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RESEARCH ARTICLE

Preparation of Graphene Oxide from Waste Pencils and Loading it on the Surface of Melamine-Formaldehyde/rGO Composite for Hydrocarbons Removal

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ABSTRACT

With the increasing technological development, the problem of hydrocarbon pollution has become one of the urgent problems of our current era. Many techniques have been proposed and applied to address this phenomenon, including the use of polymer-based nanocomposites. This study involved the preparation of a few wrinkled layers of graphene oxide from waste pencils (wp-GO) as proved by the High-resolution transmission electron microscopy HRTEM image. Besides, magnetic iron oxide nanoparticles were synthesized and showed superparamagnetic properties due to the magnetic properties of 40 emu/g saturation magnetization and close to zero coercivity, the structure is mostly spherical particles with a diameter of 8–10 nm. These nanomaterials were exploited as a composite along with melamine-formaldehyde polymer MF. The HRTEM results showed that the prepared wp-GO-MF composite has a stable structure with many porous. The wp-GO-MF composite was applied for several hydrocarbons adsorption. Hydrocarbons adsorption study was accomplished using benzene, toluene, m-xylene, hexane, heptane, kerosene, and diesel oil. The findings revealed that the wp-GO-MF composite performs the highest sorption capacity (Qe) for kerosene at 71 g g^{-1}. Adsorption capacity was maintained at around 90% for the hydrocarbons up to 10 absorption cycles. Therefore, it can be concluded that the wp-GO-MF composite successfully showed an excellent sorption capacity for all tested hydrocarbons with superb recycle performance.

Keywords: Composite, Hydrocarbons adsorption, Magnetic iron oxide nanoparticles, Melamine formaldehyde polymer, Waste pencils

Introduction

Petrol disasters involving spills of oil on oceans and seawater happen overwhelmingly in the present time and require a rapid and economically suitable solution.[1] Consequently, a critical issue herein is the removal of traces of harmful hydrocarbons organic from aqueous solutions. Recently, the pollution of water has had a substantial impact on humans and the environment, which requires critical and effective processes to deal with this issue.[2] Aromatic hydrocarbons are considered one of the usual pollutants among all dangerous environmental contaminants, which are created due to the burring of organic compounds such as coal, oil, gas, wood tobacco, etc. Organic hydrocarbons can be considered as extremely serious compounds that can cause chronic and persistent health effects.[2] Many methods were used to handle pollution problems. Diverse treatment methods such as chemical, thermal, biological and UV-vis techniques have been applied to eliminate these environmental pollutants.[3] However, these methods have

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some drawbacks such as high cost and complex post-processing. Therefore, the core point of pollution treatment is the selection of eco-friendly and effective treatment processes. Purification processes based on the adsorption method could be used effectively to remove the chemical substances in the water which are not simply treated by using chemical and biological ways.[4]

Polymer sorbents such as polystyrene,[5] polyurethane foam,[6] and melamine resins,[7] have usually been employed to reduce the impacts of these disasters. These sorbent materials are effective because they are easy to use, low density, available, and inexpensive. It can introduce some modifications to overcome some drawbacks of sorption in these sorbents. One of these modification processes is using nanomaterials, which can be employed separately as chemicals (sorbents) for the elimination of hydrocarbon products from the surface of the water as mentioned by Munyengabe et al.[8] One of the polymeric materials that can be used for removing hydrocarbons from aqueous solutions is melamine-formaldehyde polymer. It is considered a 3D network thermosetting material that has high hardness and rigidity. It has been vastly applied in foams, adhesives, fiber, photo-electric, repellents of water, and coating agents. In addition, Tang et al.,[9] pointed out that Melamine is widely used in flame-resistant substances because it helps to inhibit fire in many fields and applications by acting as a fire retardant. Melamine-formaldehyde composites have been applied in several areas such as industrial, medical, and water treatment. Reinforced melamine-formaldehyde polymer was used to enhance adsorption and mechanical and physical properties. It can use cellulosic reinforcements, novalac, amino compounds, alginate, nylon fibers, and nanoparticles to improve its properties. Moreover, some papers discussed the impact of melamine resins on the thermosets of polyisocyanate emulsifiers. Hassan et al.,[10] used prepared melamine formaldehyde/nanozeolite/nano-silica composites to determine adsorption capacity. Another study mentioned that the combination of porous nanocarbon with melamine-formaldehyde polymer led to the production of high-activity surfactants. In addition, polyvinyl chloride/melamine-formaldehyde polymer was applied to the production of composites in the adsorption of the drug. Therefore, the mechanical and physico-chemical features of melamine-formaldehyde polymer have been improved by using several methods especially based on nanomaterials.[10]

Polymer nanocomposites were employed to provide further value features to the homopolymer, without losing its process ability. Various kinds of sorbent materials have already been used for hydrocarbons pollutant elimination from solutions, such as activated carbon,[11] clay,[12] and biosorbents.[13] In this context, carbonic nanoparticles have become one of the most promising substances in nanoscience attributed to their distinctive thermal, electrical, and mechanical features. Fan et al.,[14] found that modified melamine-formaldehyde polymer was applied to remove several heavy metal ions by using adsorption. Other studies show that the modification of the surface of melamine-formaldehyde polymer using nanoparticles could facilitate separation and recycling properties. Graphene exhibited promising findings for petroleum pollutant removal from solutions. Currently, nanoparticles can be applied as proper and effective chemicals to remove contaminants from the surrounding environment. Among all these materials, graphene derivatives have shown promising findings as an influential alternative for the elimination of organic pollutants from the environment as a result of their unique chemical and physical features.[14] Among all techniques that have been suggested to eliminate contaminants, adsorption was considered worldwide as an attractive choice due to its adaptability, broad applications, and economic viability. Recent studies pointed out that GO can remove aromatic organic compounds and can be used promising alternative for conventional adsorbent compounds with excellent findings.[15]

Due to great research, many important enhancements have been achieved in the synthesis and adaptation of these compounds that have been mostly introduced as successful adsorbents of toxic chemical contaminants. The usage of novel effective adsorbents based on polymer/graphene has opened the door to sustainable eco-friendly remediation. Polymer composites with nano-materials like graphite, carbon black, graphene, or metals have recently substantial attention due to their effective applications. Homogenous filler that is appropriately dispersed in the polymer matrix can be created with a tidy filler content.[16] In this study, we used GO with melamine-formaldehyde polymer for improving mechanical, and thermal properties and surface modification. Graphene oxide and reduced graphene oxide were used as insertion compounds. Graphene oxide has a similar layered structure like natural graphene associated with functional groups containing oxygen atoms. Given this context, our study was focused on the application of a novel, successful way of producing operative sorbents depending on highly porous polymer matrices by melamine resin foam with modification of the surface by nanomaterials. It can be

possible to insert rGO sheets within the surface of the polymer to obtain hydrophobic material and sorption of hydrocarbon pollutants. Moreover, groups such as hydroxyl, carboxyl, carbonyl, and epoxy groups can provide a massive number of reactive sites for the interaction of GO.

Materials and methods

Preparing wp-GO

The waste pencils were collected from students at the University of Basrah, Firstly, they were immersed in water for 24 hours, the wood was inflated and the carbonaceous part was easily removed from the wood. After that, the carbonaceous part was heated in a furnace to 150 °C for two hours to remove the stacking wood and stains. Then, the obtained carbonaceous was ground by Frish Germany – grinder. Finally, the results were sieved by Test Elegi micro sieves 25 microns and termed (wp).

An amount of 2 g of (wp) was added to 92 mL concentrated sulfuric acid (98%, Merck) and 2 g, 0.024 M of Sodium nitrate (98%, Merck) while stirring in an ice-water bath for one hour the color turned to black. 3 g, 0.019 M potassium permanganate (98%, Merck) was progressively added while maintaining the temperature under 10 °C for 15 min and the color turned to dark green. Then, the suspension was stirred overnight at room temperature. After that, the reaction was quenched by the adding 200 mL distilled water, and then the temperature was raised to 90 °C for 30 min. An extra 400 mL of purified water was added with stirred for five minutes. Then, 200 mL of water and 18 mL of hydrogen peroxide (30%, Scharlau) were added to the mixture solution and stirred for ten minutes.[17] The obtained solution was centrifuged during washing with distilled water 10 times, followed by 6 hours of sonication in an ultrasonic bath (Guagzhou Henwei Electronics) for exfoliation.

Preparation of deep eutectic solvents

27.5 g, 0.197 M Choline chloride (Himedia laboratories private limited, Mumbai, India) was added to 24 g, 0.31 M urea (99%, Merck) in a beaker, then heated to 45 °C for 2 hours to obtain a clear solution of deep eutectic solvents.[18] This solvent was added to the exfoliated GO and sonicated for 5 hours, centrifuged during washing with distilled water 10 times then dried by a vacuum oven at 60 °C for 24 hours to acquire wp-GO powder.

Preparation of polymer melamine-formaldehyde (MF) tablets

2.27 g, 0.018 M of melamine (99%, HiMedia), and 0.0021 g of wp-GO were put in two necks round bottom flask equipped with a condenser and thermometer located on a magnetic stirrer. 6 mL formaldehyde (30%, Aqua Medikal) was added to the melamine mixture at 50 °C for 30 min. Then, after adjusting the pH at ~8.5 by the addition of 9.5 mL, 1 M sodium hydroxide (98%, Merck) 0.2 g, 0.57 mM of sodium dodecyl benzene sulfonate was added at 60 °C for 1 hour. The sticky solution was discharged in a beaker and added 0.15 mL acetic acid and 0.1 mL petroleum ether with hardy stirring. The product was placed in a silicon mold and left for 3 hours for complete polymerization.

Reduction of polymer melamine–formaldehyde tablets

0.3 g, 1.7 mM ascorbic acid was dissolved in 50 ml ethanol in a beaker, after that the prepared tablets were placed in this solution and the beaker was positioned in the sonicator at 60 °C for 1 hour while the pH was adjusted at ~ 10 by using 12 mL, 1 M ammonium hydroxide. Then, the tablets were dried at 40 °C for 24 hours.

Preparation of magnetic iron oxide nanoparticles (m-Iron oxide NPs)

In a round bottom flask equipped with a mechanical stirrer, 4.0 g, 0.0148 M $FeCl_3.6H_2O$, and 2.5 g, 0.0126 M $FeCl_2.4H_2O$ were dissolved in 100 mL double distilled water under argon atmosphere and the temperature was slowly increased to 35 °C for 10 min. Next, 50 mL NaOH (25%) was added progressively (pH~10), and the temperature was raised to 56 °C for 10 min while controlling the homogenization by overhead stirrer at a rate (700 rpm) to prevent the agglomeration of large poly-crystalline particles.[19] Then, 150 mL, 0.5 M citric acid was added to the mixture for 10 min while holding the temperature at 56 °C. Magnetic separation utilizing a neodymium magnet and washing several times with deionized water was used for the black precipitates.[20] Finally, 100 mL of double distilled water was added to the black precipitates and ultrasonicated for 30 min.

The different ratios of wp-GO powder (0.001, 0.003, 0.005, and 0.01 g) were added to the reduced melamine polymer–formaldehyde tablets in 50 mL ethanol and ultrasonicated for 1 hour after the tablets

were dried by air for 24 hours. Then, the resulted tablets were immersed in the magnetic iron oxide nanoparticles and ultrasonicated for 60 min then dried at 40 ˚C for 2 hours.

Sorption study

To reveal details on wp-GO-MF adsorption performance, several studies were utilized such as sorption capacity, sorption kinetics, and recyclability. The sorption capacities of the wp-GO-MF were conducted by following the weight of the wp-GO-MF tablet before and after adsorption. The wp-GO-MF tablet was tested for its recyclability by obtaining the weight of recovered hydrocarbons that would be divided by the weight of the wp-GO-MF tablet before adsorption for each cycle, while the sorption kinetic of solute absorbed per gram of sorbent was determined by applying Pseudo-second-order reaction.

Liquid sorbate

The adsorption performance of the wp-GO-MF tablet was investigated, and various chemicals were tested such as benzene (Sigma), toluene (Sigma) and m-xylene (R & M Chemical), hexane (Sigma), pentane (Sigma), and the study including various oils such as kerosene (Winstar enterprise SDN. BHD), diesel fuel (Basrah station).

Sorption capacity

The sorption capacity measurement was examined depended the ASTMF726-99:[21] Sorbent Performance Standard Test Method of Adsorbents. For oil adsorption tests, hydrocarbon (50 mL) was put in 100 mL beakers. The weight of the wp-GO-MF tablet absorbents was listed before dipping in the hydrocarbons (batch experiments). Pre-experiments were organized to decide adsorption time that is 10, 20, 30, and 40 min ± 5 s using wp-GO-MF tablets utilizing 0.005 g wp-GO, and the toluene as a hydrocarbon source. 30 min ± 5 s was the highest sorption capacity, which is equal to 40 min ± 5 s meaning the tablet reached saturation. Thus, in specific, the adsorbent was separated after 30 min ± 5 s of immersing and drained for a few minutes. The soaked adsorbent was weighed immediately by a weighing glass Petri dish. Sorption capacity (g g^{-1}) was stated in grams of contaminant adsorbed per gram of wp-GO-MF was determined using Eq. (1):

$$Q_t = \frac{m_t - m_0}{m_0} \quad (1)$$

Where Q_t (g g^{-1}) is the sorption capacity of the wp-GO-MF tablet at an evident time t (s), m_t (g) is the weight of wp-GO-MF tablet after adsorption, and m_0 (g) is the initial weight of the wp-GO-MF tablet. The sorption capacity reached a saturated value Q_e (g g^{-1}) when Q_t was unaffected with (t).

Sorption kinetic study

Sorption kinetics was accomplished to study the sorption mechanism of oil onto the wp-GO-MF tablet and to elucidate the investigational data obtained by utilizing the pseudo-second-order model which has proven as an appropriate model by many studies according to the correlated data obtained from the experiments.[22]

The Eq. (2) describes the linearized form of the pseudo-second-order kinetic model:

$$\frac{t}{Q_t} = \frac{1}{K_2\, Q_e^{\,2}} + \frac{1}{Q_e}\, t \quad (2)$$

Where Q_e (g g^{-1}) and Q_t (g g^{-1}) are the sorption capacities at equilibrium and at various times (t), respectively; k_2 is the second-order rate constant (g $g^{-1}$$min^{-1}$) for adsorption. Besides, the slope and intercept of the linear plot of $\frac{t}{Q_t}$ against t resulted from the values of slope $\frac{1}{Q_e}$ and intercept $\frac{1}{K_2\, Q_e^2}$ respectively.

Recyclability

To evaluate the recyclability and reproduction of the adsorbent, vacuum filtration was utilized to eliminate the hydrocarbons from the applied wp-GO-MF tablet. Then, the wp-GO-MF tablet was heated around the boiling point of adsorbate to exploit in the next hydrocarbon adsorption assessment to characterize the recycling performance.[23]

The wp-GO-MF tablet was tested for its recyclability by obtaining the weight of recovered hydrocarbon that would be divided by the weight of the wp-GO-MF tablet before adsorption Eq. (1) for each cycle.

Characterization

Field emission scanning electron microscopy (FESEM) observation realized with Energy Dispersive X-ray spectroscopy using EDX was carried out by using FESEM Instrument: TESCAN MIRAIII CZECH. High-resolution transmission electron microscopy (HRTEM) was carried out on a TECNAI G2 F20, X-TWIN model FEI. Value stream mapping (VSM) for measuring the magnetization properties using WEISTRON VSM1100 TIWAN. X-ray diffraction

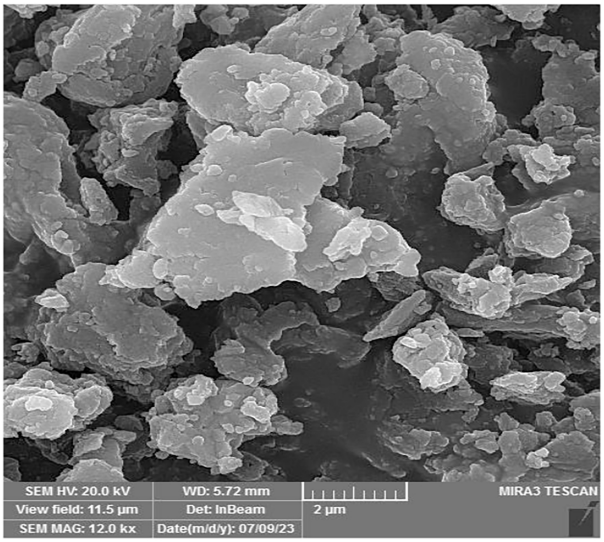

Fig. 1. FESEM image of wp-GO showing the flake-like structure with the aggregation.

analysis (XRD) for crystallographic structure details utilized PILIPS PW1730 HOLANDA.

Results and discussion

Fig. 1 shows the FESEM image of the wp-GO morphology that has small sizes and a flake-like structure with high aggregation. The aggregation of nanoparticles is a familiar phenomenon in graphene oxide, mostly driven by the high surface energy of the nanoparticles and soft exfoliation. Fig. 2 shows the HRTEM image of the wp-GO, which appeared as highly electron transparent, besides it illustrates the wrinkled structure of a few layers of graphene oxide.

Fig. 3 shows that the EDX analysis of wp-GO exhibits two main peaks of carbon 49.66% and oxygen 39.66% indicating a high value of oxygenated groups in the wp-GO.

Fig. 4 displays the FESEM image of m-Iron oxide NPs. The image exhibits a homogeneous pattern revealing a uniform organization of aggregation nanoparticles structure, the average diameter of the agglomerated particles was 25 ± 5 nm. HRTEM in Fig. 5 indicates that particles have mostly spherical shape, with a diameter of 8–10 nm. This finding is expected and it can be illustrated due to the citric acid behavior as a reducing and capping agent of the m-Iron oxide NPs. However, it can be grasped that the size of m-Iron oxide NPs expresses uniform growth over the clusters.

Fig. 6 illustrates the powdered X-ray diffraction patterns of citric acid capped m-Iron oxide NPs. The main characteristic peaks are (220) at 30.40 $2\theta^{\circ}$, (311) at 35.70 $2\theta^{\circ}$, (400) at 43.33 $2\theta^{\circ}$, (422) at 53.89 $2\theta^{\circ}$, (511) at 57.38 $2\theta^{\circ}$, and (440) at 63.01 $2\theta^{\circ}$, This finding pointed out that the γ-Fe_2O_3 was prepared and the feature of iron oxide is a cubic structure according to the (ICDD No. 00-039-1346).[19] The patterns of XRD exposed highly crystalline single phase illustrated in the diffraction peaks of γ-Fe_2O_3 nanoparticles. The average size of crystallite for prepared samples was equal to 9.43 nm approximately which was calculated from the Scherrer equation.

Fig. 7 shows the value stream mapping (VSM) analysis of citric acid capped m-Iron oxide NPs, according to the plot the nanoparticles are considered to be super-paramagnetic because the hysteresis loop does not display magnetic remanence. The magnetization of saturation (M_s) of the Iron oxide magnetic nanoparticles was 40 emu/g and the coercivity was close to zero emu/g, this indicated that the citric acid molecules are covered by the iron oxide nanoparticles leading to the increased distance between the

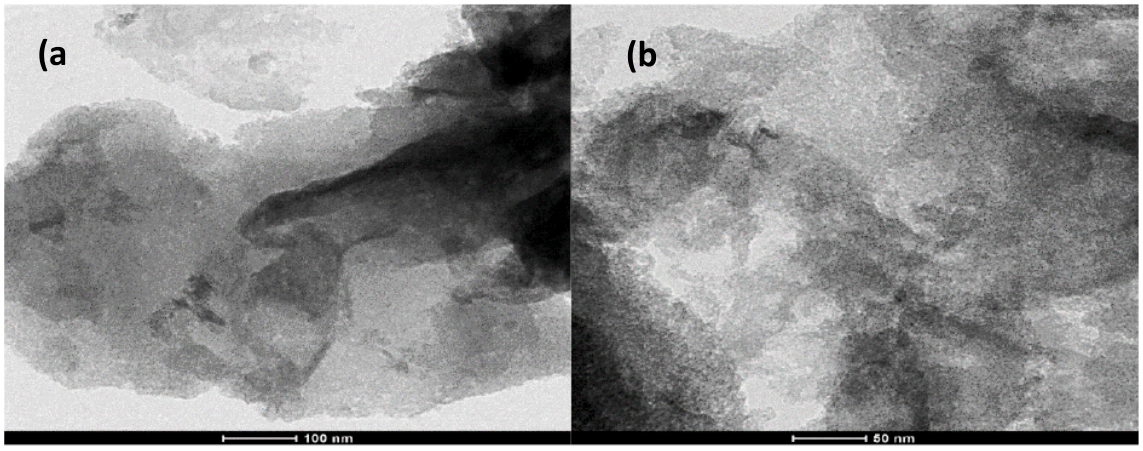

Fig. 2. HRTEM images of wp-GO showing highly electron transparent flake-like wrinkled structure.

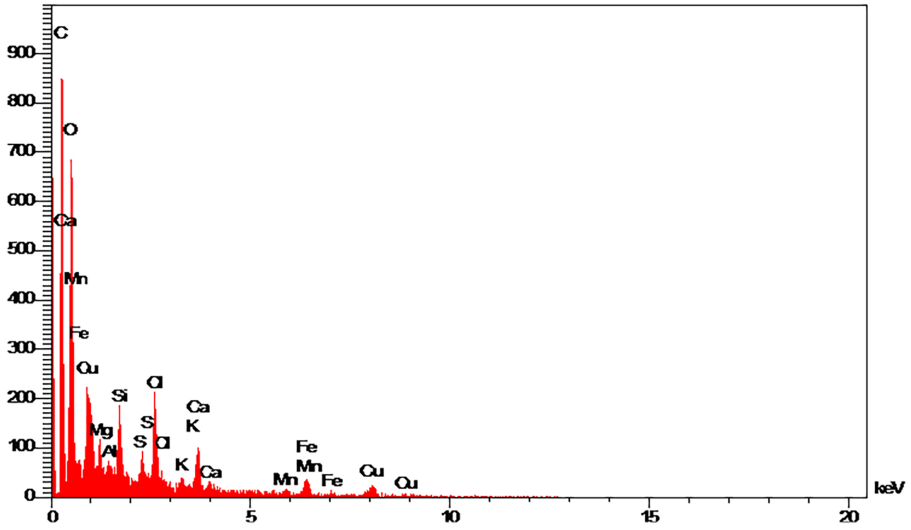

Fig. 3. EDX profile of wp-GO, showing a majority of the carbon and oxygen.

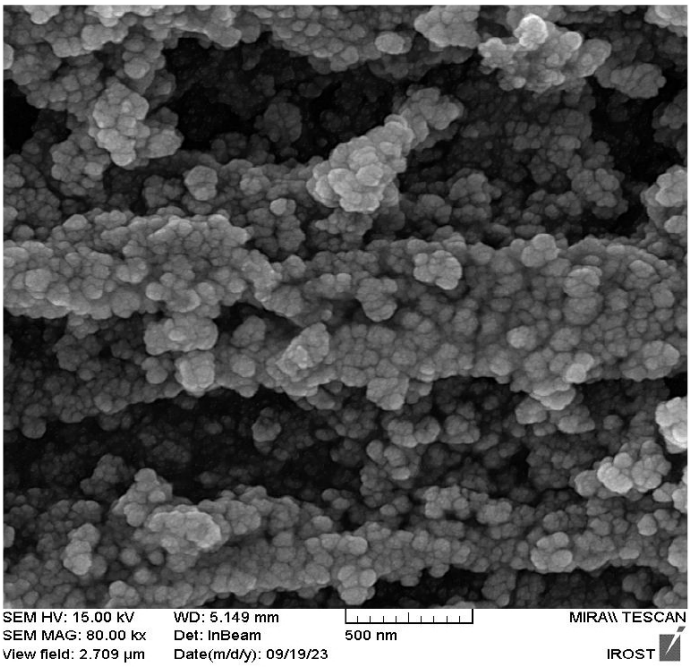

Fig. 4. FESEM image of m-Iron oxide NPs showing a homogeneous pattern of aggregate nanoparticles.

nanoparticles that cause the decrease of the dipole-dipole interaction between the inter-particles. This finding was proved by HRTEM analysis.

In this current work, several tablets of wp-GO-MF polymer are prepared by using reduced GO, vitamin C is exploited as a reducing agent to prepare rGO that could adhere to the skeleton of the polymer. Then, various ratios of wp-GO powder (0.001, 0.003, 0.005, and 0.01 g) were added to the polymer. Finally, the polymer was loaded with m-Iron oxide NPs, Fig. 8. The magnetic iron oxide nanoparticles are used to enhance the adsorption ability as well as improve the ability to withdraw the tablets after the adsorption of the hydrocarbons.

According to the morphology of the wp-GO-MF tablets characterized using FESEM, the tablet loaded with 0.005 g explicit the best homogeneously and more porosity than the other pellets, Fig. 9 illustrates the FESEM image of 0.005 g of wp-GO powder. This tablet was chosen for the adsorption study.

Fig. 10 shows the XRD of the wp-GO-MF polymer composite. The XRD pattern shows a broad peak at

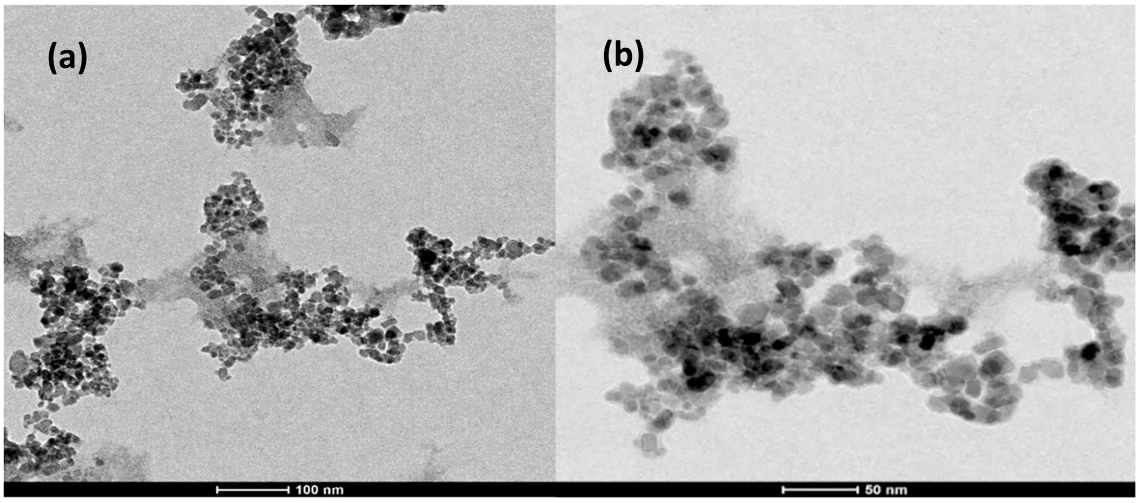

Fig. 5. HRTEM images of m-Iron oxide NPs showing spherical particles with a diameter of 8–10 nm.

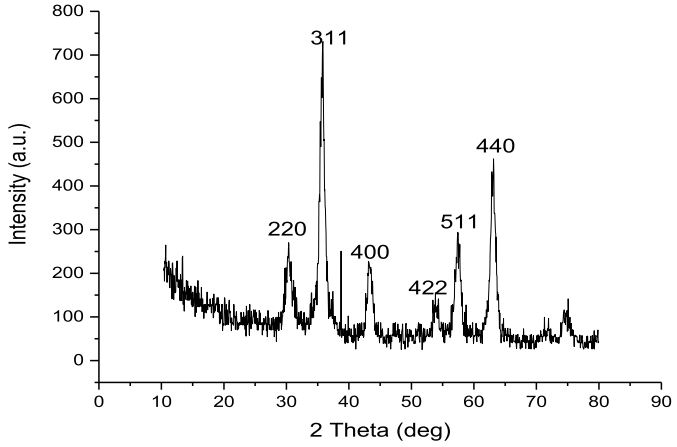

Fig. 6. XRD patterns of the citric acid capped Iron oxide magnetic nanoparticles.

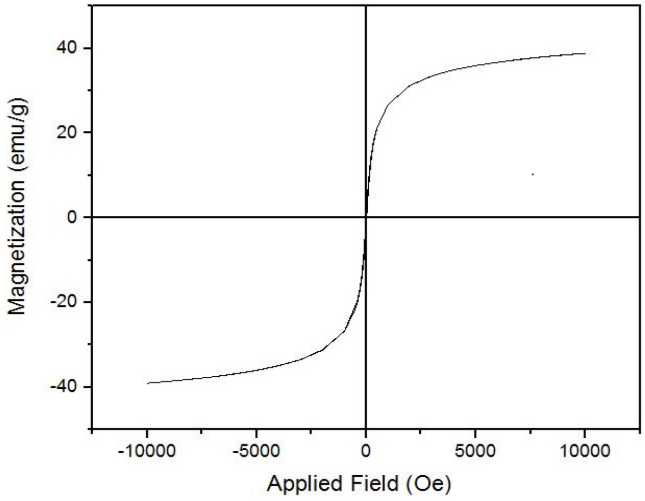

Fig. 7. Magnetometric curves for the citric acid capped iron oxide nanoparticles.

Fig. 8. Photograph of blank melamine formaldehyde tablets loaded by (0.001, 0.003, 0.005, and 0.01 g) of wp-GO powder.

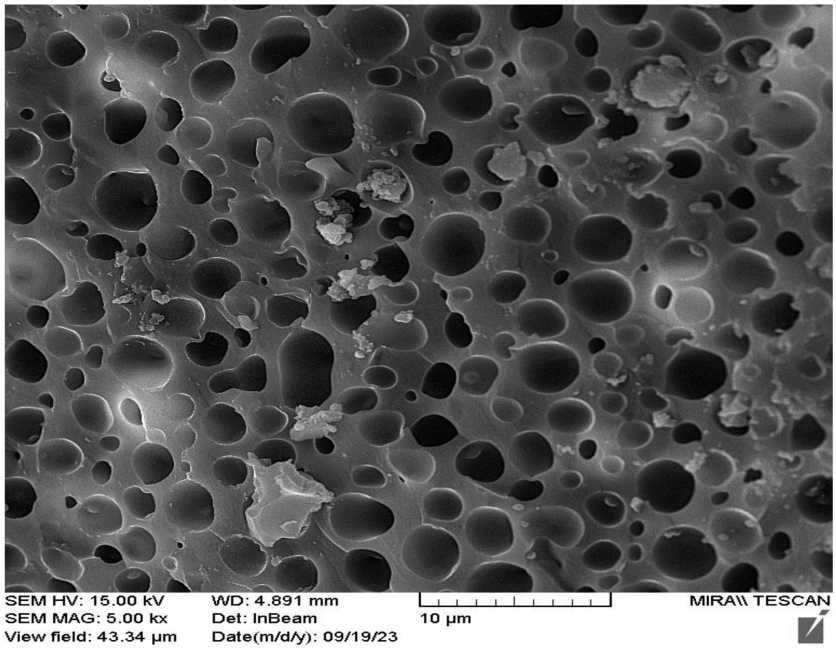

Fig. 9. FESEM image of melamine–formaldehyde polymer loaded with 0.005 g of wp-GO powder.

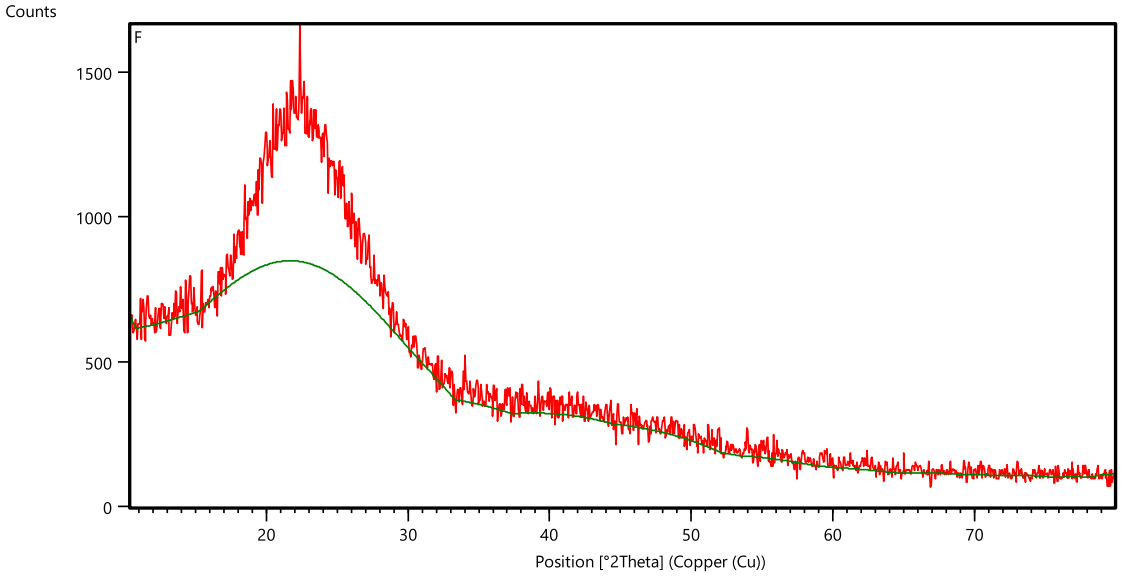

Fig. 10. The XRD of the melamine-formaldehyde polymer composite.

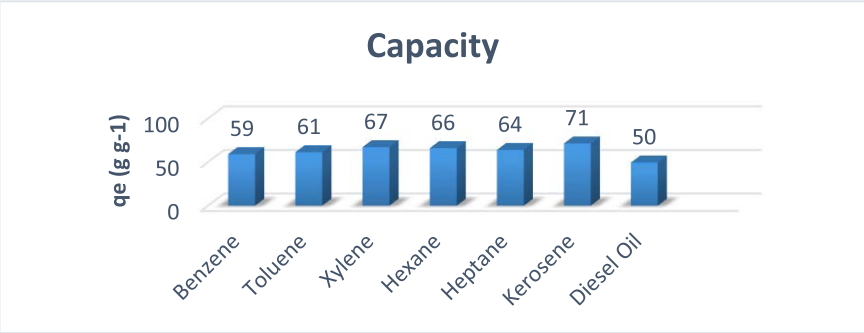

Fig. 11. The equilibrium sorption capacity of wp-GO-MF tablets for different organic solvents and oils.

24 2θ° indicating the amorphous nature of the polymer composite.

The conventional methods applied to clean oil spills include biological, chemical and physical methods, most of these techniques generally are slow, expensive, require high energies, or cause secondary pollution. Recently, most of the physical methods are focused on the use of porous materials. One of these materials is melamine/graphene sponges. All studies have shown that the most effective strategies to obtain super-hydrophobic properties in melamine foam are by transforming the hydrophilic surface of MF into a hydrophobic surface, and endowed MF with higher oil absorption capacity and excellent recyclability.[10]

To investigate the sorption performance of the wp-GO-MF polymer, a sorption study which includes sorption capacity, kinetic study, and recyclability was achieved for various hydrocarbons. The hydrocarbons used in the study are benzene, toluene, *m*-xylene, hexane, heptane, kerosene, and diesel fuel. The study was accomplished by selecting melamine formaldehyde loaded with 0.005 g of wp-GO powder for sorption study due best morphology according to the FESEM image.

A typical adsorption test was carried out to ensure the wp-GO-MF tablets able to absorb organics and oils, the wp-GO-MF tablets were immersed into the hydrocarbons. It was observed that these hydrocarbons were immediately absorbed in 2 to 15 min. The equilibrium sorption capacity (Q_e), which is described by the weight ratio of the absorbate to the wp-GO-MF polymer at saturation, was used to evaluate the adsorption performance toward various organic solvents and oils in Fig. 11 and Table 1.

The wp-GO-MF tablets had quite larger adsorption in benzene, toluene, xylene, hexane, heptane, and kerosene, though they displayed lower adsorption regarding diesel oil. The lower adsorption of the diesel oil might be clarified because the diesel oil comprises approximately 25% aromatic hydrocarbons such as benzene and styrene, and about 75% aliphatic hydrocarbons ($C_{10}H_{20}$–$C_{15}H_{28}$).[24] Therefore, the diesel oil included some relatively longer hydrocarbon chains and molecular weight than mostly adsorbed hydrocarbons on the surface of the wp-GO-MF and therefore impeded the absorbency further compared to small molecules. According to the ability of wp-GO-MF composite to adsorb low and high molecular weight hydrocarbons, it could be determined that the wp-GO-MF composite can adsorb other hydrocarbons enclosed to the tasted hydrocarbons molecular weight.

Sorption kinetic is one of the most significant characteristics that manage the solute adsorption rate and characterize the adsorption efficiency and thus, establish its potential applications. The pseudo-second-order kinetic of different hydrocarbons absorbed by the wp-GO-MF is shown in Fig. 12, where the sorption capacity Qt of each contaminant is plotted as a

Table 1. Sorption capacity and pseudo-second-order kinetic parameters for the hydrocarbons.

		Pseudo-second order parameters		
Hydrocarbons	Qe, exp (g g^{-1})	K_2 (g g^{-1} min^{-1})	Qe, cal (g g^{-1})	R^2
Benzene	59	0.0178	63.171	0.9953
Toluene	61	0.0172	65.963	0.9959
Xylene	67	0.0185	71.024	0.9996
Hexane	66	0.0175	63.331	0.9994
Heptane	64	0.0169	66.007	0.9995
Kerosene	71	0.0678	71.685	0.9974
Diesel oil	50	0.0147	54.675	0.9981

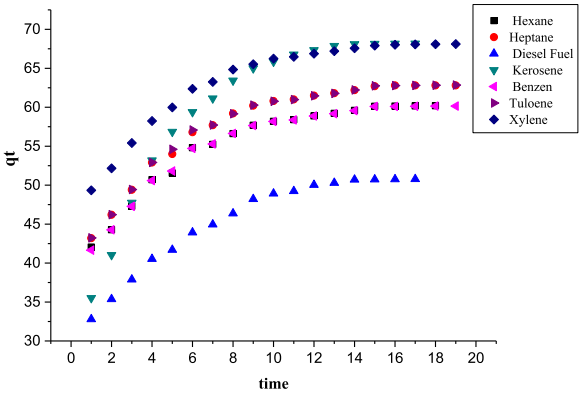

Fig. 12. Sorption kinetics of the wp-GO-MF composite for hydrocarbons adsorption.

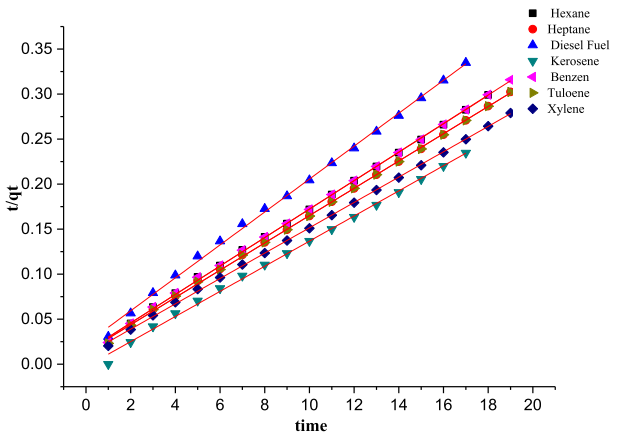

Fig. 13. Linear regression lines of kinetics plots using the pseudo-second-order model of the hydrocarbons adsorbed by wp-GO-MF composite.

function of the sorption time. From the experimental results, rapid initial adsorption from hydrocarbons to the wp-GO-MF is observed within 5–10 min, which implies a prompt initial movement of solute into the wp-GO-MF surface layers. At this step, the adsorption is larger than the equilibrium absorbed amount. Large differences in solute concentration between phases cause quick movement of the solute when the solution starts to interact with the solid. Large numbers of pores in the precise size range of wp-GO-MF contribute very well to filling the pores through capillary condensation.[25] The kinetic curve displays that the solute quantity increases slowly with time until the adsorbent has a steady value which means the solute reaches the equilibrium.

The plot of t/Qt versus time Eq. (2) gives a straight line. From the slope and intercept Qe and K_2 values are obtained, and the linear regression of hydrocarbons is shown in Fig. 13, and the results as illustrated in Table 2. The values of Qe, k_2 obtained from this rating model are given in Table 1. The high correlation coefficient (R^2) value between 0.995–0.999 implies that the collected data fit the pseudo-second-order model, furthermore, the chemical interactions of the solute and the surface of wp-GO-MF are probably involved in the adsorption processes and the adsorption capacity is relative to the number of active sites on wp-GO-MF polymer. In Table 1, the experimental sorption capacity Qe is the attained value in sorption experiments at equilibrium that is obtained at a certain time concentration, while the calculated Qe from the kinetic model is the predicted value obtained from the equation of kinetic study with increasing time. Thus, the data seem close although do not match well the precise kinetic plot.

Table 3. illustrates the comparison of the adsorption capacities of graphene-based composites reported in the literature. The adsorption capacities of the wp-GO-MF composite were close to or better than those sorbent materials previously reported.

The recyclability of the wp-GO-MF polymer was achieved by heating up at the boiling point of adsorbate to use in the next hydrocarbon adsorption test. Determining the recyclability of absorbent materials for the removal of organics or oils is an important factor in assessing the practical usage performance of the wp-GO-MF polymer to evaluate its potential application. As shown in Fig. 14, the wp-GO-MF polymer shows high performance toward absorption of the hydrocarbons. Wp-GO-MF polymer exhibited high recycle performance and sustained absorption capacity of 90% for the hydrocarbons after 10 cycles of test

Table 2. Intercept, slope, and R-Square based on linear regression lines of kinetics plots.

Hydrocarbons	value	intercept Error	slope value	Error	statistics R-Square
Benzen	0.01408	0.000921	0.01583	0.000080	0.9953
Tuloene	0.01339	0.000828	0.01516	0.0000726	0.9959
Xylene	0.01069	0.000695	0.01408	0.0000613	0.9996
Hexane	0.01426	0.00102	0.01579	0.000093	0.9994
Heptane	0.01358	0.00088	0.01515	0.000077	0.9995
Kerosene	0.00287	0.00181	0.01395	0.000175	0.9974
Diesel oil	0.02271	0.00202	0.01829	0.000197	0.9981

Table 3. Comparison of adsorption capacities of graphene-based composites.

Sorbents	Type of oil	Qe, cal (g g^{-1})	Ref.
Graphene/poly-pyrrole foam	Kerosene	101	26
Graphene sponge	Crude oil	85–95	27
Silane-functionalized graphene sponge	Diesel oil	86	28
	Hexane	25	
Graphene-coated melamine sponge	Diesel oil	105	29
graphene-based sponges	Hexane	60	30
rGO@ melamine formaldehyde sponge	Kerosene	93	31
	Toluene	104	
Graphene/Cellulose/Silica Aerogel	xylene	58	32
	Toluene	55	
	benzene	68	
melamine sponge/silver-reduced graphene oxide	Diesel oil	57	33
PVC/SiO_2 nanoparticles/melamine sponge	xylene	45	34
Isocyanate-Modified Melamine Sponges	Heptane	75	35

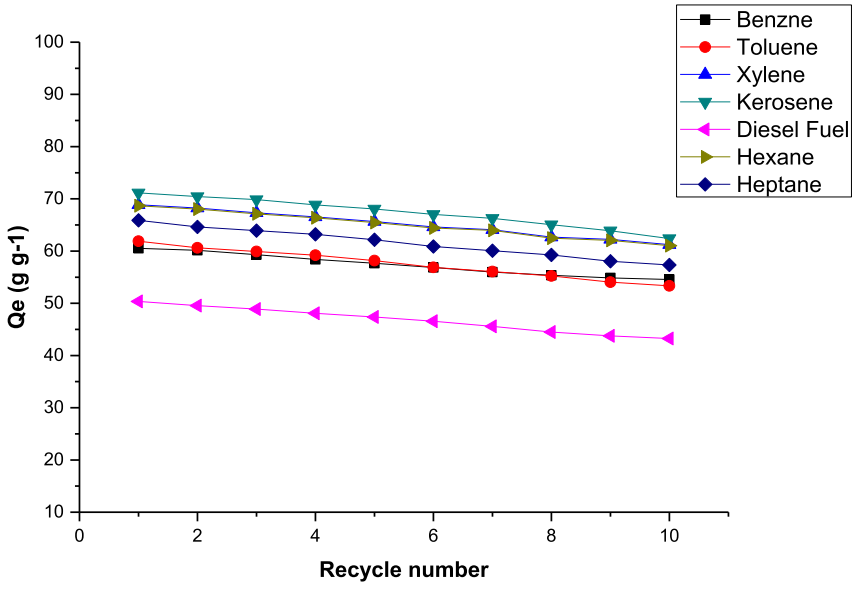

Fig. 14. Recyclability performances of the hydrocarbons adsorption capacities of wp-GO-MF tablets.

where the adsorbent was removed after 30 min ± 5 s of dipping in each cycle. The reason for its efficient adsorption was due to its robust pores as illustrated in the FESEM image. Most of the wp-GO-MF tablets were damaged or smashed after 13–16 cycles due to repetition of heating, while, no damage to the wp-GO-MF tablets was observed after the 10 cycles process indicating the successful reusability and recycling of this composite.

Conclusion

The composite of wp-GO, iron oxide magnetic nanoparticles, and melamine–formaldehyde polymer have been successfully prepared as tablets for oil adsorption. The wp-GO morphology had small sizes and flake-like structures with high aggregation and appeared highly electron transparent, as well as the graphene oxide demonstrated a wrinkled structure of a single or few layers. The m-Iron oxide NPs exhibit mostly spherical particles with a diameter of 8–10 nm and 40 emu/g saturation magnetization of no account coercivity revealing super-paramagnetic properties. It was found that the wp-GO-MF is highly porous according to the FESEM image. These desirable characteristics of the wp-GO-MF composite are the practical candidates for hydrocarbons adsorption. Sorption capacity for several hydrocarbons were examined utilizing the wp-GO-MF tablet. The wp-GO-MF have comparatively larger adsorption in benzene, toluene, xylene, hexane, heptane, and kerosene, while they exhibited lower absorption toward diesel oil. Conforming to the recent studded adsorption capacities, the prepared wp-GO-MF composite is considered advantageous to or better than

those sorbent materials previously reported. The pseudo-second-order kinetic model of different hydrocarbons absorbed by the wp-GO-MF composite expresses a high correlation coefficient (R^2) value (equal to 0.99) indicating that the experimental data fits the pseudo-second-order model, and the sorption capacity for the hydrocarbons is matched to experimental sorption capacity. Moreover, the wp-GO-MF composite establishes high recycling ability and reusability for the hydrocarbons tested in an oil system of 10 cycles.

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Authors' declaration

- Conflicts of Interest: None.
- We hereby confirm that all the figures and tables in the manuscript are ours. Furthermore, any figures and images, that are not ours, have been included with the necessary permission for republication, which is attached to the manuscript.
- No animal studies are presented in manuscript
- No human studies are presented in manuscript
- Ethical Clearance: The project was approved by the local ethical committee at the University of Basrah.

Authors' contribution statement

The proposed idea was conceived by M. Q. M. and H.B.A. The data was acquired by A. M. A and M. Q. M. The analysis was carried out by H. B. A, M. Q. M, and A. M. A. The data were interpreted by M. Q. M. and H.B.A The paper's drafter is H.B.A. The article was revised and proofread by M. Q. MK and H.B.A. All authors read the manuscript carefully and approved the final version of their MS.

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تحضير أوكسيد الغرافين من اقلام الغرافايت المستعمل و تحميله على سطح متراكب ميلامين فورمالديهايد/ غرافين و استخدامه لأزالة الهايدروكاربونات

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المستخلص

التطورالتكنولوجي الذي يشهده عصرنا الحاضر غالباً ما يصاحبه بعض الجوانب السلبية ومنها التلوث الهيدروكاربوني, هذا التلوث اصبح مشكلة ملحة تتطلب معالجة ملائمة وفعالة. تم اقتراح و تطبيق تقنيات متعددة لحل هذه الظاهرة تتضمن متراكب نانوي بوليمري. تضمنت الدراسة تحضيرطبقات قليلة متعرجة من أوكسيد الغرافين المشتق من أقلام الغرافيت (wp-GO) المستعملة حيث تم توصيفها باستخدام المجهر الإلكتروني النافذ ذي الدقة العالية (HRTEM). كذلك تم تحضير اوكسيد الحديد النانوي المغناطيسي اذ أضهر صفات مغناطيسية فائقة من خلال درجة تمغط مشبعة تبلغ 40 emu/g و قوة ممانعة مغناطيسية قريبة من الصفر, كان التركيب ذو اشكال كروية يبلغ القطر حواي 8-10 nm. تم استخدام هذه التراكيب النانوية كمتراكب مع بوليمر الميلامين فورمالديهايد. اضهر تشخيص المتراكب wp-GO-MF بوساطة HRTEM تركيب مستقر مسامي. تم تطبيق المتراكب wp-GO-MF لامدصاص عدد من الهايدروكاربونات, انجزت دراسة امدصاص الهايدروكاربونات و هي البنزين و التولوين و الزايلين و و الهكسان و الهبتان و الكيروسين و زيت الديزل. عرضت النتائج أن المتراكب wp-GO-MF قدرة امدصاص عالية (Qe) للكيروسين تصل بلغت 71 -g g 1. كذلك استقرت قدرة امدصاص الهايدروكاربونات بحدود 90% لعشر دورات امدصاص. لذلك يمكن استنتاج أن المتراكب wp-GO-MF أضهر قدرة امدصاص عالية لجميع الهايدروكاربونات المدروسة و بأداء تكرار ممتاز.

الكلمات المفتاحية: المتراكب، امدصاص الهايدروكاربونات، أوكسيد الحديد النانوي المغناطيسي، بوليمر الميلامين فورمالديهايد، أقلام الغرافيت المستعملة.