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Synthesis and analysis of flakes graphene oxide

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The presented article is devoted to the synthesis and analysis of flakes graphene oxide obtained by the Hammers method. The synthesized flakes graphene oxide was studied using SEM, EDX, X-ray diffraction, Raman spectroscopy, element analysis, temperature dependent of resistance and IR spectroscopy. As a result of calculating the results of X-ray analysis according to the Debye-Scherer's formula, the thickness of graphite flakes was about 12,9nm and the number of layers was 38. The result of the Raman analysis show that high quality flakes grapheme oxide was obtained. Based on the result of elementary analysis of grapheme oxide mass, the C/O ratio was determined to be 1,42. The grapheme layers inside the sample were 3,31nm thick and 14,8nm long by scanning electron microscope. The temperature variation of the resistance was determined. IR spectroscopy shows the results of the absorption of electromagnetic radiation in the infrared range by atomic groups of reduced grapheme oxide and the excitation of the molecule by light quanta. When a molecule is irradiated with infrared radiation, it is shown that only quantum absorption quantities are formed according to the frequencies of the molecules.

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1. Introduction

Modern research is mainly devoted to the study of limited-size systems. Thus, graphite and its modifications are considered to be the core of these systems and have unique features. Graphite is a system formed by layers of graphene. In addition, graphene oxide (GO) is one of the most widely studied and promising derivatives of graphene due to its electronic, mechanical, thermal and optical properties. Graphene oxide should be thought of as graphene with defects by applying oxygen groups. The grapheme quasi, which has been widely studied in moder times, is a two-dimensional system, for the first time in 2004, A.K.Game and K.S.Novoselov graphite using the simple band method by obtained [1]. In [2], the authors analyzed some graphene-based properties using X-ray diffraction analysis, Raman scattering, and IR luminescence. Addition structure, purity, quality and surface morphology, as well as homogeneity or heterogeneity of graphene-based samples were analyzed. Analysis of graphene-based samples was performed using a scanning electron microscope (SEM), energy dispersion analysis (EXD), X-ray diffraction analysis, Raman scattering, and IR luminescence. It was found that the addition of carboxyl group had a strong effect on the physical properties of the sample [3]. The physical properties of graphene-based samples have been comparatively studied [4]. In, [5] the synthesized a graphene-based sample and analyzed its structural, morphological and electrical properties and identified the possibilities of application. The recently developed pitch-based graphitic foams have a very high thermal conductivity to weight ratio. This property allows graphitic foams to be used in several thermal management applications, especially in the aeronautics and aerospace industries. Raman spectroscopy studies were performed on the different structural regions of the foam, yielding important information on the structural properties of the graphitic foams, as well as on the physical properties of graphite. The graphitic foam was found to be composed of two intermixed graphitic structures, one with stacked planes and one with a turbo stratic structure. This special structure allowed for a simultaneous study of the properties of two-dimensional (2D) and three-dimensional (3D) graphitic structures. The dispersion of the G′ band was found to be different for 2D and 3D

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graphite. The intensity of the D-band Raman feature was used to probe the density of defects in the structure, leading to the conclusion that the defects are mainly localized in the 2D structures. A simple model is proposed to explain the origin of the two different structures within the graphitic foam. Also, the dependence of the G′ band on polarization is addressed [6]. Recent Raman scattering studies in different types of graphene samples are eviewed here. We first discuss the first-order and the double resonance Raman scattering mechanisms in graphene, which give rise to the most prominent Raman features. The determination of the number of layers in few-layer graphene is discussed, giving special emphasis to the possibility of using Raman spectroscopy to distinguish a monolayer from few-layer graphene stacked in the Bernal (AB) configuration [7]. Highly oxidative debris (OD) was obtained by aqueous ammonia wash of as-prepared grapheme oxide (GO) which composed of OD and lightly oxidative GO sheets. The magnetic properties of OD and GO were studied [8]. The application of the kinetic properties of graphene and its derivatives in spintronics is of particular interest. Long spin diffusion lengths and associated times resulting from the weak spin-orbit of graphene and high subtle interactions can provide ideal conditions for coherent spin manipulation [9]. The electrochemical performance of $Ni(OH)_2$/ reduced graphene oxide nanocomposite obtained by ultrasonic treatment of hydrothermal synthesized β-$Ni(OH)_2$ and rGO has been investigated in alkaline medium. The material demonstrates good specific capacitance and is quite cost effective. The kinetics of proton migration in electrode materials has been investigated. The increasing of capacitance for $Ni(OH)_2$ / reduced graphene oxide composite up to 494 F/g in comparison with 121 and 133 F/g for rGO and $Ni(OH)_2$ at current density 0.08 A/g are attributed to the synergistic effect due to conductive system of grapheme nanosheets formation and enhancement of both electron and ion transport [10]. The aim of this paper is the comparison of structural, morphological and electrical properties of thermally extended graphite synthesized by chemical oxidation of graphite with sulfur of nitric acids at all other same conditions. Thermal treatments of graphite intercalation compounds were performed at a temperature of 600°C on the air for 10 min but additional annealing in temperature range of 100 - 600°C for 1 hour was done. The obtained materials were characterized by XRD, Raman spectroscopy and impedance spectroscopy. The evolution of structural ordering of thermally extended graphite samples at increasing of annealing temperature was traced. It was determined that the additional annealing allows to control the electrical conductivity and structural disordering degree of extended graphite samples that is useful for preparation of efficient current collectors for electrochemical capacitors [11]. In this study, mixed matrix membranes (MMMs) consisting of graphene oxide (GO) and functionalized graphene oxide (FGO) incorporated in a polymer of intrinsic micro porosity (PIM-1) serving as a polymer matrix have been fabricated by dip-coating method, and their single gas transport properties were investigated. Successfully surface-modified GOs were characterized by Fourier transform infrared spectroscopy (FTIR), UV-Vis spectroscopy, Raman spectroscopy, scanning electron microscopy (SEM), and thermo gravimetric analysis (TGA). The effect of FGO loading on MMM morphology and performance was investigated by varying the FGO content in polymer matrix from 9 to 84 wt.%. Use of high FGO content in the polymer matrix helped to reveal difference in interaction of functionalized fillers with PIM-1 and even to discuss the change of FGO stiffness and filler alignment to the membrane surface depending on functional group nature [12]. Raman spectroscopy is one of the most accurate tools for characterizing the structure, disorder level, and modification of carbon-based materials [13].

2. Experimental details

2.1. Synthesis and characterization

The synthesis process can be described as follows: A certain amount of 10ml of 96% solid sulfuric acid is added to a beaker. 1g of high-dispersion graphite (99.9995% purity) is poured mechanically with 0.5g of sodium hydroxide in a petri dish well and on top of solid sulfuric acid. The remaining amount (13g) is added to the system with a magnetic stirrer, adding solid sulfuric acid. Potassium permanganate (3g) is added to the system in portions within 2 hours. During this period, the mixture comes into contact with the ice bath and the temperature is maintained at 20 degrees. After mixing is stopped, the system comes into contact with the hot water bath steam. This time the temperature is maintained at 35 degrees. Thus, the mixture is obtained in the form of a chestnut-gray paste. 46ml of distilled water is added to the mixture. Immediately put to boil in a

boiling water bath. After 1 hour, mix by adding 250ml of distilled water. After a while, 100ml of perhydrol is added and mixed. A change in color is noted. Thus, the formation of a yellowish compound in chestnuts visually proves the production of graphene oxide. The mixture is filtered after the temperature drops. The remaining substance on the filter paper is washed with distilled water. The substance obtained at a temperature of 50 degrees for 2 hours dried in a vacuum dryer during the process. In the end, the substance is obtained in powder form when completely dry. 1.8grams of the substance is obtained, which corresponds to a loss of 40% [4,12].

The structure and purity of graphene oxide, quality and surface morphology, SEM, XRD, Raman spectroscopy, IR-luminescent, and temperature dependence of resistance properties of graphene oxide were analyzed.

2.1.1. SEM and EDX analysis

Figure 1 shows images of grapheme oxide samples according to the scanning electron microscope (SEM). The images prove it graphite oxidation, the number of layers decreases the crystalline decreases and amorphization accurse.

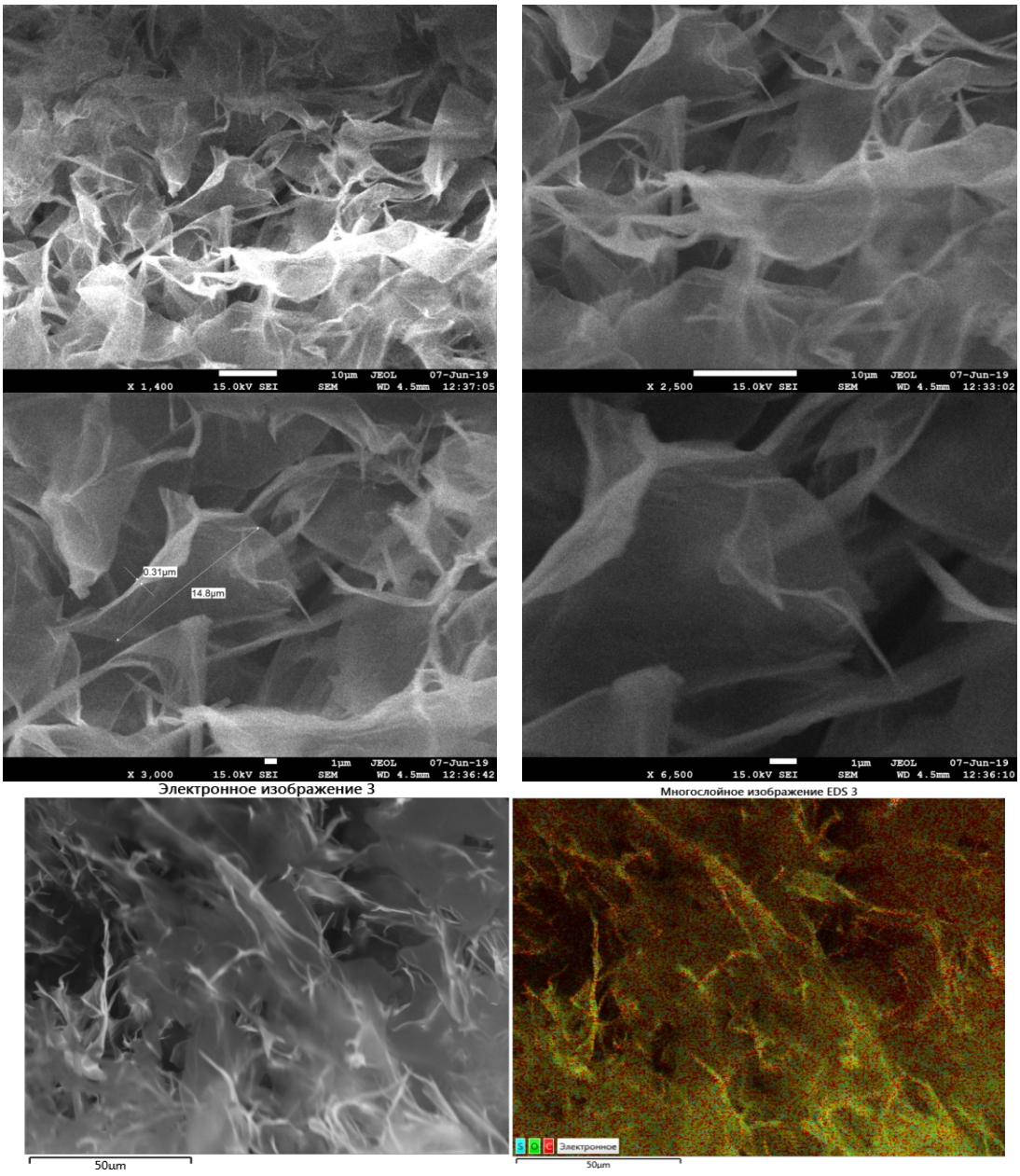

Fig. 1. Scanning Electron Microscopy (SEM) images of graphene oxide flakes (a), Size distribution and thickness of the flakes grapheme oxide (b).

Table 1. Elemental analysis of graphene oxide.

Samples	Element content [wt.]%				C/O ratio	C/H ratio	C/N ratio
	C	H	ON	N			
Graphene oxide	48.1	3.06	45.2	-	1.42	1.32	-

Table 1 above shows the result of the elemental analysis of grapheme oxide. According to the results of elementary analysis of graphene oxide mass, the C/O ratio is 1.42. This is a high degree oxidation, which was accompanied by the largest interlayer distance. The thickness and length of the graphene layer are shown in the 1400 - 4000 magnification range of the microscope, as shown in Fig. Thus, the layer is visually visible with a thickness of 0.31 nm and a length of 14.8 nm. According to the results of energy-dispersion analysis (EDX) of graphene oxide sample, it was determined that the ratio of C=70%, O=30%, as can be seen from the spectrum of the synthesized sample.

2.1.2. XRD analysis

The structure of low-graphene oxide obtained by the Hammers method was studied using X-ray phase analysis. The spectra of the graphene oxide sample were analyzed by X-ray diffraction analysis using an D2 Phaser (Bruker) diffractometer with CuK_{α} rays (λ=1.5406 Å) at an angle of $2\theta = 0.5^0$-80^0. Diffraction peaks, the intensity of which depends on the morphological characteristics of the samples, allow to obtain information about the distance between the graphene oxide sample and the layers.

Describes the XRD analysis of the GO sample as shown in the graph. Graphite flakes exhibit a sharp 002 reflection at ~26.3° corresponding to its intermediate layer. If we use Debye-Scherer's expression [15] for this sharp peak, the thickness of the graphite flakes is about 12.9 nm and the number of layers here while we would have received 38.

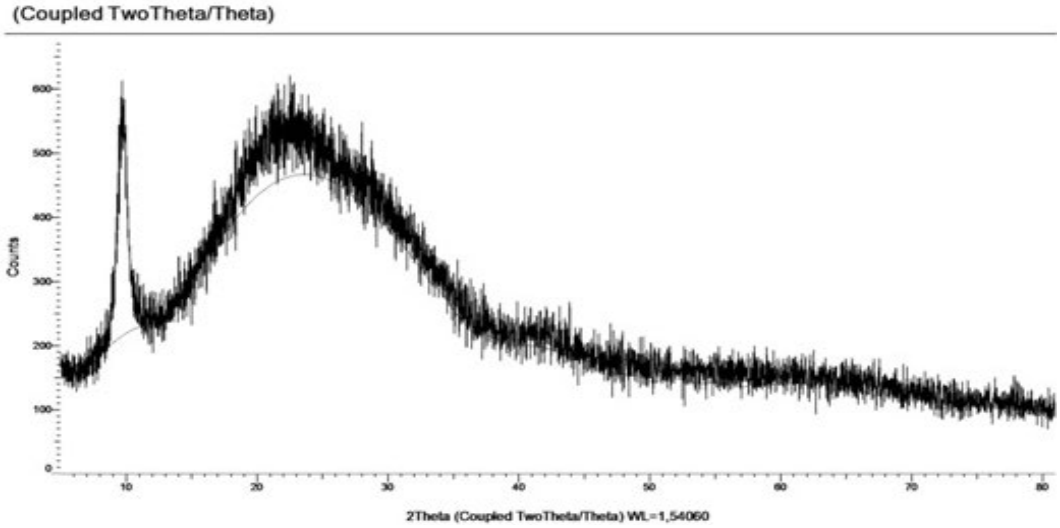

Fig. 2. X-Ray diffraction pattern of the flakes grapheme oxide [4].

2.1.3. Raman analysis

Raman scattering spectra were measured on a Nanfinder 30 (Tokyo Instrument, Japan) confocal Raman micro spectrometer. An Nd: YAG laser with a wavelength of 532 nm and a maximum power of 10 mW was used as an excitation source. In this case, the spectral resolution is

0.5 sm^{-1}. A CCD camera cooled to (-70^{0}S) and operating in phonon counting mode was used as a detector. Raman spectroscopy is a method of molecular spectroscopy to observe in elastically scattered light and is important for determining phonons. The first peak observed in the spectrum is the D peak, which is related to the defects of the sample. characterizes the oxygen groups present inside graphene oxide when examined. The second peak observed is the G peak. this peak is due to the scattering of light from the Brillion zone. This peak has a double shear, which is typical for all specimens with sp^2 hybridization. The third peak corresponds to the resonance scattering of light consisting of two phonons with the same energy and momentum in the opposite direction. A sharp peak observed in the 2D range, indicates the presence of a layered graphene.

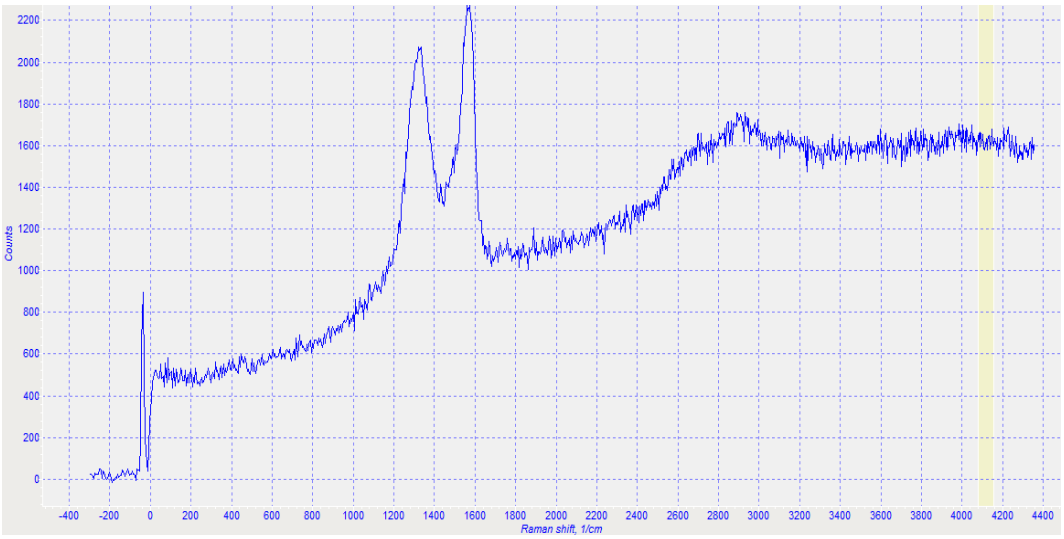

Fig. 3. Raman spectroscopy of the flakes graphene oxide [4].

According to the Raman scattering curve, graphene oxide varies depending on the thickness of the different layers. Thus, as shown in Figure 4, according to the Raman analysis, the D peak is 1320 sm^{-1}, the G peak is 1582 sm^{-1}, the 2D peak is 2720 sm^{-1}, the D+D/ peak 2910 sm^{-1}, the 2D/ peak 3190 sm^{-1} , the ID/IG~ 0.83 and the I2D/IG~ 0.496 respectively. The 2D peak observed here shows the number of graphene layers. The results of the Raman analysis show that high quality low-graphene oxide was obtained. The results of the Raman analysis confirm the results of the X-ray phase analysis. Thus, it should be noted that using the Hammer method is high low-quality graphene oxide was obtained.

2.1.4. Temperature dependence of resistance

To measure the temperature dependence of the resistance, the sample was continuously heated and then cooled. Then the values of resistance corresponding to changes in temperature are noted. The figure shows the results of the curves of graphene oxide heating 1 and cooling 2 in the temperature range 1-220^{0}C. As can be seen from the graph, the analysis of the heating and cooling curves showed that the temperature was 9.84-6.5·10^9 Ohm, respectively. Approximate stability is observed in the graphene oxide sample in the cooling curve in the range of 10-25^{0}C. In the range of 120-178^{0}C, a sharp decrease is observed in accordance with the law of activation. With the help of other properties of graphene oxide, such as Raman, X-ray phase and IR spectroscopy, the nature of the carbon-carbon bonds that make up the layer helps to obtain information about the acoustic and optical phonon lines that form the elemental core of the molecule.

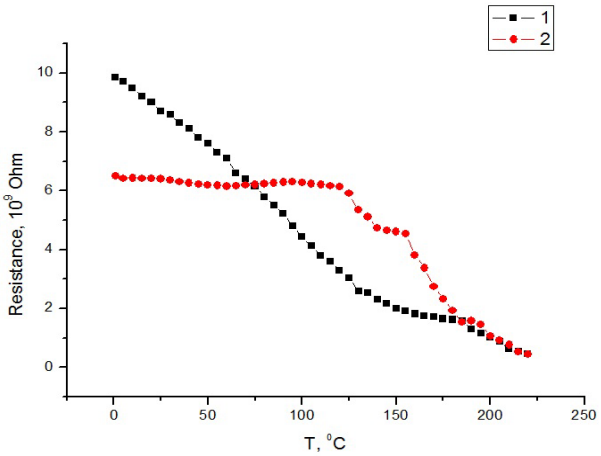

Fig. 4.Temperature dependence of resistance of flakes graphene oxide.

2.1.5. Luminescent analysis

IR spectroscopy is one of the most intensively studied methods for determining and evaluating the chemical composition of a material. It also helps to identify the functional groups that exist within graphene oxide. Fourier-IR absorption spectra of carbon nanotubes samples were obtained in the range of 4000÷400 sm^{-1} on a Varian-640IR IR spectrometer.

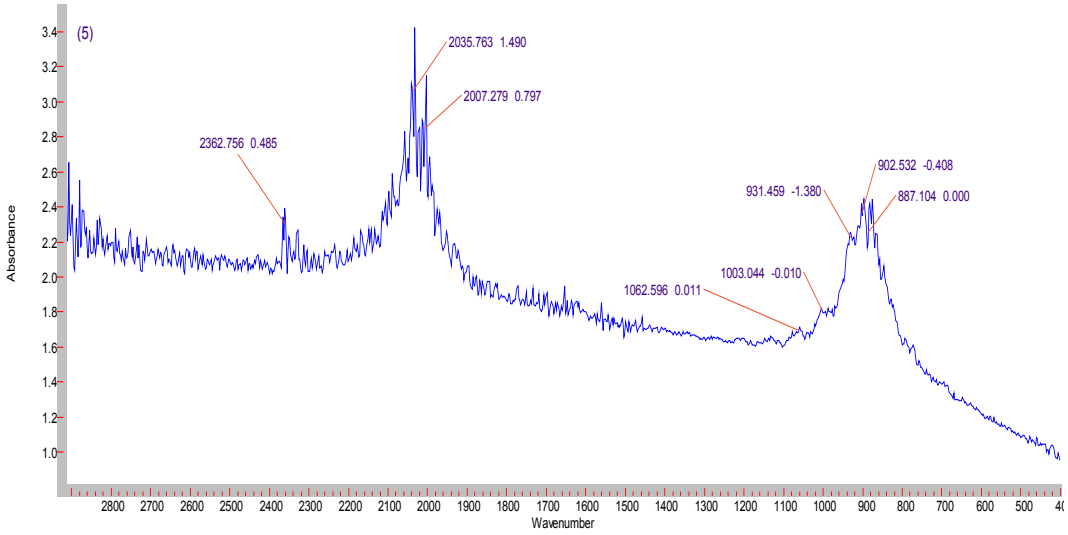

Fig. 5. Luminescent analysis of flakes graphene oxide.

Helps to obtain information about the functional groups present inside the graphene oxide using the method of IR spectroscopy. According to the Lerf-Klinowski model, graphene oxide belongs to some oxygen groups, along with alcohol and epoxy groups, to carboxyl and carbonyl groups [15]. According to the luminescence analysis of flakes graphene oxide, it consists of carbon-carbon, carboxyl, carbonyl, alcohol, aromatic esters, alcohol and epoxy groups. The peaks observed in the board bands observed in the spectrum ~2007-2363sm^{-1} – mainly characterize the carbon-carbon, carboxyl and alkane groups. The peaks at 1524, 1525, 1526 və 1558 sm^{-1} belong to the benzene rings. Peaks observed in the range of ~1003-1063sm^{-1} indicate aromatic ether and alcohol groups, and peaks observed in the ~887-932sm^{-1} indicate epoxy groups, respectively.

3. Conclusions

Based on the results of SEM and EDX analysis, the elemental composition of the mass, the thickness and length of the layers present within the mass were determined. The thicness of the flakes graphite layer and the number of layers were calculated using the Debye-Sherer expression. Based on the Raman scattering curve, flakes were found to vary depending on the thickness of the different layers of graphene oxide. The prices of D, G, 2D, D+D, $2D^{/}$, ID/IG and I2D/IG were determined accordingly. The temperature dependence was determined by preparing a thin layer of the obtained flakes graphene oxide mass. With the help of infrared spectroscopy, electromagnetic radiation in the infrared range is absorbed by the atomic groups of graphene oxide, and molecular vibrations are excited by light quanta. When a molecule is irradiated with infrared radiation, only quantum quantities of absorption arise, corresponding to the frequencies of elongated and deformation vibrations of molecules.

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