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SYNTHESIS AND CHARACTERIZATION OF FUNCTIONALIZED EPOXY/SIO_2 HYBRID WITH GRAPHENE OXIDE NANOSHEETS

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Abstract. Nanocomposites were prepared and characterized with a functionalized epoxy resin hybrid (REF) with SiO_2 nanoparticles, synthesized by the *in-situ* sol-gel process, and graphene oxide (GO) nanosheets. The epoxy resin is synthesized with bisphenol A and epichlorohydrin for its subsequent functionalization with abietic acid, providing –OH groups having a greater number of active chemical sites on the surface so that they can join with the SiO_2 particles synthesized *in situ* from TEOS and modified-Hummers GO. The nanocomposites were prepared with REF and a solution of TEOS 40 v/v%; to this hybrid material (HREF), two concentrations of GO at 1 wt% (HREF1) and 5 wt% (HREF5) were added. All materials were characterized by spectroscopic techniques FT-IR and Raman: showing groups -(COOH) from abietic acid, silanol -OH, which will bond with the same groups in the GO sheets. Thermogravimetric analysis (TGA) revealed that SiO_2 nanoparticles decorated the basal plane of GO by covalent bonding TGA, increasing the thermal stability at 50 °C, HREF5 being the material with the highest degradation temperature. A homogeneous dispersion of SiO_2/GO decorated sheets in the functionalized epoxy was studied using the SEM technique, with HREF1 as the most homogeneous. ASTM D2369 establishes that volatile organic content should not surpass 3.4 g/mL, and the materials prepared have only 0.23 g/mL, which marks the first step to achieve real applications in several industries.

Keywords. Functionalization epoxy resin, Graphene oxide nanosheets, sol-gel process, SiO_2-epoxy hybrid

1. Introduction

With the continuous ambition of improving and modifying the properties of polymers, research has been carried out where nanofillers materials are introduced as modifiers of the matrix thermal and mechanical properties.[1] Modification by elastomeric materials has also been studied to improve the properties of several epoxy networks[2] and a good synthesis can be found in the review made by Frigione.[3] On the other hand, the epoxidation of phenolic Novolac and Cresol Novolac resins are studied, which are also forms of modified epoxy resins, and their properties are improved in terms of adhesion and chemical resistance.[4] By incorporating specialized additives, commonly nanoparticles, and modified chemical structures, the study of Brzozowski demonstrated enhanced fire resistance while maintaining the mechanical integrity of the resins,[5] being deeply relevant for aerospace, automotive, and construction applications where fire safety is a critical concern.

Other investigations have added rigid inorganic particles creating a coarse composite material in order to increase hardness.[6] Or the studies to devise a way to embed inorganic particles of nanometric size for the manufacture of resins resistant to tarnishing and scratching.[7] However, a greater scientific approach has been presented in the epoxy resin treated with a sol-gel polymerization process, which allows the creation of a porous material that admits small particles of inorganic materials, generally ceramics. Introducing itself like this, hybrid materials are a type of nanocomposite material.[8] Having these nano size fillers permits the use of the new nanocomposite as the continuous phase for macro composite materials, *i.e.* glass or carbon fiber composites.[9]

In China,[10] research continued to evolve, and the appropriate concentrations of silicone-epoxy-silica are being investigated to make coatings on aluminum 2024. Also, an approach similar to the objective of this work is presented, studying the interfacial bond between titania and graphene modifying an epoxy resin applied to con-

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crete.[11] Advances in epoxy resin formulations aim to enhance their mechanical performance, fire resistance, viscoelastic properties, and durability.

In the last decade, graphene oxide (GO) sheets have been growing in applications of all types owing to their high specific surface area and unique structure, consisting of a quasi-two-dimensional honeycomb network of carbons that make up the structure (sp2 domains) that is altered because these GO sheets are highly functionalized with different oxygenated groups such as epoxide, carboxyl, hydroxyl, and carbonyl. Basal planes are linked with epoxide and hydroxyl groups, in addition to hydrogen bonds and Van der Waals-type interactions, while the edges are specially functionalized with carboxyl groups that provide the material with colloidal stability and a negative surface charge that is pH dependent.[12] Having high thermal and electric conductivity, excellent mechanical properties, low density, and ion impermeability has become a good option for polymer reinforcement.[13] Recent studies show how GO can act as an ion barrier to delay corrosion and simultaneously toughening the material.[14,15] Despite all the improving in nanocomposites, there is no reported research on using an abietic acid-epoxy matrix nor *in situ* synthesis with SiO_2.

In the same line of carbon allotropes, nanotubes have been studied in combination with different nanoparticles as nanocomposite fillers, commonly as heat stabilizers with electric properties. Materials can be obtained from high purity chemicals or bio-waste with a well established process.[15]

Epoxy-novolac copolymers incorporating polycyclic bisphenols of norbornane explore the development of new modified resins that exhibit superior thermal stability and mechanical performance due to the rigid structure of norbornane, making them suitable for coatings of high-performance requirements and structural applications.[16] Moreover, research about the incorporation of polysulphide rubber enhances the toughness and flexibility of epoxy composites has been investigated, focusing on dynamic mechanical properties, improved impact resistance and elongation without significantly compromising stiffness and thermal stability, which is crucial for applications requiring high durability and elasticity.[17]

On the other hand, crucial advancement is presented in epoxy polymer-inorganic composites with modified minerals,[18] which investigates the reinforcement of epoxy resins with modified oxide minerals. This modification, often called fillers, improves the mechanical strength, thermal resistance, and barrier properties of the composite materials, making them suitable for protective coatings and high-stress structural components, which is further discussed in articles related to inorganic fillers, mainly Si, Ti and Zr.

More recently, hydrophobic spherical micro-/nanostructures comprising α-Al_2O_3/ZrO_2 particles, prepared using combustion synthesis-assisted water atomization and chemical modification methods, along with a one-step spray coating process embedded in epoxy resin, had great results in antifriction and corrosion resistance.[19] Parallel to this investigation, a study of the effect of titania (TiO_2) nanoparticles on the viscoelastic behavior of epoxy composites.[20] Using a sol-gel approach for nanoparticle dispersion, the study reveals significant improvements in thermal stability and modulus retention under dynamic mechanical conditions, indicating potential applications in electronics and high-temperature environments. Mechanical and electrochemical testing of an epoxy hybrid material with SiO_2 can be found in the work of Tesillo *et al.*[21,22]

Finally, not only mechanical and corrosion resistance is pursued but also microbial protection; the research of these materials integrates antimicrobial agents with epoxy matrices, offering long-term durability and mechanical corrosion resistance in harsh environments, such as marine and industrial settings.[23] These studies collectively contribute to the advancement of epoxy-based materials, demonstrating significant improvements in mechanical performance, durability, fire resistance, and specialized applications. By modifying the epoxy resin formulations with various additives and reinforcements, researchers continue to expand the utility of these materials across diverse industries.

The objective of this study is to elaborate a covalently bonded interface between epoxy functionalized with SiO_2/GO hybrid and characterize the material with diverse techniques to confirm the correct bonding as well as to investigate the effect of GO concentration on the mechanical properties as a coating material. The synthesis is an *in-situ* sol-gel synthesis based on tetraethyl orthosilicate (TEOS) as a silica precursor, mineral graphite as a precursor GO nanosheets and abietic acid functionalized epoxy resin (FER). The volatile organic content of liquid nanocomposites with 0%, 0.1 wt% and 0.5 wt% GO concentrations was also measured for further possible applications complying with the normativity.

2. Experimental

2.1. Materials

Mineral graphite (~90%) grounded powder was characterized and used. Sulfuric acid 98 wt% (H_2SO_4), chlorohydric acid 30% (HCl), hydrogen peroxide 30% (H_2O_2) and potassium permanganate 99.5 wt% ($KMnO_4$) for the synthesis of graphene oxide (GO) were acquired from Sigma-Aldrich. Ethanol 98 wt% (C_2H_5OH), tetraethyl orthosilicate 98 wt% (TEOS), and sodium hydroxide 98% (NaOH) were purchased from Sigma-Aldrich. For epoxy resin, bisphenol A 97%, and Epichlorohydrin 99% and to functionalize it, abietic acid 97% ($C_{20}H_{30}O_2$) were purchased from Sigma-Aldrich.

2.2. Methodology

2.2.1. Synthesis of GO

Using a modified Hummers method[24] to synthesize graphite oxide (GrO) from mineral graphite, the process began with the dispersion of 2 g of graphite powder in 45 mL of H_2SO_4 at -5°C in an ice bath under constant stirring. Gradually, 6 g of $KMnO_4$ was added while maintaining the temperature below 20°C. After homogenization, the mixture was removed from the ice bath and heated to 35°C to initiate the reaction, which was sustained for 2 hours. Subsequently, 92 mL of distilled water was added dropwise while stirring continued for 15 minutes. The reaction terminated by pouring the mixture into a vessel containing 270 mL of distilled water and 10 mL of H_2O_2 (30% v/v).

The product was then washed and centrifugated using 400 mL of an HCl solution (2.5% v/v) to remove metal ions, followed by rinsing with distilled water until a neutral pH was achieved. Graphene oxide was then obtained by exfoliating the stacked graphite oxide sheets. This was done by dispersing 300 mg batches of GrO in distilled water (10 mg/mL) and subjecting them to ultrasonic bath treatment (42 kHz ± 6%) for 3 hours. The resulting product was dried to obtain black graphene oxide (GO) powder.

2.2.2. Functionalized and Synthesis Epoxy Resin Hybrids

The synthesis was carried out with 22.8 g (0.1 mol) of bisphenol A and 20 mL (0.24 mol) of epichlorohydrin; as catalyst agent 20 mL of 0.86M NaOH solution was used,[25] which was incorporated dropwise into the reaction. The three reagents are kept under vigorous stirring and at a temperature of 95 °C for 30 minutes with reflux and distillation. It resembles a commercial formulation for the epoxy resin of Mn=700. Subsequently, 0.85g (0.003 mol) of abietic acid is added, and the mixture is left under reflux for another 20 minutes at a temperature between 95 °C to 115 °C. Obtaining 20mL of volume distilled and 30mL of FER.

The sol-gel[26] hydrolysis process was made with an EtOH : TEOS solution (5 : 13.75 v/v) mixed with a Water:EtOH:GO (5:20:x v/v/m) solution previously dispersed, with constant stirring and heating up to 95 °C, where x represents the concentration of GO, specifically 0 g, 0.04 g and 0.2 g of GO powder for the three hybrids. Passed 30 minutes, 40 g of FER was added dropwise, and the mixture was refluxed for 1 hour at a temperature of 105 °C verifying the viscosity.

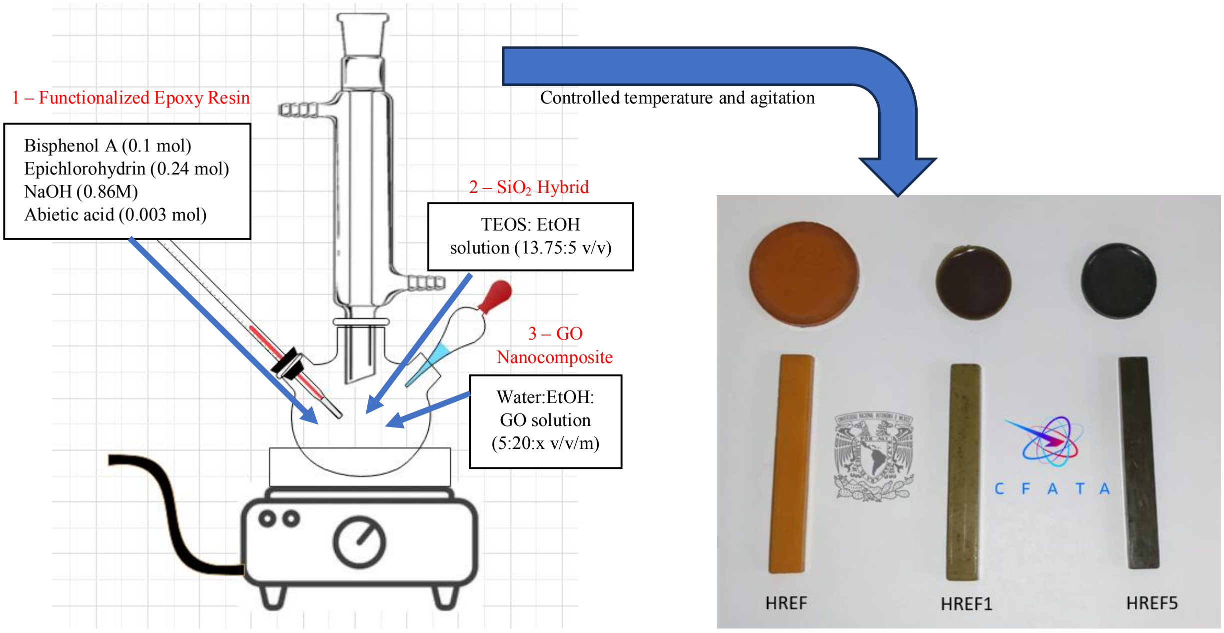

Fig 1. Schematic of the three-step synthesis to obtain the functionalized epoxy resin (REF), the hybrid with SiO_2 (HREF) and the nanocomposites with SiO_2 and 1 and 5% of GO (HREF1 and HREF5 respectively)

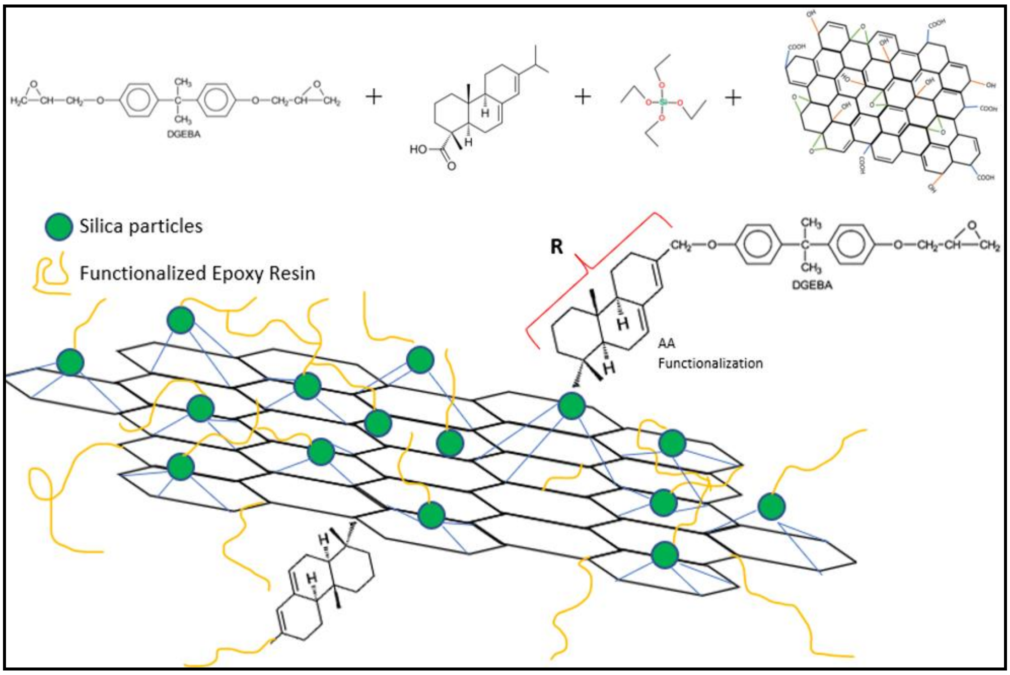

Fig 2. Schematic representation of the structure and interaction between the SiO$_2$ particles, the GO sheets and FER

2.3. Characterization

For X-Ray Diffraction (XRD), a Rigaku Ultima IV diffractometer, equipped with an X-ray generator with a max power of 3 kW, voltage of 40kV, and current output of 2-60 mA from 5° to 80° was used to verify the oxidation of mineral graphite and the exfoliation to obtain GO sheets.

Spectroscopically, for the Fourier transformed infrared (FT-IR) spectra measurements were performed on a Thermo Scientific Model 6700 Nicolet with ATR technique using a scan speed of 0.32s, a gain of 8, and a voltage (Vpp) of 5.75V and recorded in the 4000-400 cm^{-1} wavenumber range at a resolution of 4cm^{-1}. For Raman dispersive spectra, a Bruker Model Senterra with a 20x optical microscope was used, with a resolution of 9-15 cm^{-1}, an integration time of 2s, 8 scans, power of 25 mW and recorded at the 100-3400 cm^{-1} range.

For scanning electron microscopy (SEM), a SU8230 Hitachi SEM/TEM CFE was used and the samples were AMS gold coated to fully observe the dispersion quality and the morphology of the hybrids.

Thermogravimetric analysis (TGA) measures were obtained using a TGA/DSC Mettler Toledo 0-1000 °C thermal analyzer. The experiments were performed under a nitrogen atmosphere at a flow rate of 40 mL/min and a 10 °C/min heating rate from room temperature (25 °C) to 250 °C.

3. Results and Discussion

3.1. GO nanosheets characterization

X-ray diffraction (XRD) and dispersive Raman spectroscopy techniques are performed to confirm the graphene oxide synthesis. For mineral graphite as obtained, we can observe in Figure 3 the graphite characteristic peak at $2\theta=26.45°$ (002) and the peaks with lower intensity (110) and (222) also assigned to graphite with hexagonal structure.[27]

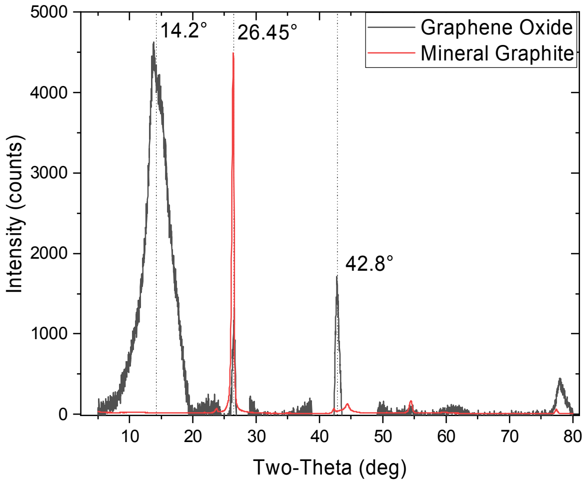

Fig 3. X Ray Diffractogram for mineral graphite and graphene oxide to confirm the synthesis

Figure 3 shows the XRD for GO, after the modified Hummers method, with the presence of the peak at $2\theta=14.2°$ (d-spacing 6.49Å) characteristic of Graphene Oxide (GO), the peaks that appear at $2\theta=26.45°$ and $2\theta=42.8°$ indicate that there is still a presence of mineral graphite in the sample.

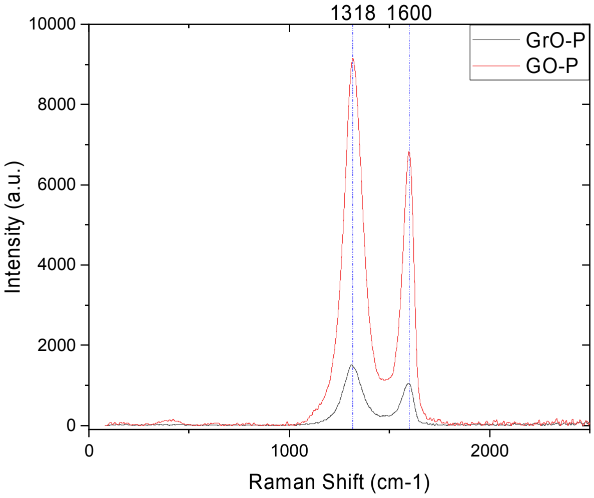

Fig 4. Raman Spectra for Graphene Oxide (GO-P) and Graphite Oxide (GrO-P)

Fig. 4 shows the Raman spectrum and makes clear the difference for Graphite Oxide and Graphene Oxide, where the D band is shown at (1318 cm^{-1}), and the G band is shown at (1600 cm^{-1}) for both GrO and GO.[28]

3.2. FT-IR analysis

Figure 5a shows the FT-IR spectra of the neat epoxy resin (RE) and the functionalized epoxy resin (REF) in a liquid state, the absorption band at 1701 cm^{-1} is present only in the functionalized epoxy resin and corresponds to the group C=O of abietic acid, which indicates the functionalization, and the presence of this group (C=O) allows the union with the SiO_2 particles and graphene oxide (GO).

Figure 5b shows the infrared spectra of the different hybrid materials to determine the functional groups present in each one. The labels correspond to Functionalized Epoxy Resin Hybrid with 0% GO (HREF), Functionalized Epoxy Resin Hybrid with 0.1 wt% GO (HREF1), and Functionalized Epoxy Resin Hybrid with 0.5 wt% GO (HREF5). The band at 3342 cm^{-1} is attributed to the secondary alcohol resulting from the opening of the epoxy ring by the Bisphenol A salt. The bands at (2968, 2830 cm^{-1}) and (1717, 1370 cm^{-1}) are due to the vibration of the $-CH_2$ and abietic acid C=O groups, respectively. The DGEBA molecule is characterized by three bands at 1250 cm^{-1} that represent the aromatic carbon-oxygen vibration of Bisphenol, 1038 cm^{-1} resulting from the aliphatic carbon-oxygen bond ($-O-CH_2-$), and a small band at 950 cm-1 which is attributed to the epoxy ether group. The band at 1381 cm^{-1} is attributed to carbon with dimethyl groups bonded to the benzenes of bisphenol A. The bands at (1510, 829 cm^{-1}) are due to the phenyl groups. The bands with the greatest difference occur at (1083, 1041 cm^{-1}), which are attributed to the presence of graphene oxide and bonds from TEOS, with Si-O-Si being present, stretching of Si-O-C bonds and Si-OH bonds.[29]

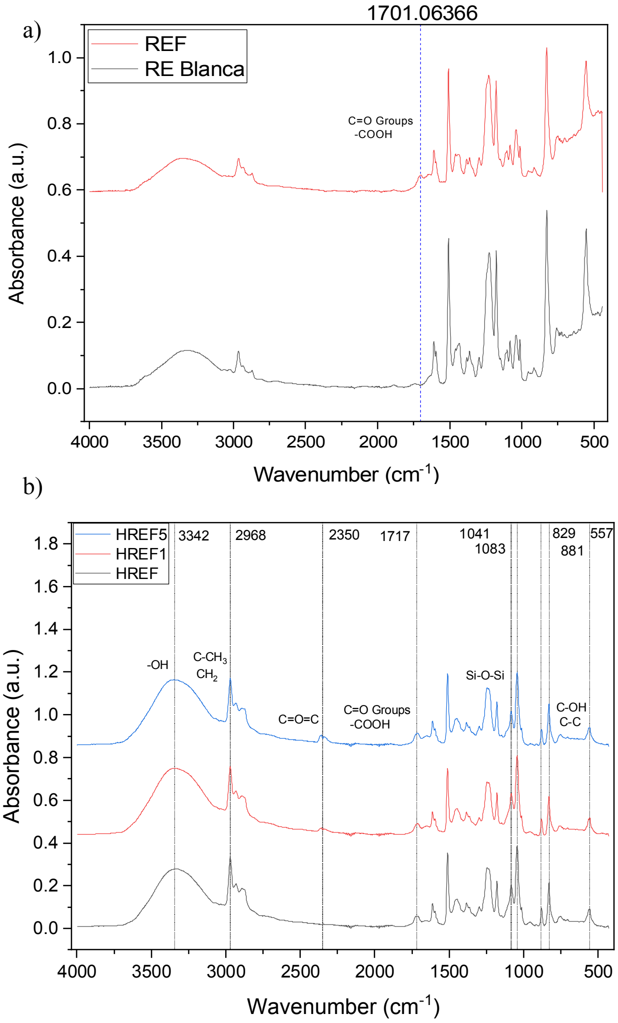

Fig 5. a) FT-IR Spectra of Neat Epoxy (RE) and Functionalized Epoxy Resin (REF) and b) FT-IR Spectra of the hybrid materials

3.3. Raman analysis

In Figure 6 the Raman spectra for the hybrid materials are shown, neat epoxy nature bands are distinguished at (1607 cm^{-1}) due to carbon-carbon double bonds vibrations. Between the range of (1230 cm^{-1} to 1280 cm^{-1}) the bands correspond to the common vibrations of the epoxy radical and the band at (916 cm^{-1})

is due to the deformed epoxy rings, being less intense. The bands at (643 cm^{-1} and 1458 cm^{-1}) identify the aromatic rings present in the polymeric resin and the bands after (2800 cm^{-1}) correspond to the CH_2 and CH_3 groups. Additional characterization is needed to provide a solid indication of the functionalization; this is due to the concentration and the location of the bands for abietic acid, which practically coincides with the characteristic bands of the clear epoxy resin.

Bands of interest are in the locations at (1183 cm^{-1}) caused by the Si-O-Si bond and bands at (1050 cm^{-1}) assigned to Si-O bonds. The band found at (~390 cm^{-1}) corresponds to the formation of the SiO_2 network.[30] The intensity of the band at (1608 cm^{-1}) decreases meanwhile broadening is also perceived as a function of the increase in GO concentration, indicating an interaction between the resin and the SiO_2/GO in the hybrid materials. This behavior indicates that there is graphene oxide with silica particles decoration inside in an epoxy medium.

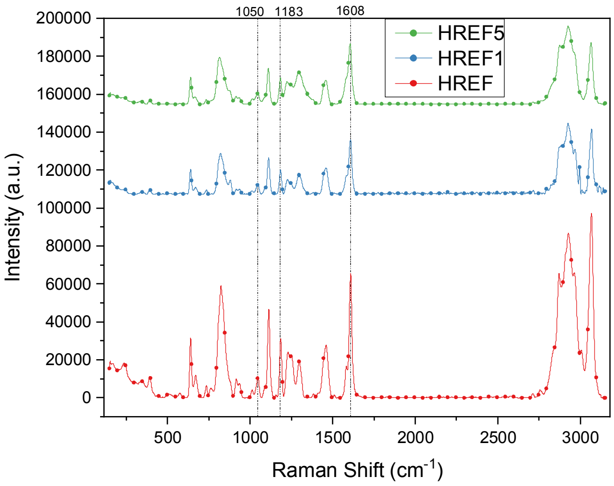

Fig 6. Raman Spectra of the hybrid materials

3.4. TGA analysis

For thermal characterization, a TGA was conducted for Epoxy Resin (RE), Abietic Acid Functionalized Epoxy Resin (REF), HREF, HREF1, and HREF5, the results are shown in Figure 7. The thermogram curves of RE and REF show only 1 step of fast degradation starting at 310 °C, the residual weight of 20 % at 700 °C indicates that most parts of the resins were volatile gases at high temperatures. On the other hand, the three hybrid materials presented a two-step weight loss at 100-380 °C and 380-750 °C temperature regions. The first step is related to the release of physiosorbed water molecules, and some excess reagents attached in the sol-gel process, and the second one is due to the organic volatile content nature of the epoxy matrix. All samples present a rapid degradation after 300 °C, highlighting the viability for aeronautical operational conditions. The incorporation of SiO_2 and GO improves noticeably the thermal stability of the epoxy resin at about 50 °C. This can be explained by the SiO_2 decoration of GO sheets lowering the hydrophilic nature and enhancing interfacial adhesion between phases.

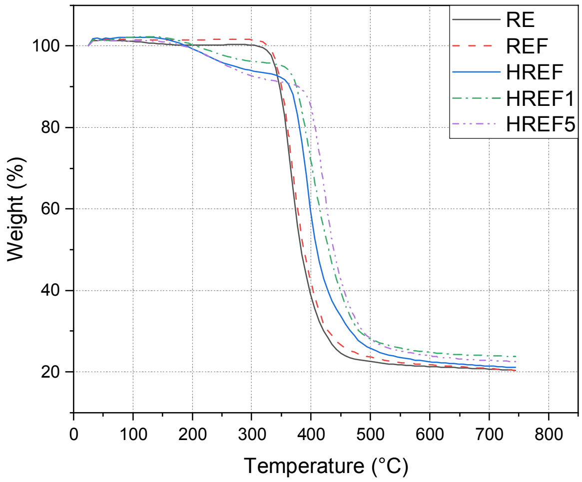

Fig. 7. Thermogram for RE, REF, and nanocomposites materials

3.5. Volatile Organic Content (VOC)

A regulation required to be a commercial product in all sectors and especially aeronautical is the *ASTM D-2369-20 Standard Test Method for Volatile Content of Coatings*,[31] which represents the mass loss of a container containing the coating in a liquid/gel state after 2 separate intervals of 48 hours. The calculation of the volatile matter (V_A) percentage in liquid coatings is determined as follows:

$$\%Volatile\ matter\left(V_A\right)=100-\left[\frac{W_2-W_1}{S_A}\cdot 100\right] \qquad (1)$$

Where, $V_A =$ %Volatile matter (first 48 hours); $W_1 =$ weight of dish; $W_2 =$ weight of dish plus specimen after heating; = specimen weight; V_B %Volatile matter (second 48h interval, calculated in the same manner as V_A).

These results must be compared with the aeronautical requirement of volatile organic content and must not exceed 3.4 g/mL. A summary is presented in Table 1.

Table 1. Volatile Organic Content Results for Hybrid Materials

MATERIAL	HREF	HREF1	HREF5
Volatile matter (V) [%]	21.19	21.72	21.46
Not volatile matter (N) [%]	78.81	78.28	78.54
VOC [g/10mL]	2.31	2.33	2.32

3.6. SEM analysis

To compare morphological and dispersion rate between the hybrid materials and to study the effect of GO structures within the materials, the SEM technique and analysis were carried out. Figure 8a shows the microphotography of HREF where the dispersion of silica spheres between 528 nm and 1.22 µm embedded in the polymeric matrix can be observed, and spheres are made up of a single unit or an agglomerate of smaller particles as shown in the close-up of Figure 8b. This dispersion is exhibited in 80% of the material surface, and the remaining surface presents a homogeneous phase.

On the other hand, in Figure 8c we can notice that the hybrid materials containing GO display a structure where the surface of the GO sheets have smaller silica particles around 24-34 nm in size adsorbed to them; in Figure 8d a close-up is shown. Also, we can observe that the dispersion of the sheets is not completely homogeneous but remains stacked, which indicates that the GO sheets held strongly together due to Van der Waals interactions and hydrogen bonds even after the curing and drying process. This indicates that during the sol-gel synthesis GO dispersion also played the role as a size modifier, and a structure where the silica particles can bond. This SiO_2 decoration is present in both materials HREF1 and HREF5 with no big difference in GO concentration.

These decorated sheets can intuitively explain why GO and silica can be presented as a corrosion protection mechanism, it represents a hydrophobic barrier role where corrosive agents as H_2O, Cl^- and O_2 cannot penetrate easily due to the blocked micro-pores and free volume (common in organic coatings) that acts as a diffusion pathway for corrosive species toward the metal interface, improving the protective performance. Moreover, adhesion can also be enhanced due to the high surface area provided by GO sheets and the chemical affinity between decorating Si and the Si-contained substrate, like glass or aluminum alloy 6061.

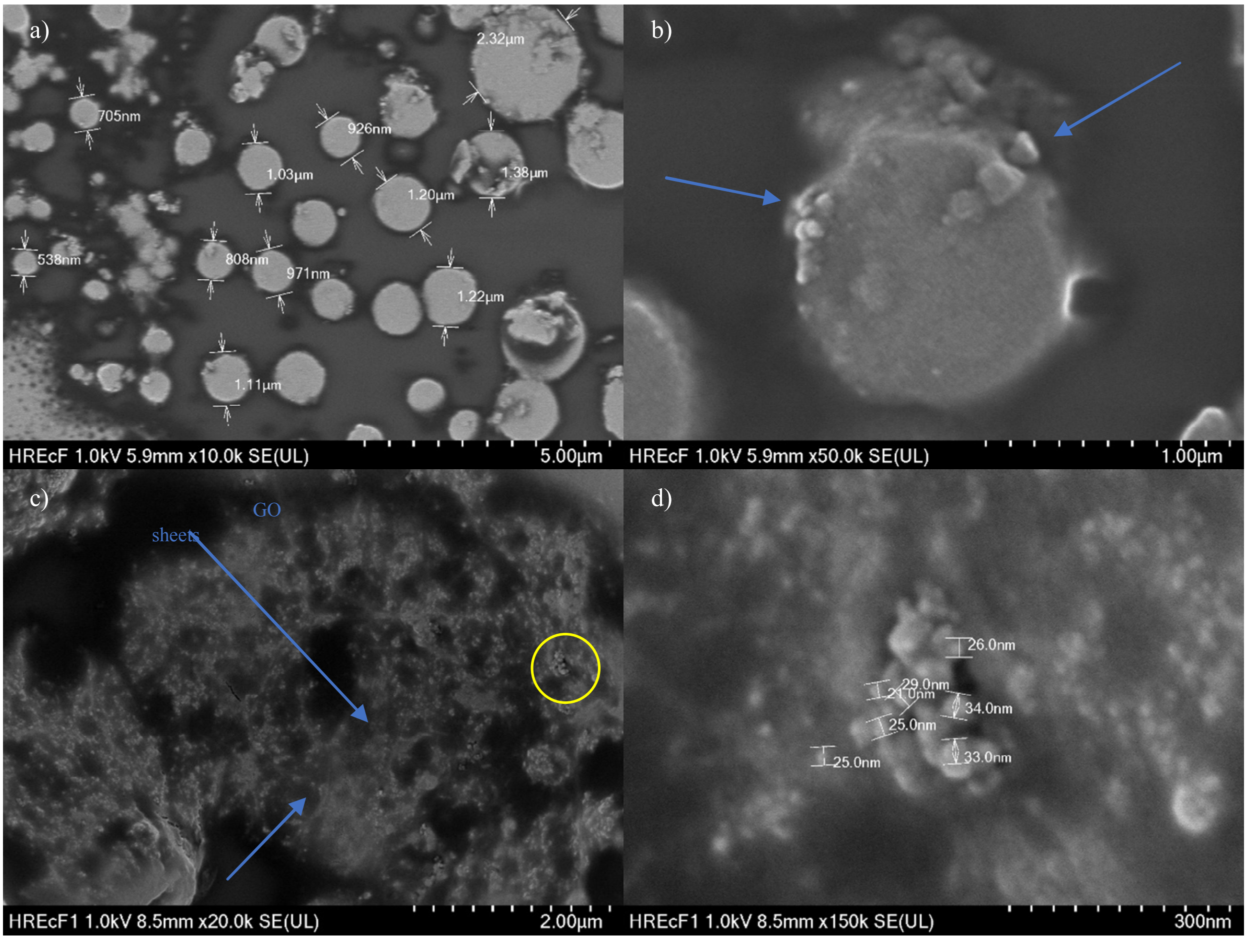

Fig. 8. a) SiO_2 particles in the hybrid material without GO with digital measures; b) close-up of one of the spheres; c) mesh of GO nanosheets with SiO_2 on its surface in the hybrid materials and d) a close-up with digital measures on the encircled area

4. Conclusions

The Raman and FT-IR spectroscopy results corroborate that the functionalization of the RE was carried out with abietic acid due to the presence of the absorption band at 1701 cm^{-1} of the carboxyl functional group, while for the composite materials, GO appears in the band at 2350 cm^{-1} in the IR spectra. In the Raman analysis, GO is not observable due to its low concentration; the concentration of abietic acid should be recalculated to obtain a better signal in the IR spectra and Raman.

The morphology showed that the SiO_2 particles are embedded in the polymeric epoxy matrix. Hybrid coatings with GO, presented nano and microlayers decorated with silica particles which could promote greater adhesion and hydrophobicity on different substrates, such as glass, aluminum alloys, steel, and copper.

Real potential applications in several industries are possible, as the thermal degradation results improved from 40°C to 60°C compared to the functionalized epoxy resin, this is a thermal improvement of 8% to 11.5% compared to the non-functionalized resin. Regarding volatile content, the results comply with the ASTM D2369 standard for application as an industrial coating, presenting only 0.23 g/mL of VOC. It is concluded that further tests are necessary to evaluate the enhancement of the mechanical properties and corrosion resistance to be used as a protective coating.

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СИНТЕЗ І ХАРАКТЕРИСТИКА ФУНКЦІОНАЛІЗОВАНОГО ГІБРИДУ ЕПОКСИДНОЇ СМОЛИ/SIO_2 З НАНОЛИСТАМИ ОКСИДУ ГРАФЕНУ

Анотація. *Одержано й охарактеризовано нанокомпозити з функціоналізованим гібридом епоксидної смоли (REF) з наночастинками* SiO_2*, синтезованими за допомогою in-situ золь-гель процесу, і нанолистами оксиду графену (GO). Епоксидну смолу синтезовано з бісфенолу А й епіхлорогідрину для подальшої функціоналізації абієтиновою кислотою, як джерело –OH-груп для забезпечення більшої кількості активних хімічних центрів на поверхні, щоб вони могли з'єднуватися з частинками* SiO_2*, синтезованими in situ з TEOS, і модифікованого Гаммерс-GO. Нанокомпозити були одержані з REF і розчину TEOS 40% об.; до цього гібридного матеріалу (HREF) додавали GO в двох концентраціях: 1% мас. (HREF1) і 5 % мас. (HREF5). Усі матеріали були охарактеризовані методами ІЧ- з перетворенням Фур'є та Раман-спектроскопії: виявлено групи –COOH абієтинової кислоти, силанольні –OH, які будуть зв'язуватися з такими ж групами в листах GO. Термогравіметричний аналіз показав, що наночастинки* SiO_2 *декорують базальну площину GO через ковалентне зв'язування, підвищуючи термостабільність за 50°C, і HREF5 є матеріалом з найвищою температурою деградації. Гомогенна дисперсія декорованих* SiO_2*/GO листів у функціоналізованій епоксидній смолі була досліджена за допомогою методу SEM, і HREF1 виявився найбільш гомогенним. Стандарт ASTM D2369 встановлює, що вміст летких органічних речовин не повинен перевищувати 3,4 г/мл, а в отриманих матеріалах він становить лише 0,23 г/мл, що є першим кроком до досягнення реальних застосувань у кількох галузях промисловості.*

Ключові слова: *функціоналізація епоксидної смоли, нанолисти оксиду графену, золь-гель процес,* SiO_2*-епоксидний гібрид.*