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Essential oil impregnation into graphene sponges with electric desorption control

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Abstract. This work shows the impregnation of scents using a graphene sponge (GS). This was functionalized by the modified Hummers method, pursuing to add different functional groups. It is proposed to achieve the release and seek to control it through electrical potential applied to the graphene sponge with essential oils. The graphene sponge was functionalized and steeped with two kinds of oil. The electrochemical study demonstrates the variation in the electrochemical behaviour of the functionalized graphene sponge without and impregnated with oil. The release of the oil and its aromatic scents was carried out by applying an electrical potential of 30 V, with a release rate of 1.86 mg/min. The heating of the sample that causes the release of oil, associated with the electrical resistance of the system, reaches temperatures of about 150°C. The essential oils, graphene sponge, surfactant, graphene sponge with essential oils, graphene sponge recuperated after applying electric potential, graphene sponge recovered by temperature and dipropylene glycol (DPG) were characterized using Fourier transformed infrared spectroscopy (FTIR), digital microscopy, and x-ray photoelectron spectroscopy (XPS).
Keywords: fragrance; graphene sponge; impregnation; odour; scent

1. Introduction

The controlled and sustained release of fragrances and different compounds is an application widely sought in fragrances (Ali *et al.* 2019, Ishiguro *et al.* 2018, Liu *et al.* 2018, 2019, Xiao *et al.* 2019), textile (Gonçalves *et al.* 2019, Sharkawy *et al.* 2017), food, and pharmaceutical industries (Paulo and Santos 2017), or simply to lengthen the release time of the fragrance (Ishiguro *et al.* 2019). The impregnation protects the active ingredients providing thermal stability and consumers are attracted to articles with a perception of lasting fragrance. Commercial essential oils are composed of a large number of compounds, in some cases, above 100 different organic molecules. Some can be lost due to their high volatility and oxidation in different environments, either heat or air exposure. Hence, the importance of implementing new methods of releasing essential oils that increase the lifetime of the scents in commercial products.

Encapsulation of scents can be attained using dextrins, which are heterogeneous, amorphous, and hygroscopic substances produced in large quantities for food, textile, paper, and other industries. They are used in products such as beer (Poór *et al.* 2018) and bread (Bär *et al.* 2020, Zhou *et al.* 2019a, b). β-cyclodextrin hydrate is a compound widely studied for encapsulation, primarily in food (Zhou *et al.* 2019a, b), textile (Xiao *et al.* 2019), and pharmaceutical industries (Andreadelis *et al.* 2020, Jaimes *et al.* 2016).

On the other hand, cleaning, remediation, or containment of spills is another application for various absorbent materials. Because of its absorbent features, it is a highly extended application for micro-porous natural materials, such as sawdust, perlite, wool, cellulose, and zeolite (Hadji *et al.* 2020). However, the natural characteristics of these micro-porous materials make them absorb a small oil load and are not selective. The extraction and recycling of these absorbent materials can be highly complicated (Joy *et al.* 2020, Nyssanbayeva *et al.* 2020, Panahi *et al.* 2020, Tayeb *et al.* 2020, Thakkar *et al.* 2020, Shin *et al.* 2020).

The absorbent polymers can absorb 5 to 25 times their weight in oils and organic solvents and have been used for load scents. These can be recycled and recover about 80% of their immediate previous capacity from absorbing in subsequent loads (Kizil and Bulbul Sommez 2017, 2020, Wang *et al.* 2019, Xia *et al.* 2019).

Nowadays, the development of absorbent and scent-releasing materials requires, in addition to the chemical

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structure for this purpose, characteristics such as high absorption, reuse, selectivity, high conductivity, and low production costs (Fan *et al.* 2020, Feng *et al.* 2019).

There is a growing interest in carbon materials, such as graphene (Ebrahimi *et al.* 2017, 2019, Kumar and Srivastava 2016, Moradi-Dastjerdi and Behdinan, 2019, Poór *et al.* 2018, Ramírez-González *et al.* 2020, Rodriguez-Pérez *et al.* 2017), which can absorb a load of up to 83 times its weight and, by vacuum filtration, allows for a recovery of up to 70% of the oil collected (Chabot *et al.* 2014). Graphenic materials have been used as adsorbents for various contaminants, such as dyes (Sun *et al.* 2020a, Thompson *et al.* 2018), antibiotics, and other pharmaceuticals (Ma *et al.* 2020, Mohamed *et al.* 2018, Solís *et al.* 2020, Yao *et al.* 2020), as well as organic solutions and metallic ions (Ma *et al.* 2020, Sun *et al.* 2020b, Yao *et al.* 2020).

Graphene-derived materials, such as graphene aerogel (Kovtun *et al.* 2020), graphene sponge (Sun *et al.* 2020a, Zhang *et al.* 2020), and graphene (Feng *et al.* 2019), are used for, among other things, the removal of contaminants. These absorbent materials, composed of carbon, show features such as high absorption efficiency, good reusability, and high selectivity to remove oils and organic compounds (Feng *et al.* 2019). The development of a quality spongy graphene for various functional applications is one of the technological trends of recent years in fields such as oil and organic solvents adsorption (Anand *et al.* 2020, Zhang *et al.* 2020), sensors (Mohamed *et al.* 2018), and super-capacitors (El-Gendy *et al.* 2019a, b).

In this work, a graphene sponge (GS) is used to absorb essential oils. The release is proposed by applying an electric potential using the low electric conductivity of this material, which causes heating.

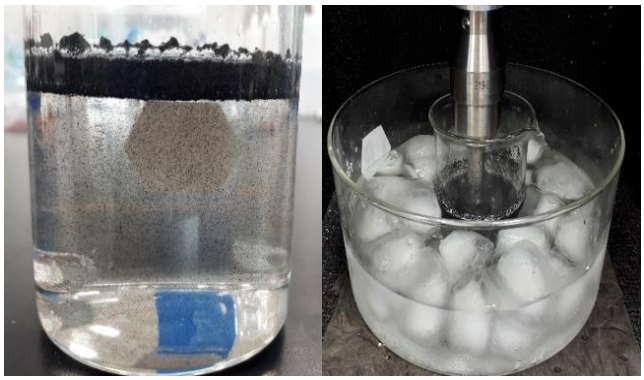
This temperature increment first causes the release of the more volatile molecules, most of the odor components, and finally, the oil as an aerosol of micrometric droplets.

2. Experimental

2.1 Preparation of the functionalized graphene sponge

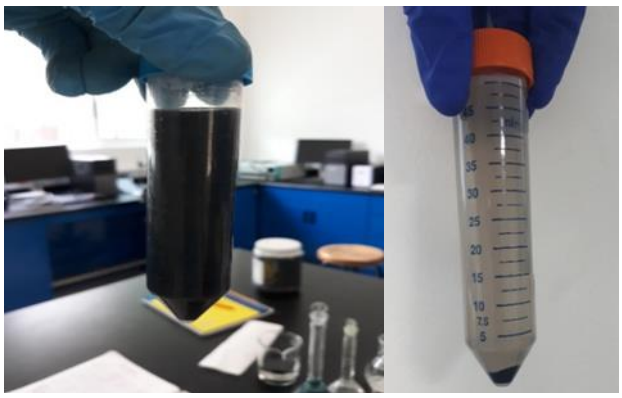
The modified Hummers method (Feicht *et al.* 2019, Mohammad *et al.* 2019, Yoo and Park 2019, Zaaba *et al.* 2017) was used to carry out the functionalization of the graphene sponge (obtained from *ID-nano Investigación y Desarrollo de Nanomateriales S.A. de C.V.*). The substances $NaNO_3$ (sodium nitrate), $KMnO_4$ (potassium permanganate), H_2SO_4 (sulphuric acid), and HCl (hydrochloric acid) used for experimentation were reagent grade.

A solution of 100 mL of 78% H_2SO_4 with 300 mg of graphene sponge and 300 mg of $NaNO_3$ was prepared (Fig. 1(a)). Also, a solution of 50 mL of 78% H_2SO_4 and 600 mg of $KMnO_4$ was prepared. The H_2SO_4/GS/$NaNO_3$ solution was treated with an ultrasonic



(a) (b)

Fig. 1 Process steps for the functionalization of GS samples: a) Acidic solution with graphene sponge having flocculated and suspended particles, b) ultrasonic bath in an ice bath of the GS solution



(a) (b)

Fig. 2 Solutions having graphene sponge a) before and b) after centrifugation

sonotrode for 2 min at an amplitude of 70% in an ice bath (Fig. 1(b)). Then, the $KMnO_4$ solution was slowly added, and the solution was treated with an ultrasonic sonotrode for 8 min. The washings were done by adding 20 mL of 5% HCl to the solution from the ultrasonic treatment and centrifuged for 20 min using 3800 rpm (Figs. 2(a) and 2(b)). After three washes with 5% HCl, the functionalized graphene sponge (FGS) was obtained and washed with deionized water.

2.2 Absorption of essential oils in functionalized graphene sponge (FGS)

A sample of 0.4 g of functionalized graphene sponge with 10 mL of water was added and subsequently treated in an ultrasonic bath for 20 min. This graphene sponge solution was mixed with 10 mL of water, 0.2 mL of *Triton-X100*® surfactant (Merck, CAS Number: 9002-93-1), and one mL of oil. The oils used were light mineral oil (Sigma-Aldrich, CAS: 8042-47-5) and a commercial oil solution of the *EPAF Monarca* brand, *Daydream*, which contained approximately 60% of propylene glycol and about 1% *Tween 20*®. The sample was centrifuged for 20 min,

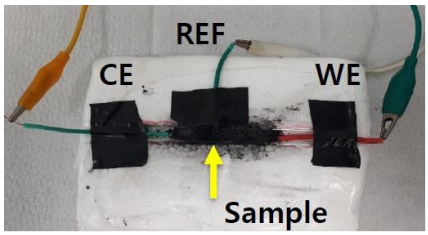
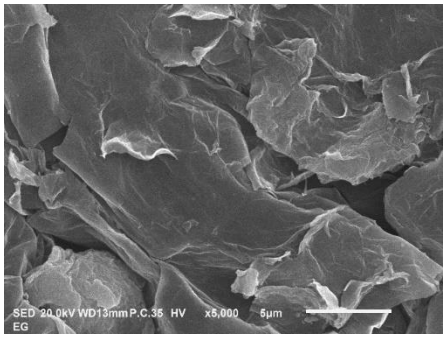
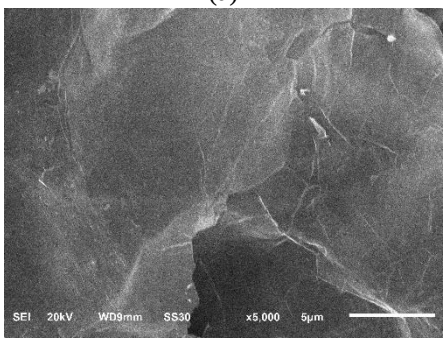


Fig. 3 Experimental setup of a graphene sponge functionalized and impregnated with an essential oil on a half-cylinder made of glass and electrically connected to a DC power supply



(a)



(b)

Fig. 4 SEM images obtained of (a) graphene sponge and (b) functionalized graphene sponge (FGS)

separated, and heat-treated in a furnace for 24 h at a temperature of 70°C.

2.3 Electrochemical tests

The functionalized graphene sponge impregnated with oil (FGSIO) was electrochemically tested with chronopotentiometry and pulse chronoamperometry using a BioLogic brand SP-300 potentiostat. The electrochemical system is shown in Fig. 3. A half-cylinder of glass was used as a base for the graphene sponge, using a reference pseudo-electrode (REF) with a distance of 3 cm between the counter electrode (CE) and the working electrode (WE). The FGSIO was compacted on the base. The chronopotentiometry tests were performed by applying a current of 240 µA for 60 min and recording its variation in potential. Pulse chronoamperometry tests were carried out by applying 0.1 and -0.1 V with pulses of 1 min for 60 min and evaluating their response in the current.

2.4 Release of essential oils with electrical potential

A laboratory DC power supply, GW brand model GPR-7510HD, was used to test the release of essential oils in the graphene sponge. The temperature measurement was done with a thermographic camera, PCE Group brand, model PCE-TC 4. A potential of 30 V was applied to the samples for 10 min, using a distance between electrodes of 3 cm, and the weight loss of the graphene sponge related to the release of the oil and other substances was recorded.

2.5 Characterization

The samples were characterized by Digital Microscopy. A Keyence brand microscope, model VHX-5000, was used. The magnification was done with 500x, where the sizes and distributions of the particles obtained were examined. A PerkinElmer brand Spectrum Two FTIR spectrometer model L160000A was used where a scan of wavenumber in the range of 4000 cm^{-1} to 450 cm^{-1} was performed. Also, an XPS characterization was done with a Thermo Scientific™ K-Alpha™ X-ray Photoelectron Spectrometer System, with an operating pressure of about 10^{-9} mbar, using an Al Kα monochromatic X-ray source (hν = 1486.6 eV), The XPS was used with a spot size of 400 µm, a number of scans of 10 and analyzer mode: CAE: Pass Energy 20.0 eV.

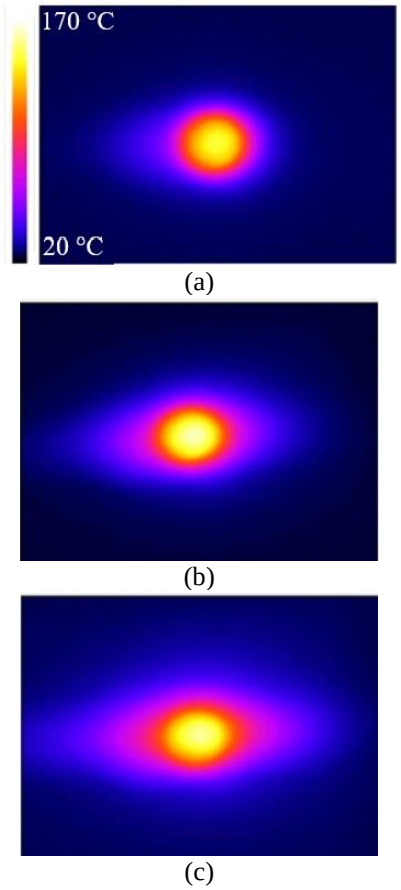
3. Results and discussion

The graphene sponge was functionalized and impregnated with oil. The observations were made with a digital optical microscope for each process stage to obtain the graphene sponge and subsequent impregnation with oil. Fig. 4(a) shows the non-functionalized graphene sponge, which was more heterogeneous than the functionalized sample.

The modified Hummers method allows us to have a more compact sample with fewer voids between particles, favoring the electrochemical study and the current flow for oil release. This can be seen in Figs. 4(a) and 4(b). In the first one, there is a slightly homogeneous surface with a large number of edges and reliefs. While, in the surface treated with the Hummers method, the surface of the particle has a more homogeneous and compact surface.

The treatment effectively reduces the number of holes in the graphene sponge. Figs. 4(b)-4(d) show the functionalized and oil-impregnated graphene foam. These were more compact and moist containing in appearance, such as a sponge. These types of samples had a consistency or viscosity similar to that of toothpaste (about 60,000 cps).

Fig. 5 shows the temperature variation. The release of oil can be attributed indirectly to the potential application due to the increase in temperature when the 30 V were applied to a sample.



(a)

(b)

(c)

Fig. 5 Thermographic images of the graphene sponge impregnated with *Daydream* oil showing the hot spots attributed to lower conductivity zones, which were taken at different times

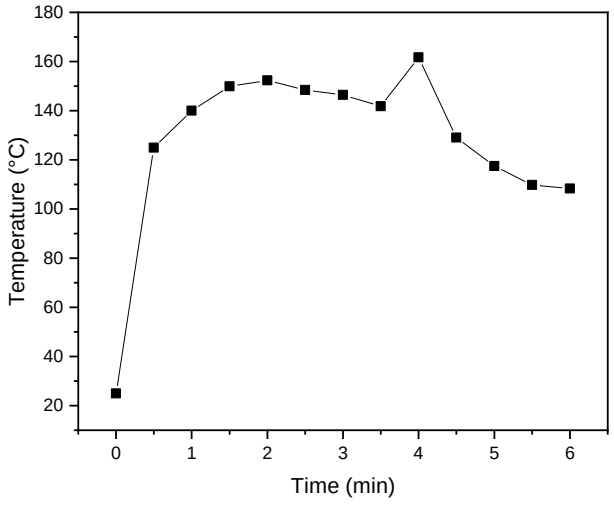
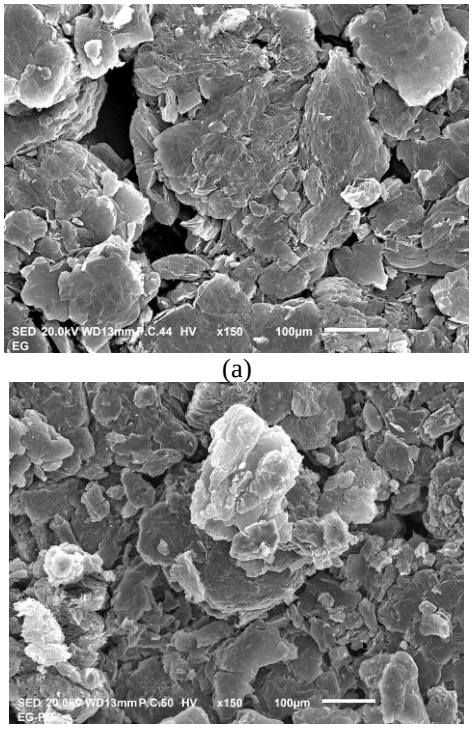


Fig. 6 Time vs. temperature graph

The initial temperature was 26°C, and when applying the potential, a rapid increase in temperature was observed due to the flow of current on the sample and the resistance of the functionalized graphene sponge impregnated with *Daydream* oil. At 30 s, the sample reaches 125°C (Fig. 5(a)). The temperature reaches its maximum peak, 148.4°C, at 2.5 min (Fig. 5(b)), and at 6 min drops to 108.3°C (Fig. 5(c)) because of the rearrangement and the increase of voids between particles.

Table 1 Values of the FGS weight changes under an electrical potential

Parameter	Value
Potential	30 V
Time	10 min
Initial Weight (W_i)	2.5104 g
Final Weight (W_f)	2.4918 g
$\Delta W = W_i - W_f$	0.0186 g
$\frac{W_{loss}}{t}$	1.86 $\frac{mg}{min}$
% $W_{loss} = \frac{Wi-Wf}{wi} * 100$	0.32%



(a)

(b)

Fig. 7 Scanning Electron Microscopy images obtained of graphene sponge (a) before applying the electric potential and, (b) after applying 30 V

Fig. 6 shows the temperature behavior in response to the application of potential. A flame was observed around the anode for a couple of seconds. Between minutes 4 and 4.5, the formation of a flame was observed for a moment, which was represented by a temperature peak at minute 4.

In the oil release analysis, by applying a potential, the amount of oil lost due to its possible release was measured. Table 1 shows the weight loss results of the graphene sponge due to the imposition of an electrical potential.

The application of an electric potential to the functionalized graphene sponge sample, having a load of *Daydream* oil, caused the volatilization of the oil, which was measured as a weight loss of 1.86 mg/min.

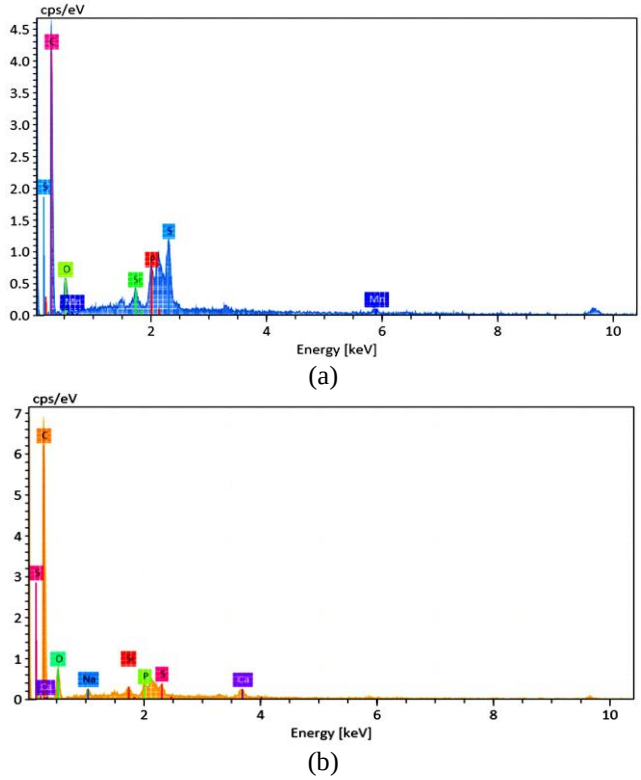


Fig. 8 EDS spectrum obtained of graphene sponge (a) before applying the electric potential and, (b) after applying 30 V

Table 2 Analysis of composition by EDS in wt.%

Element	GS	GS After potential
Carbon	72.40	71.71
Oxygen	20.51	24.37
Calcium	-	1.37
Sulphur	3.89	0.47
Phosphorus	2.24	0.64
Sodium	-	0.90
Silicon	0.04	0.90
Manganese	0.91	-

Since the beginning of applying electric potential, a white smoke release from the sample was observed, with a strong aroma of the scent of the *Daydream* oil.

SEM and EDS analyses were conducted to know the change in GS particles after applying the electric potential. Figs. 7(a) and 7(b) show FG before impregnating with oil and applying electric potential, and FGS impregnated with oil after potential application. SEM micrographs show no distinguishable differences or physical changes in the particles before and after the tests applying 30 V. Figs. 8(a) and 8(b) show the EDS spectra and Table 2 shows the composition results. There were no representative differences between samples. The major elements, carbon, and oxygen, have similar concentrations and the differences are in minoritarian elements that can be

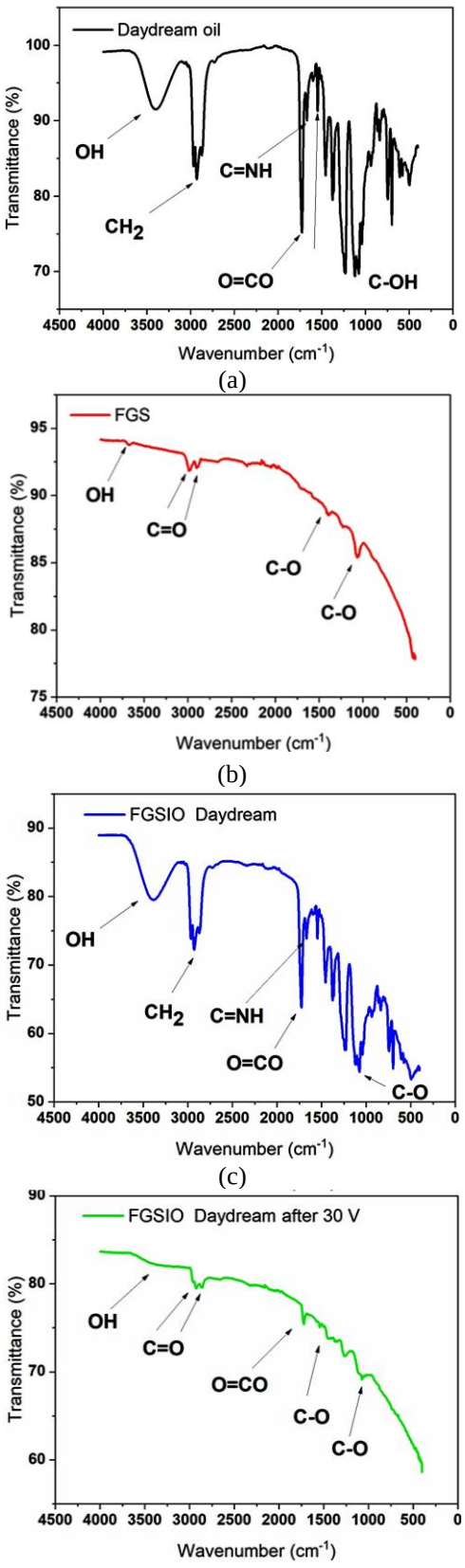
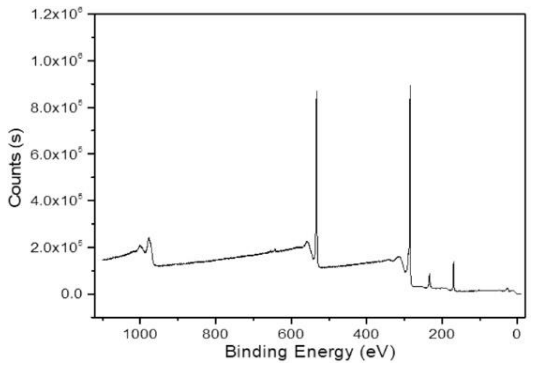


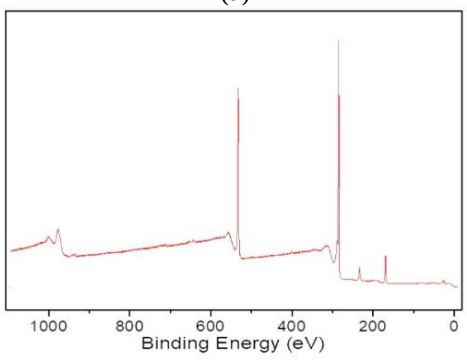
Fig. 9 Spectra of FTIR corresponding to (a) Dipropylenglycol (DPG), (b) Daydream oil, (c) FGSIO impregnated with Daydream, and (d) the functionalized graphene sponge with Daydream oil recovered after the application of electrical potential

Table 3 Values of the At. % in the FGSIO, FGSIO recovered after the application of electrical potential, and FGSIO after chronopotentiometry tests

	FGS	After 240 µA	After 30 V
O 1s	25.24	21.85	21.89
C 1s	69.72	73.86	76.9
S 2p	5.04	4.30	0.59



(a)



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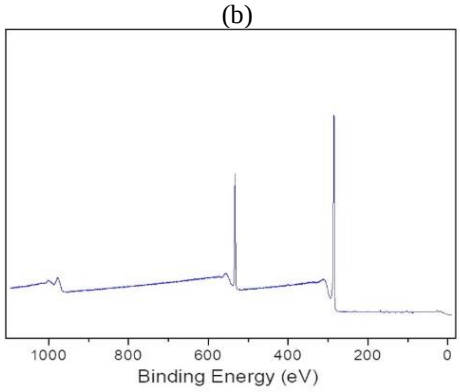

(c)

Fig. 10 XPS spectra corresponding to (a) FGSIO (before), (b) FGSIO after chronopotentiometry test (after 240 µA), and (c) FGSIO recovered after the application of electrical potential (after 30 V)

attributed to the oil in the sample after impregnation and application of potential.

The *Daydream* oil, the functionalized graphene sponge, functionalized graphene sponge with oil, and the graphene sponge after the release of oil by electric potential were characterized using a Fourier transform infrared spectroscopy (FTIR).

In Fig. 9(a), the spectrum of *Daydream* oil is shown, which has its characteristic peaks that belong to -OH, -CH_2, and C-OH, located at about 3399 cm^{-1}, 2966 cm^{-1}, and 1100 cm^{-1}, respectively. These are the vibration peaks reported in the bibliography for propylene glycol (Suganthi *et al*. 2013), which is the main component of this aromatic oil. There is a peak at 1730 cm^{-1} corresponding to a stretching of a carbonyl group – esters, specifically O=CO. At 1670 cm^{-1}, the peak belongs to a bending of the bond C=NH, and there is a tension in the C-OH bond that belongs to the ether group at 1070 cm^{-1}. These peaks are attributed to the aromatic compounds in *Daydream* oil.

Fig. 9(b) shows the spectrum of the functionalized graphene sponge, which indicates that FGS absorbs low energy, lowering the molecular vibration and the spectrum tends to go down. The absorption band at 3670 cm^{-1} is related to stretching vibrations of the O-H group. The absorption peaks at 2988 cm^{-1} and 2900 cm^{-1} can be attributed to carboxylic and/or carbonyl functional groups stretching. The absorption peaks at 1383 cm^{-1} and 1062 cm^{-1} are attributed to the C-O stretching vibrations. These absorption peaks are reported for graphene oxide by *Rattana et al*. (2012).

The presence of representative peaks found in *Daydream* oil can be observed in the infrared spectrum of the functionalized graphene sponge impregnated, Fig. 9(c), but with a minor intensity. These peaks are attributed to these groups being attached to the bond C-C due to the free electrons in this bond.

In the infrared spectrum of FGSIO, it recovered after applying an electrical potential (Fig. 9(d)). The spectrum is more like FGS without impregnation of *Daydream* oil, the characteristic peaks of the FGS spectrum remain in the same region, and the representative peaks found in *Daydream* oil disappeared. This indicates the release of absorbed oil because of the heating caused by the resistance to the applied electric current.

In the chemical composition analysis by XPS, the representative components of the samples were O, C, and S, with C1s being the main component of the graphene sponge.

Variations in the composition are observed in Table 3. After the chronopotentiometry tests, the graphene sponge sample impregnated with *Daydream* oil had a slight decrease in the At. % of O1s going from 25.24% to 21.89% and shows a reduction of S2p.

The most significant changes occurred in the composition of the sample after applying 30 V. In this sample, the atomic percentage of O 1s was 21.89%.

Then, it remained the same as in the sample after 240 µA, since it was 21.85%. After 30 V, due to the evaporation of the oil, the atomic percentage drops to 0.59%.

Fig. 10 shows the spectra of the samples, which are alike, comparing the functionalized graphene sponge samples and the samples after the chronopotentiometry tests (Fig. 10(b)). This is because, in the electrochemical tests, the potential values were too low, so they have no

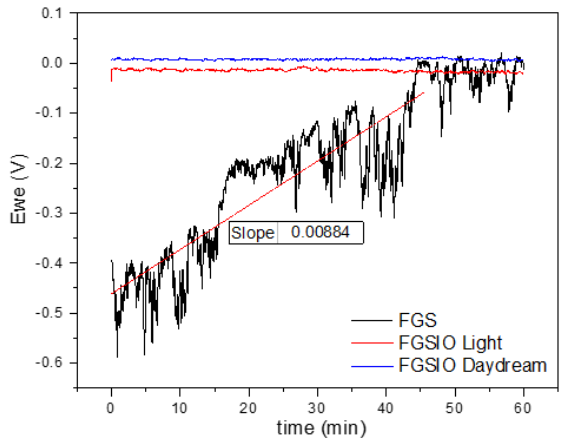

Fig. 11 Chronopotentiometry tests of functionalized graphene sponge

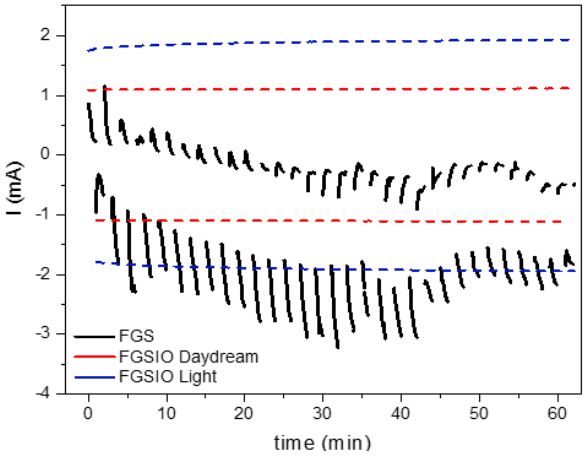

Fig. 12 Pulsed chronoamperometry tests of functionalized graphene sponge

heating of the sample, and there is no release of oil and essential. After applying a potential of 30 V (Fig. 10c), the spectrum of the sample no longer shows the peak of S2p below 200 eV due to composition changes by the oil release.

There were no chemical changes because of the low temperatures that the material reached during the flow. This allowed several impregnation-release cycles to be carried out.

Electrochemical tests evaluated the behaviour of functionalized graphene sponge impregnated with the mineral light-oil and *Daydream* oil. Figs. 11 and 12 show the results of the electrochemical tests.

In chronopotentiometry tests (Fig. 11), the behaviour of the functionalized graphene sponge shows potentials of one order of magnitude greater than the oil samples. The graph shows a growing profile with a slope of 0.00884 and high fluctuations, which was attributed to a reduction in surface area that reaches a zero slope in a time of 45 min. Both samples with oils had stable behaviour, with a zero-slope profile and small fluctuations. The FGS with *Daydream* oil remained at 0.008 V, while the FGS with the mineral light oil had a potential of -0.015 V throughout the entire test. The difference between them can be attributed to the difference in composition since the mineral light oil is a pure oil and *Daydream* oil is an essential oil solution with a great diversity of chemical composition (polypropylene glycol, *Tween 20*®, and others).

The difference in behaviour, between the functionalized graphene sponge samples and those containing oils, was attributed to the high resistance to the current that the oil gives to the impregnated samples.

A layer of oil around the particles increases the electrical resistance because of the compatibility between the liquid and solid.

In Fig. 12, the pulsed chronoamperometry graph is shown. It is observed that the two samples impregnated with oil have similar behaviour. FGSIO Light presents current values of 1.89 and -1.9 mA, while FGSIO *Daydream* has a response to currents of 1.10 and -1.10 mA. The mineral light-oil impregnated graphene sponge has a 0.8 mA current window higher than the *Daydream* oil. This difference between the samples is due to the composition of the oils since the components of the *Daydream* oil (propylene glycol, *Tween 20*®, and others) increase the electrical resistance of the material, and the mineral light-oil, being simpler in composition, has a lower resistance to the current flow. This increase in resistance to current flow favors the heating of the sample by applying higher voltages and, thus, indirectly achieves the liberation of the oil and its scent by applying electrical power.

In the case of the FGS sample, irregular electrochemical behaviour was observed. In the first three minutes, the system goes through a transition period until it reaches a behaviour with repeatability for the rest of the test. In these three minutes, a drop in current from 0.9 mA to 0.3 mA was observed on the anode side and an increase in current on the cathodic side from -0.9 mA to -0.3 mA. Correspondingly, there was a current drop to -0.5 mA in the same pulse. After this, there was a decrease in current in the anode part from 1.1 mA to 0.3 mA. The system acted differently in these three pulses than in subsequent pulses. In the successive pulses, a tendency towards the negative direction was found. This tendency is proposed to occur because of the accumulation of charge (e^-) in the cathodic part and its subsequent release in the anode part. From minute 3 to minute 45, the system tended to the cathodic side. From minutes 45 to 60, there was a slight variation in both directions. This was attributed to a rearrangement of the functionalized graphene particles.

4. Conclusions

This work proposes a methodology for absorbing and releasing essential oils with a graphene sponge by applying an electric potential. There was heating in the graphene sponge loaded with the oils by applying an electrical current. The temperature reached 147.4°C when applying 30 V for 10 min, using a distance between electrodes of 3 cm, to a compact sample on a

grooved system. This heating causes the release of the scent, which was observed as a mass loss of the graphene sponge sample. A scent release rate of 1.86 mg/m was obtained, which was controlled by fluctuating the process parameters. The SEM micrographs allowed to identify that none of the treatments applied to the graphene sponge samples implied a change in morphology or chemical composition. In the composition analyses, the decrease in the atomic percentage of S2p was attributed to the oil loss after applying a potential of 30 V. Therefore, the releases of aroma and oil sequentially were achieved by heating the functionalized graphene sponge samples by applying an electric potential. FT-IR spectra show that the application of electrical potential slightly decreased the concentration of essential oil in the graphene sponge and was demonstrated that the application of potential is a feasible way to liberate aromatic essentials absorbed in the graphene sponge without provoking changes in the base material.

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