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Synthesis of Graphene Oxide Functionalized with Amio Methyl Phosphonic Acid (AMPA) and its Structural Characterization

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(Received 7 Apr. 2021; Revised 8 May 2021; Accepted 23 May 2021; Published 15 Jun. 2021)

Abstract: In this paper, graphene oxide (GO) and its functionalization with “Amio Methyl Phosphonic Acid” (AMPA) are synthesized using modified Hummers method. Crystal structure of the compounds is investigated by X-ray diffraction pattern (XRD). Fourier transform infrared spectroscopy (FTIR) clearly shows that the AMPA agent does really enters into the GO. Transmission electron microscope (TEM) images of the compounds reveal that they are in the form of nano-sheets. High resolution TEM (HRTEM) microscope is also used to observe and study the nanoscopic morphology of the structures. In addition, the samples are element-analyzed by energy dispersive X-ray spectroscopy (EDS), and X-ray photoelectron spectroscopy (XPS) to get more information. Finally, the functionalization mechanism of GO with AMPA is studied and the mechanisms of “nucleophilic displacement” as well as “condensation reaction” are suggested and experimentally confirmed. Due to the favorable properties of the synthesized material AMPA-GO, its use was suggested for water treatment and removal of heavy metals such as lead and strontium.

Keywords: Graphene oxide, Amino Methyl Phosphonic Acid, Modified Hummers Method, Structural Analysis.

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1- INTRODUCTION

Humans are familiar with carbon and its different allotropes from ancient times and it is more than thousands of years that people's life is engaged in different forms of carbons such as coal, soot and graphite. Carbon is the most important element in the organic structures and organism's bodies and is actually the building-block element in the life skeleton. However, this life-giving element can sometimes be the origin of the ecosystem pollution and also cause of killer poisons such as the green-house CO₂ gas and the CO deadly gas. The carbon element is not only one of the foremost elements in the life and living cycles on the earth, it is also the first and most well-known element in the nanotechnology. In other words, carbon nano-tubes which was discovered by Iijima in 1993, is indeed the main key into the nano-world [1]. After this discovery and subsequent introduction with features of the carbon nanotubes, the nanotechnology with its strange and interesting properties was started and has been continued and, at present, is going ahead with a very high speed. Of course, few years before the discovery of carbon nanotubes, the ball-like structures of the carbon (known as fullerene or Buckyball such as C₆₀) had been discovered which are also one form the carbon allotropes [2]. After that, the mono-layer and multi-layers of the graphite known as graphene were discovered [3]. In other words, the carbon element which has very old and ancient usage has entered to the nanotechnology and has allocated a very large portion to itself in today's modern world. Many studies were accomplished on these nanomaterials to incorporate them in different scientific and technology fields which their citation is impossible here. Even if one wants to limit himself to one of these three nano-materials such as graphene, then the volume of citations and discussion will be very huge [4].

Graphene is a mono- or multi-layer structure from graphite sheets which consists of millions of carbon elements placed near each other in a honey-comb two-dimensional lattice [5-7]. Graphene has many potential and practical applications which one can find a full address of that in the review papers [8-10]. Recently, new compounds and composites of graphene have been introduced or are being synthesized which theoretical predictions and also experimental confirmation show that they should have exclusive properties. Among these cases, one can mention the graphene oxide. GO, contrary to its name, does not contain only carbon and oxygen but it also includes hydrogen and its oxygen content is variable. The GO has many applications in nano-thickness papers, chemical and biological membranes, and sensors as well as in pollution removing from water and a lot of literature is devoted to them [11-14].

GO can be functionalized with different chemical groups or be composited with different compounds and get increased and improved properties [15].

There are different methods to synthesize GO and its functionalization which chemical methods are one of them [16]. The Hummers method among these methods is a very efficient and environment-lover method which has the ability in both synthesis of GO and its functionalization [17]. By Hummers method, different chemical agents such as epoxy, hydroxyl, carboxyl and etc. can be added to GO [18, 19]. Beside these oxygen-containing agents, other chemical groups such as aromatics, amino-acids and also aliphatics have the ability to be functionalized to GO with this method [20]. One of the most important chemical groups which its functionalization with GO is valuable and no scientific reports can be found regarding it in the literature is “Amino Methyl Phosphonic Acid” (or AMPA in brief). AMPA is an organic molecule and has a lot of applications in water, soil, herbal and agricultural products and also in ecology. It seems that, conjugation of AMPA with GO can have improved properties due to the nanotechnology.

In the present paper, the synthesis of GO and its functionalization with AMPA is considered. These compounds are prepared by improved Hummers method and are studied with different characterization and microscopic tools such as XRD, FTIR, XPS, and HRETM. The paper is organized as follows: In the first section the synthesis of the compounds is reported. In the second section, the experimental analyses are mentioned. The third section is devoted to the results and discussion. Summary and conclusion are drawn in the last section.

2- SYNTHESIS of GO and AMPA-GO

In this paper, improved Hummers method is used to synthesize GO and graphene oxide functionalized with “Amino Methyl Phosphonic Acid” (AMPA-GO). In the continuation the procedure is described.

2-1- Synthesis of GO

To prepare GO, 2 g of graphite powder (C, purchased from Merck) with 2 g sodium nitrate (NaNO_3 from Scharlo) and 50 mL of concentrated sulfuric acid (H_2SO_4 98% from Merck) are purred into a beaker containing a magnet and then is magnetic-stirred below 10 °C using an ice bath. Then, 6 g of potassium permanganate (KMnO_4 from Scharlo) is slowly added to the mixture in 2 hours while the mixture is continuously stirred under 10 °C. After two hours, the beaker is taken out form the ice bath and is transferred to another bath with 35 °C temperature and is stirred in that condition for 1 hour. Then, 100 mL of water is added to the mixture and the beaker is putted in a 98 °C bath for 15-20 minutes. Gradual color change of the solution to yellow is seen which a sign of GO production. In the continuation, 300 mL of distilled water is added to the

solution and after reaching to the medium temperature (for completeness of the oxidation) 20 mL of 30% hydrogen peroxide (H_2O_2 from Merck) is added. The mixture is ultra-sounded for 1 h. The prepared GO is washed with 5% hydrochloric acid (HCl from Merck) and also with distilled water until no SO_4^{2-} ion is recognizable with BaCl_2 . After removal of SO_4^{2-} ion, the color changes to brown. Finally, the solid product remained in the beaker is dried in an oven at 60°C . The final product is GO which will be analyzed in the following.

2-2- Synthesis of AMPA-GO

An amount of 0.2 g GO obtained from the previous section is dispersed in a 400 mL water and is stirred and ultra-sounded. Then, 0.2 g of “Amino Methyl Phosphonic Acid” ($\text{CH}_6\text{NO}_3\text{P}$ from Scharlo) is added to the mixture. In the continuation, the mixture is transferred to a cooling cycle balloon putted in an oil bath with 80°C for 40 hours and is stirred with a suitable speed until the desired reaction (i.e. attachment of AMPA to GO) is conducted (Note: the air circulation suffices for water evaporation and also there is no need to circulation of water in the cooler). The obtained deposition is separated by a centrifuge and is washed with water several times and finally is dried in an oven with 70°C temperature. The product is indeed the AMPA-GO which will be structure-analyzed in the next section.

3- STRUCTURAL AND ELEMENTAL CHARACTERIZATION

For identification of crystal structure of the synthesized materials, their XRD pattern is obtained. In Fig. 1 the XRD of GO and AMPA-GO is shown:

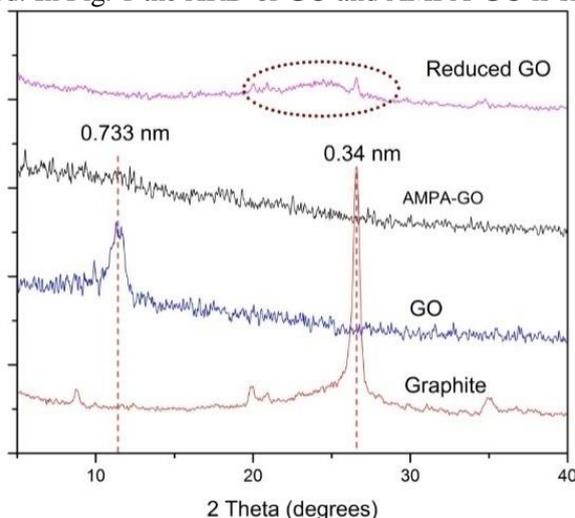


Fig. 1. XRD patterns of GO and AMPA-GO.

From the XRD pattern of the GO it is clear that a diffraction peak at $2\theta=12^\circ$ is present. This peak predicts through the following Bragg relation [21] that the distance between graphite-like plates of the GO is 0.73 nm:

$$n\lambda = 2d\sin\theta \quad (n = 1, \lambda = 1.5419 \text{ \AA}, \theta = 6^\circ) \rightarrow d$$

$$= \frac{1.5419}{2 \times \sin(6)} = 0.73 \text{ nm}$$

Of course, corresponding diffraction peak of the graphite (and also the diffraction peak of reduced graphene oxide, RGO) yields the distance to be 0.34 nm [22]. This small distance in graphite and RGO can be attributed to the Van der Waals force and π - π binding present between graphite sheets in the bulk graphite and also the reduced and less content of oxygen in RGO. In other words, presence of oxygen and hydrogen atoms in the GO structure is the main reason that the intra-sheet distance in GO is nearly twice more than that in graphite and RGO. From XRD of the ANPA-GO it is evident that the product has no crystal structure and is indeed an amorph. The finding can be related to the positioning of AMPA molecules in random places within and between GO layers.

In order to identify the chemical bonds in the products and also presence of chemical groups, the FTIR spectroscopy is used. The FTIR spectra of the GO and AMPA-GO is shown in Fig. 2:

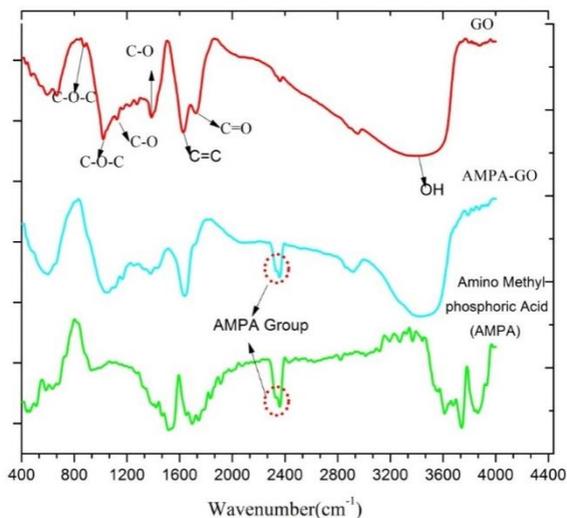


Fig. 2. FTIR spectra of GO and AMPA-GO.

From FTIR spectrum of the pure GO it is clear that all absorption peaks related to oxygen-containing bonds and chemical groups (present in the hexagonal lattice of the carbon) are present. From the FTIR spectrum of the

AMPA-GO it is evident that the finger-print absorption of AMPA at 2386 cm^{-1} is clearly seen [18, 23, 24]. Presence of this characteristic absorption is proving the fact that the AMPA agent is really attached to the GO structure. More inspection of the FTIR spectrum of GO and AMPA-GO also shows that the absorption peak at 1740 cm^{-1} (which is related to carboxyl group present in GO) is completely removed. In addition, absorption bands at 880 cm^{-1} and 1020 cm^{-1} which are attributed to epoxy group are also rather removed upon functionalization. The absorption at 1381 cm^{-1} which is related to C-O bond of the carboxyl group is also considerably eliminated in AMPA-GO.

To study the microscopic details of the AMPA-GO sample, the TEM and HRTEM electron microscopes are gotten and are shown in Fig. 3. The TEM images of AMPA-GO with smaller magnifications indicate that transparent sheets with wavy and wrinkled surface (especially at the edges which is due to out-folding of the planes) are formed. In addition, as is seen from Fig. 3, the AMPA group has been attached to the surface of the GO sheet. From HRTEM images, the crystalline order of the carbon atoms in the GO and AMPA-GO body is rather seen [22, 23].

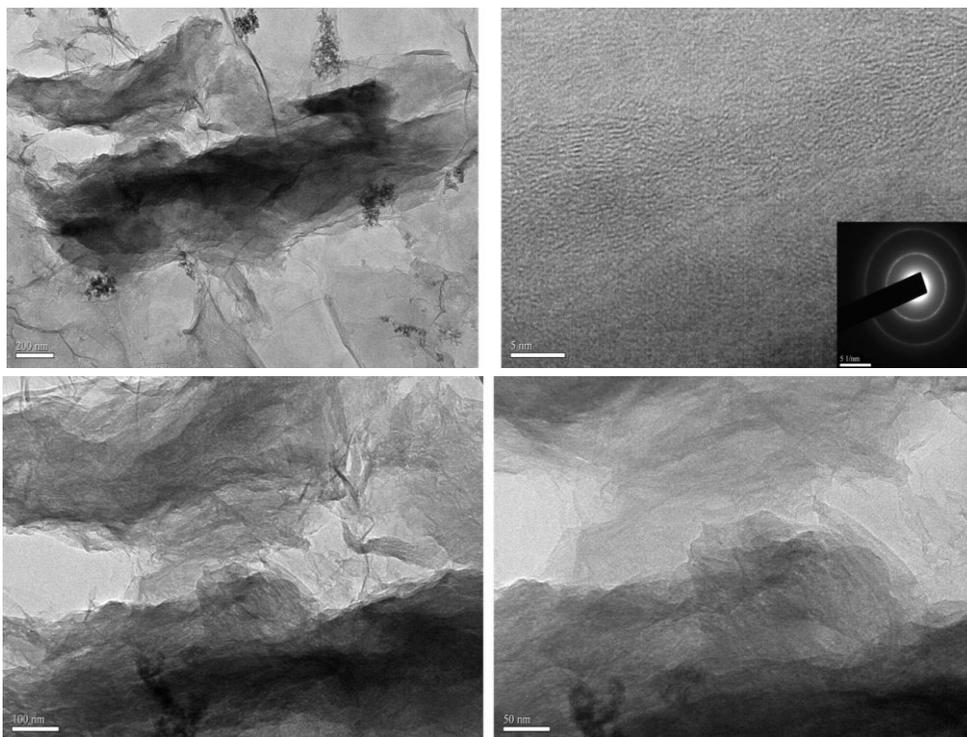


Fig. 3. TEM and HRTEM images of the AMPA-GO sample with different magnifications.

To identify the chemical elements of the GO and AMPA-GO, their EDS spectra were obtained which are shown in Fig. 4. The presence of copper in the spectrum is related to the substrate that has been used during the imaging and spectroscopy process.

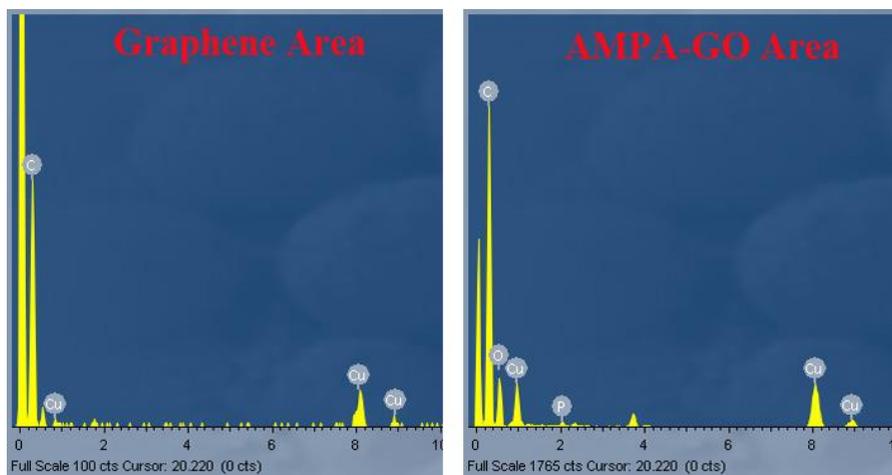


Fig. 4. EDS spectrum of the GO and AMPA-GO.

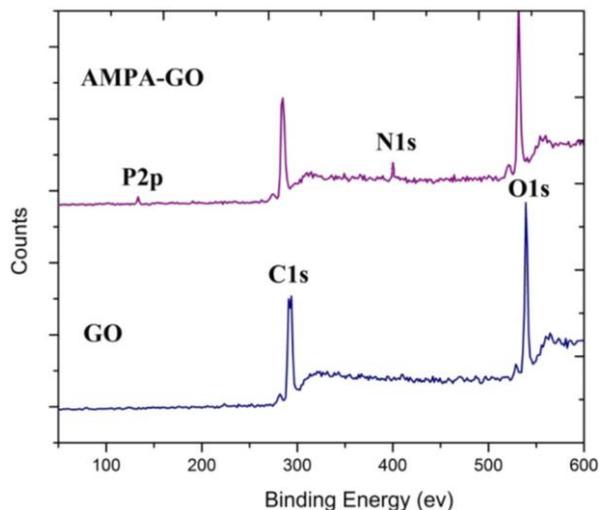


Fig. 5. XPS spectrum of the GO and AMPA-GO for determination of chemical compositions at the surfaces. The ratio of C1s peak to O1s peak can be a good criterion.

To study the chemical compositions at the GO and AMPA-GO, the elemental analysis of XPS is used. Such analysis is seen in Fig. 5.

XPS data can be adjusted and corrected by assigning of the absorption peak at 284.6 eV to C1s. It is clear from the XPS plot that after functionalization with AMPA two new absorption peaks at 133.5 eV and 405 eV appears which are related to P2p and N1s transitions [22]. Such observation shows that the AMPA is appended to the GO surface and is chemically bounded to it. For determination of elemental compositions at the GO and AMPA-GO surface, a good idea is to relate the effective area under each peak to the percent of the corresponding element. Summary of the results are reported in Table 1. Due to the Table, percentage of the nitrogen (i.e. N1s) and phosphor (P2p) are nearly the same. This is an acceptable result because the AMPA molecule has equal numbers of nitrogen and phosphor. Moreover, it is seen from the Table that there are no impurities at the surface proving that the samples have high purities.

TABLE 1
ELEMENTAL ANALYSIS OF GO AND AMPA-GO OBTAINED FROM XPS.

sample	Elemental percent			
	C	O	N	P
GO	69.3	30.7	0	0
AMPA-GO	60.7	35.4	1.6	1.7

From the Table it is also clear that there are no nitrogen and phosphor in the GO which is true because no AMPA is added to it yet. On the other hand, presence of nitrogen and phosphor in AMPA-GO is confirming the attachment of AMPA to GO after functionalization.

As the last analysis, the high resolution XPS spectrum of GO and AMPA-GO is obtained. Such spectra are shown in Fig. 6:

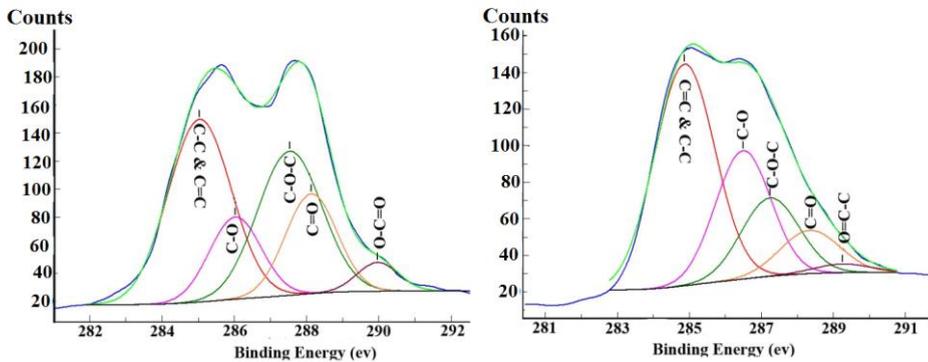


Fig. 6. High resolution XPS from GO (right) and AMPA-GO (left) for identification of atomic bonds.

The spectra clearly confirm the presence of C=C, C-OH, C-O-C, C=O and O=C-OH bonds at respectively, 285 eV, 286.2 eV, 278.1 eV, 288.2 eV and 289.1 eV in GO [25]. In other words, high resolution XPS of GO shows that all of oxygen containing chemical groups is present. However, after the functionalization with AMPA some of the groups, especially the C-O-C and O=C-O bonds are considerably reduced. This reduction can be related to the opening of epoxy group of GO upon functionalization which is due to replacement of OH of carboxyl group with AMPA.

For exact inspection of the AMPA functionalization effect on the deoxygenizing of the mentioned chemical groups, the area under the oxygen-containing peaks and also the area under the C-C and C=C bonds are obtained and their ratio to the total area under the spectrum are calculated. This simple method can give a good approximate about the percentage of the chemical bonds. Such percentages are shown in Table 2:

TABLE 2
RATIO OF OXYGEN CONTAINING GROUPS TO C-C AND C=O BONDS.

Sample	GO	AMPA-GO
C=C and C-C	37%	47%
C-O	13%	25%
C-O-C	29%	16%
C=O	16%	10%
O=C-O	5%	2%

From such a quantitative discussion one can understand that there is a reduction of about 50% in some of the oxygen-containing groups. It is also seen that the percentage of the epoxy, carbonyl and carboxyl groups are considerably reduced due to liberation of free OH⁻ ions. With this interpretation, the mechanism of functionalization of GO with AMPA agent can be investigated. It can be said that, the functionalization process is carried out in two different ways: 1) Nucleophilic substitution, and 2) condensation reaction [26]. As is seen from Fig. 7, the epoxy functional groups present at the surface of the GO are in fact the more active sites for functionalization mechanism with nucleophilic substitution channel. In this mechanism, the amine group (i.e. -NH₂) present in AMPA has a lone electron pair and, hence, can attack the epoxy group and open their ring and be bonded to the carbon atoms of the GO body. Decrease of oxygen-containing groups in GO after functionalization with AMPA (Table 2 and Fig. 7) does confirm the fact that the epoxy groups are active sites of functionalization with nucleophilic substitution mechanism. The second mechanism is based on the condensation reaction. The condensation

reaction is a phenomenon that occurs when two molecules meet each one and under suitable condition are attached to each other and form a single molecule with less entropy. Attachment of AMPA to the GO can be regarded as the condensation reaction between carboxyl group of GO and $-NH_2$ group of AMPA.

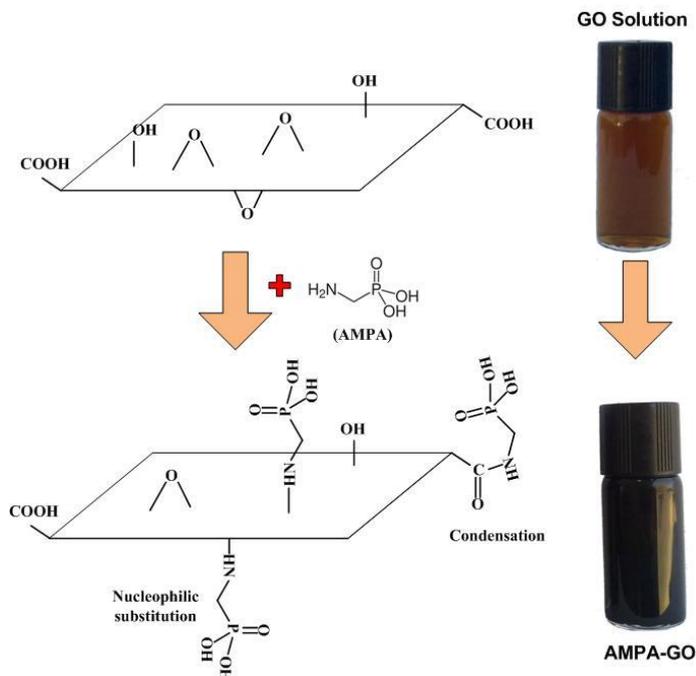


Fig. 7. Schematics of mechanisms of GO functionalization with AMPA agent.

To confirm the above-mentioned mechanisms and justify the formation of N-C bond on the GO surface, the N1s peak of XPS for the AMPA-GO is analyzed in more details. Fig. 8 reveals that the N1s peak is in fact composed of two adjacent peaks with energies 399.2 eV and 401 eV. These energies can be related to the N-C and N-H bonds, respectively [25]. This clearly shows that the amine groups (i.e. $-NH_2$) are attached to the flat surface of the GO through opening of epoxy group which is the nucleophilic substitution. The amine groups can also be grafted to the edges of GO due to condensation of carboxyl groups as is seen from Fig. 7.

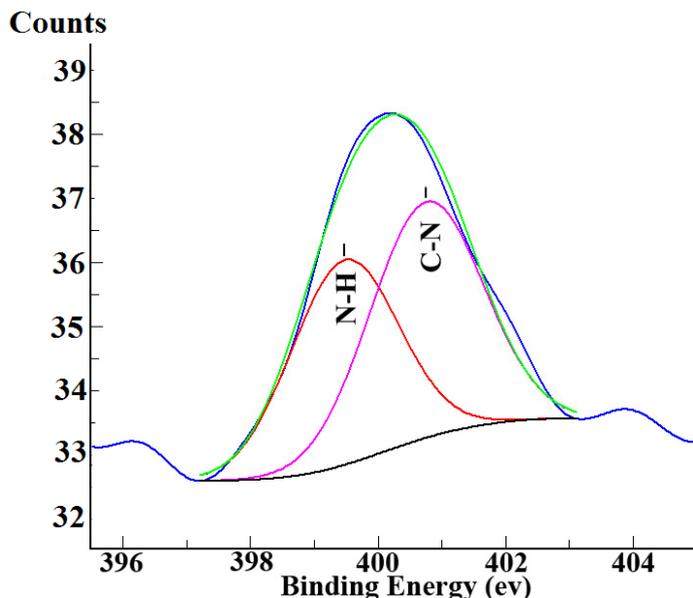


Fig. 8. More details of N1s absorption in AMPA-GO.

4- DISCUSSION

In the previous section, synthesis of graphene oxide (GO) and “Amino Methyl Phosphonic Acid” functionalized graphene oxide (AMPA-GO) was considered and its characterization through different analytical methods including XRD, FTIR, EDS, HRTEM and XPS was accomplished. Most of the obtained results were discussed in some details in the previous section, but some more discussion is considered here.

Due to XRD patterns of GO and AMPA-GO it is known that the GO has a crystal structure with intra-plane distance of 0.73 nm but the AMPA-GO has a non-crystalline structure and is amorphous. There are many reasons that can destroy the crystalline structure such as high level of defects and dislocations and also high concentrations of impurities with random distributions. It seems that attachment of AMPA agent to GO has occurred at high concentrations and high levels which is the main origin to disarrange the crystalline atomic positions of the GO.

From the FTIR spectrum of the AMPA-GO sample and its comparison with that of GO it is completely clear that the presence of AMPA and its attachment to the GO has an outstanding fingerprint absorption at 2385 cm^{-1} . On the other hand, it was seen that attachment of AMPA agent to GO leads to reduction of C=O and C-O bonds. This finding means that the AMPA agent has made a bond with carbon atoms of GO and has excluded oxygen atoms from some

bonds. Such phenomenon can be considered as another reason for destroying of the atomic crystalline order in GO and converting it to the amorphous AMPA-GO.

Elemental distribution of compounds was obtained using XPS analysis. High resolution XPS also yields the number of oxygen-containing bond in GO before and after functionalization with AMPA. The only thing that is not resolved yet is the percentage of C-O bond because, FTIR data says that the content of this bond for AMPA-GO must be less than that in GO but, due to XPS data in Table 2, it is higher for GO. This observation needs more study but it is not the aim here.

Investigation of N-C and N-H bonds in AMPA-GO shows that two different mechanisms are engaged in the functionalization process of AMPA on GO. The first mechanism is nucleophilic substitution and the second one is condensation reaction. Nucleophilic substitution leads to opening of epoxy ring at the GO surface and attachment of AMPA to it. The condensation reaction is also responsible for attachment of AMPA via carboxyl group present in the GO edge. Perhaps, the wrinkling of GO sheets at its edges and its back-folding seen in the HRTEM images can be related to the condensation reaction which, of course, needs more study.

5-SUMMARY AND CONCLUSION

In this paper, the synthesis and characterization of graphene oxide (GO) in pure form and as well as functionalized with “Amino Methyl Phosphonic Acid” (AMPA-GO) with modified Hummers method and various analytical tools was considered. The following final conclusions can be drawn:

- 1- The XRD patterns of GO and AMPA-GO showed that GO has crystal structure but AMPA-GO is amorphous. The amorphous-structure of the matter is originated to be the attachment of AMPA agent at high levels and also to random positions in GOES structure and, thus, destroy of atomic order.
- 2- HRTEM images shows that the AMPA agent is really grafted to the GO in random sites.
- 3- High resolution XPS data indicated that there are two mechanism for functionalization of AMPA on GO which includes ‘nucleophilic substitution’ and ‘condensation reaction’. The main mechanism for this attachment to GO surfaces is nucleophilic substitution, but the process at the edges is mainly conducted through condensation reaction.

As the final conclusion, it can be said that the modified Hummers method is a suitable and successful method for GO synthesis and its functionalization with AMPA. Now, the AMPA-GO is synthesized and it can be used for different

applications. The authors are using the AMPA-GO for water purification and removal of heavy metals such as lead (Pb) from it. The obtained results are very good and will be presented elsewhere.

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