stretching vibrations) clearly indicates that in GO – Melamine composite, the Melamine molecules are covalently bonded to the GO via amine linkage. The SEM image (Fig. 2) shows that few layered GO is formed, although the SEM image does not surmise the layer numbers of the GO Nano sheets precisely. Pure graphite presents ordered flakes of a 2–4 μm range. The scanning electron micrograph for the GO–Melamine hybrid materials (Fig. 2B) displays that a homogeneous system with a micrometer order of magnitude was gained. On functionalization, GO presents a three-dimensional network of randomly oriented sheet like structures with a wrinkled texture and hierarchical pores with a wide size distribution [13–15].

2. 7. Adsorption experiments

To optimize the adsorption efficiency, adsorption experiments were performed at various pH levels of the aqueous solution of Lead (II) ion. All experiments were run in triplicate. Experiments were carried out using 1 g/L of adsorbents each time 0.025 g of adsorbents' mass were added to 25 mL of deionized water in a conical flask). The

pH-effect tests were initially adjusted with aqueous media of acid and base (0.01 M H_3PO_4 and NaOH). Buffer solution is applied to control the pH (Fig. 4). Considering Lead precipitation in higher concentration, the equilibrium adsorption tests were conducted at pH 6, 8 in batch modes. The process of removing metal was performed. The Lead solution was shaken on an orbital shaker at 180 rpm for a specified period of time.

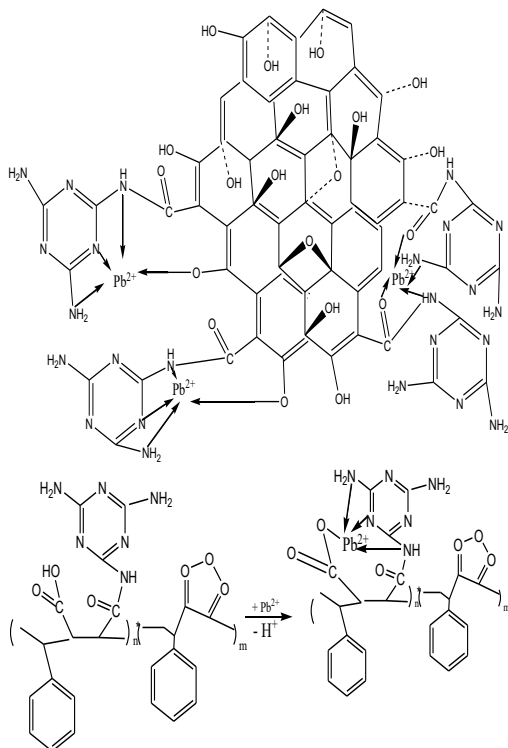


Fig. 4 . Adsorption of Lead (II) ion by GO-M and SMA-M.

Samples were collected from the supernatant and filtered. The equilibrium concentrations of Lead ions that remained in solution were measured using AAS. The effect of contact time on the Lead ion uptake was studied at constant pH (pH=6 for SMA-M and pH=8 for GO-M). The outcomes showed that adsorption increased rapidly during the first 20 min. After 20 min, the adsorption reached to the equilibrium state. As evident from Fig. 5, 20 min contact time was required to attain adsorption equilibrium for the Lead solution. During this time, more than 98% and 97% of the Pb (II) ions were adsorbed by GO-M and SMA-M respectively. At equilibrium, coordinate interaction between GO-M, SMA-M and Lead (II) ion occurred, and adsorption became almost constant up to the end of the test [16-19].

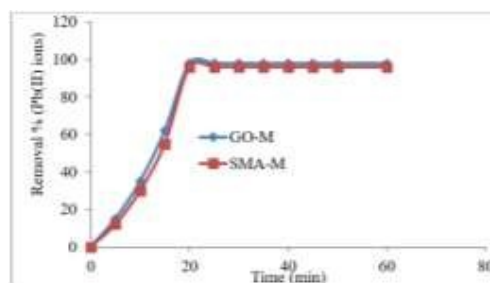


Fig. 5. Removal percentage of Lead (II) ions by 0.05 g of GO-M composite and 0.05g of SMA-M in 50 ml of 50 ppm Lead (II) ions aqueous solution at pH= 8 for GO-M and pH=6 for SMA-M in the period of 60 min.

It can be resulted that the binding of Lead (II) ion with GO-M and SMA-M are high during the initial stages and that, the reactive functional groups of GO-M and SMA-M were integrated after a certain period of time (optimized conditions shown in Table 1)[[20].

3. RESULTS AND DISCUSSION

3. 1. Desorption and repeated use

Desorption of Lead (II) ion was administered by 0.5M HNO_3 solution. The SMA-M– Lead ion, complexes were immersed in 0.5 M HNO_3 solutions and the mixture was shaken until equilibrium was reached (30 min). Then the mixture was filtrated and the final concentrations of Lead (II) ion in the aqueous solution was measured by AAs. The desorption ratio (D %) of toxic Lead (II) ion from the (CSMA-M and GO-M) adsorbents were computed by following equation:

$$D\% = \frac{V_d C_d}{V(C_i - C_e)} \times 100 \quad (2)$$

Where V is the volume of the solution (L); C_d is the concentration of the toxic Lead (II) ion in the desorption solutions (mg/L); and V_d is the volume of the desorption solution (L).The assembled (SMA-M and GO-M from the desorption process was washed thoroughly with deionized water and dried by vacuum pump at 65°C for repeated use [21-22].

3. 2. Preparation of Sample solutions

Various water samples (Ground water and Tap water) were compiled from different places in around Urmia and Salmas (Iran). The samples (100 mL) were saved at 4°C in metal free polyethylene bottles. Water samples were filtered through what man filter paper and collected into 200 mL beakers. All the filtered environmental water samples were evaporated nearly to dryness with a mixture of 5 mL con HNO_3 and 2 mL of con H_2SO_4 in a fume cupboard and then cooled to room temperature. The sample was digested in the presence of an excess potassium permanganate

solution according the method recommended. The residues were then heated with 6 mL of deionized water in order to dissolve the salts. The solutions were cooled and neutralized with dilute NH_4OH . The digest was transferred into a 20 mL calibrated

flask and diluted up to the mark with deionized water, then measured value of Lead (II) ions in solutions by AAS (Table 2).

Table 1. Optimized characters for removal of Lead (II) ion with SMA-M and GO-M.

Adsorbents	pH	Contact time(min)	Concentration of Lead(II)ion (ppm)	Amount of Sorbents(g L^{-1})	Temperature ($^{\circ}\text{C}$)
GO-M	8	20	20	1	25
CSMA-M	6	20	20	1	25

Table 2. Determination of Lead (II) ion in real samples.

Sample Name	Pb(II) found $\mu\text{g/mL}$								
	AAS	S.D	R.S.D(%)	Desorbed from GO-M	S.D	R.S.D(%)	Desorbed from SMA-M	S.D	R.S.D(%)
Ground water ^A	0.920	0.0002	0.01752	0.914	0.0003	0.01954	0.903	0.0003	0.018
Ground water ^B	0.835	0.00008	0.01182	0.829	0.0001	0.0029	0.832	0.008	0.020
Industrial waste water ^A	1.428	0.0014	0.0057	1.402	0.0015	0.0018	1.387	0.0013	0.021
Industrial waste water ^B	1.254	0.0011	0.023	1.255	0.0014	0.00169	1.265	0.0018	0.0231

A: Urmia B: Salmas

3. 3. Equilibrium Isotherm

The interrogated of equilibrium isotherms was administered by agitating of 50mg SMA-M resin and GO-M adsorbent in a solution containing different concentrations of Lead(II) ion for 60 min at pH =6 for SMA-M and pH=8 for GO-M. After filtration, the AAS was applied to measure the Lead (II) ion concentration in the filtrate. The amount of Lead (II) ion adsorbed q_e (mg/g) was measured using the following mass balance relationship:

$$q_e = \frac{V(C_i - C_e)}{S} \quad (3)$$

Where V is the volume of the solution (L); and S is the mass (g) of the adsorbent.

3. 4. Langmuir Isotherm.

Langmuir model has been widely used to some process of metal ions adsorption. The fundamental supposition of the Langmuir theory is that uptake of metal ions happens on a monotonous surface by monolayer adsorption without any interplay between adsorbed metal ions that is all the adsorption sites have equal adsorbate continuity and that the adsorption at one site does not affect the adsorption at a neighbor site. So, the Langmuir isotherm is exactly for monolayer adsorption onto a surface containing a determinate number of identical sites[23]. The Langmuir adsorption isotherm model is given by:

$$q_e = \frac{q_m C_e K_L}{1 + C_e K_L} \quad (4)$$

Where C_e is the equilibrium concentration (mg/L), q_e is the amount adsorbed at equilibrium

(mg/g), q_m is the maximum adsorption capacity corresponding to the complete monolayer coverage (mg/g), and K_L is the Langmuir constant affiliated to the adsorption energy[24]. The model of Langmuir adsorption isotherm can be rearranged as follows:

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m} \quad (5)$$

The linear plot of Langmuir adsorption isotherm (C_e/q_e vs. C_e) demonstrated the applicability of Langmuir adsorption isotherm for this work that was shown in Fig. 6.

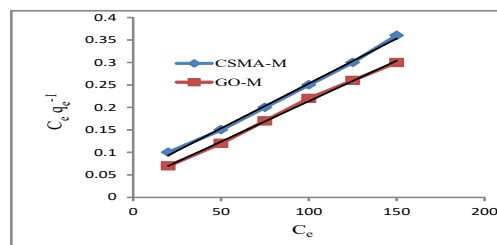


Fig. 6. Langmuir isotherms for Pb (II) adsorption onto CSMA-M and GO-M.

The values of q_m and K_L for Pb (II) ions were calculated from the slope and the intercept of the linear plots C_e/q_e versus C_e . The results are listed in Table 2. The results showed that good fit was achieved with the Langmuir isotherm equation. The Langmuir parameters can also be used to predict affinity between the adsorbate and the adsorbent using the without dimension separation factor (R_L), which has been defined as below:

$$R_L = \frac{1}{1 + C_0 K_L} \quad (6)$$

$$\Delta G = -RT \ln K_L \quad (7)$$

Where K_L is the Langmuir constant (L/mg) and C_o is the concentration of toxic heavy metal ions, in mg/L. The amount of R_L illustrated the type of the isotherm to be either undesirable ($R_L > 1$), linear ($R_L = 1$), desirable ($0 < R_L < 1$), or irreversible ($R_L = 0$). Fig. 7 illustrated that adsorption of Lead (II) ion increased continually with a concentration of Lead (II) ion. The computed R_L values for Lead (II) ion adsorption illustrate that adsorption is desirable even for higher concentrations of Lead ion (Langmuir constants exhibited in Table 3) [25-26].

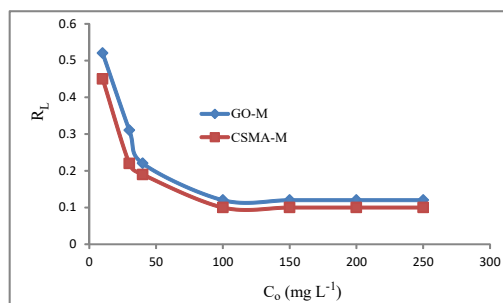


Fig. 7. Separation factor (R_L) for adsorption of lead (II) ion as function of concentration of lead ion by CSMA-M and GO-M.

Table 3. Langmuir isotherm parameters and regression data of Lead (II) ion using GO-M and CSMA-M as adsorbents.

Langmuir isotherm	GO-M	CSMA-M
q_m (mg g ⁻¹)	555.56	526.32
K_L (L mg ⁻¹)	0.053	0.035
R_L	0.386	0.489
R^2	0.9981	0.9968
ΔG (Kj mol ⁻¹)	-81.2	-71.3

4. CONCLUSION

In this study, new composites were synthesized based on the melamine (M) stabilized with GO and poly (Styrene- alternative- Maleic Anhydride) (SMA) as Sorbents for the removal of amounts Lead (II) ions from aqueous solutions. Based on the results, it is clear that the Lead-removal values achieved with GO-Melamine composites were higher than GO and SMA-M were higher than SMA [27-28]. The main advantages of this elimination method include simplicity, cost effectiveness, rapidity, and higher elimination efficiency of toxic Lead (II) ions. The GO-M composites can be produced readily by a solution of EDTA and they have longer lifetime and also a desorption of Lead (II) ion was administered by 0.5M HNO_3 solution. The SMA-M- Lead ion and GO-M complexes were immersed in 0.5 M HNO_3 solutions and the mixture was shaken until equilibrium was reached (30 min). Then the

mixture was filtrated and the final concentrations of Lead (II) ions in the aqueous solution were measured by AAs. The adsorbents were resulted to be reproducible with a good Lead (II) ion selectivity over other potentially interfering ions. Data of these adsorbents very set with Langmuir isotherm. These adsorbents were tested on various water samples, which were shown high adsorption capacity for the elimination of amounts Lead (II) ions. These adsorbents are very proper for removing heavy metal ions specially Lead (II) ions from aqueous solutions[29].

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چکیده

در این مطالعه یک روش جدید بایکالگری نانو کامپوزیت های گرافن اکسید- ملامین و مشتقی از پلی استایرن - آلترناتیو- مالییک انیدرید- ملامین به عنوان جاذب برای حذف یونهای سرب از محلولهای آبی مورد استفاده قرار گرفته اند. جاذب ها برای حذف یونهای سرب قابلیت لازم را دارند. با نشان دادن و ثابت کردن ملامین روی نانورقه های گرافن اکسید ترکیب مورد نظر ساخته شده و بوسیله تکنیک های میکروسکوپ الکترونی روبشی نشری، اسپکتروسکوپی تبدیل فوریه فروسرخ و اسپکتروسکوپی جذب اتمی مورد ارزیابی و شناسایی قرار گرفت. پارامترهای مختلف نظیر غلظت محلولهای سرب، مقدار جاذبها، زمان تماس با جاذبها و pH بهینه شده اند. در این مطالعه نشان داده شده است که با نشان دادن ملامین روی جاذبها کارایی آنها افزایش پیدا کرده است.

واژه های کلیدی

گرافن اکسید؛ حذف؛ نانورقه ها؛ ملامین؛ نانو کامپوزیت.