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Green Synthesis and High Efficacy Method for Reduced Graphene Oxide by Zataria Multiflora Extract

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Abstract

There are many different methods for producing nanoparticles, but the use of plants due to the low cost and environmentally friendly nature of nanoparticle synthesis is very much considered. The biological synthesis of reduced graphene oxide by extracts of Zataria multiflora cultivar is reported in this work. In this study, Zataria multiflora extract was used as a reducing agent for the production of nano-graphene oxide in high yield and avoids the use of toxic and environmentally hazardous reducing agents commonly used in the reduction processes of GO. The reduced graphene oxide and optimization of reaction conditions were monitored by UV and analyzed for determination of size and morphology of reduced graphene oxide with Transmission Electron Microscopy (TEM), Raman spectra, Fourier Transform-infrared spectroscopy and X-ray diffraction devices (XRD) respectively.

Keyword: Green synthesis, Zataria multiflora, Reduced graphene oxide, Graphene oxide

1 Introduction

Graphene and graphene oxide have enormous potential for a broad category of biomedical, catalytic and optical applications, electronic, bioimaging, etc. due to their excellent characteristics and their biocompatibility (1-6). Thus, there is a growing need to develop the processes which are not harmful to the environment of the graphene synthesis that does not use toxic chemicals. Recent articles on the reduction of graphene oxide discuss the substitution of natural reducing agents for toxic hydrazine4–8and biologically reduction of graphene oxide by microorganisms and leaves, seed or peels of a plant extract have been suggested as possible eco-friendly alternatives to chemical and physical methods (7-11).

The reduction of graphene oxide under specific conditions (temperature and pressure) are costly and tedious and have potential dangers for the earth, thus it's required to have minimal effort, non-dangerous and non-lethal strategies. One of the strategies to produce reduced graphene oxide is the green reduction, which is noticeable and has expanded nowdays (12-15). Graphite oxide was first created in 1860, in view of known techniques for Hummer and Hoffman. It is a blend of carbon, hydrogen and oxygen molecules. Graphite oxide is extremely hydrophilic and comprises a layered structure of graphene oxide sheets. The most widely recognized technique for the preparation of graphite oxide is to utilize at least one-concentrated acid within the strong oxidizers. Brody utilized this strategy in 1859 (16-18). Researchers trust that graphene oxide is a standout amongst the toughest materials at any point known, because of its C-C bond strengths. Graphene oxide has specific properties, for example, nanometric size, high hardness, and mechanical quality, high electrical and warm conductivity, adaptability and attraction. Along these lines, numerous utilizations will be made, incorporating applications in nanoelectronic, sun oriented cells, vitality stockpiling gadgets, for example, batteries and supercapacitors. Graphene and graphene oxide have high conductivity and are extremely reasonable for use in circuits and electronic devices. While graphene oxide has been introduced as the primary component in all materials discussed thus far, nanocomposites of graphene oxide use these sheets as a minor filler component fixed within either a polymer or an inorganic matrix. Delivery their high oxygen content, graphene oxide layers are the carbon analogs, whose nanocomposites with polymers have been completely investigated for a broad range of applications (6, 19). In contrary, the huge surface to volume ratio of graphene oxide, in addition to its high ability to dispersion in both water and non-aqueous solvents and its broad spectrum of reactive surface-bound functional groups, discourages aggregation, make it easy for processing in solution and promoting gross interaction between fillers and polymers. A diversity of graphene oxide-based nanocomposites has been manufactured as thin films, although they are usually reduced to graphene for conductivity surveys.

The most common path toward bulk quantities of reduced graphene Oxide starts with the conversion of graphite to graphene Oxide [GO]. The first procedures for the synthesis of GO were done many years ago by Brodie (20), Staudenmeier (21) and Hummers et al. (16) and persist with only minor changes. The degree of graphite oxidation, as quantified by the carbon to oxygen atomic ratio, is dependent on the synthetic

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method as well as the reaction procedure (22-24). The Staudenmaier method 20 usually produces the most oxidized GO, although the oxidation of graphite to GO breaks up the sp2-hybridized unity of the stacked graphene layers, generating errors that manifest as clear notches in the stack (18, 25) and become more the distance between adjacent sheets (23, 24). This increased spacing is different depending on the amount of water interacted within the Stacked-sheet structure (25, 26) and reduces sheets interaction, so helping the delamination of GO to individual graphene oxide sheets by exposure to low-power sonication through the water. At a little basic pH, negatively charged, hydrophilic oxygenated functional groups on the graphene oxide surface can settle down dispersions of these layers in water-based media (18, 27). Within shortly of being available in bulk quantities, graphene oxide, reduced graphene oxide, and graphene have become highly practical, inexpensive building blocks for the creation of several advanced carbonaceous materials, with applications to be restricted only by the imagination. For example, it is not difficult to imagine the internalization of different additives into thin layer composites of graphene and graphene oxide to create excellent material compositions.

Thus, polymers have already been put together to graphene papers and films, either in nanoparticle form (9, 28, 29), or by in situ electro polymerization (1, 18, 22) for stabilizing mechanically and to facilitate processing. The development of ways to completely exfoliate graphite powder, in a wide range of solvents (30, 31), and the without the presence of reducing or stabilizing agents, will be essential to the generation of high-quality graphene materials. Improved synthesis and processing of graphene oxide and graphene being these highly versatile materials can be achieved by more researchers and engineers, so ensuring the quick development of many new materials with amazing properties. For instance, monodisperse colloidal solutions of single graphene sheets and few-layer graphene piles have recently been separated from bulk graphene nanosheet dispersions via density gradient ultracentrifugation (1, 32) and used to make thin films that have lower sheet resistance than those made from bulk graphene dispersions. The third frontier for later development hides in the production of huge surface graphene thin films for use in electronic and energy usage. However, to produce the highly conductive and translucent monolayers necessary for these purposes, new methods for large-area thin-film preparation must be mixed with advances in the synthesis of large quantity, spitless graphene sheets, as discussed in the next paragraph. As graphene can also be a template for catalytic nanoparticles (33, 35), having high procedures and turnover, the production of catalytic thin films would afford large surface area heterogeneous catalysts for utilization in fuel units. In another application, graphene drop cast from a dispersion has recently been utilized as a thin support film for TEM images to prepare unmatched atomic resolution (36). With such decent applications in one's head, the development of new graphene oxide and graphene-based materials will lead to many future developments in science and technology (1, 37). While graphene particles can be detached from reduced slightly dissolve of graphene oxide, thermal exfoliation prepares an alternative way for its preparation directly from GO and circumvents the usage solvents (1, 18, 25).

This method involves the speedily heating of GO, which gets complete delaminated via the evolution of carbon dioxide as surface hydroxyl and epoxy groups decompose (1, 23). With its fully joined together, firm 2D structure, graphene has both electrical and thermal ability to conduct (38, 39) and mechanical possessions (40) that are superior to those of other carbon allotropes. So, graphene has attracted significant interest in the world as the most successful carbon allotrope for the growth of next-generation carbon-based materials. On the other hand, the number of papers that explain graphene and its related materials (41, 42) has eminently exponentially from 2004 isolation of single graphene by use of mechanical exfoliation3,42. Graphene can be made from different graphite derivatives (43), bulk number of completely exfoliated graphene-like sheets are commonly driven by the reduction of graphene oxide distribution or powders. Graphene has attracted significant interest in the scientific community has the most promising carbon allotrope for the development of next-generation carbon-based materials. Indeed, the number of peer-reviewed articles that discuss graphene and its associated materials (41, 42).

Zataria multiflora as a reducing agentZataria multiflora is a kind of thyme. Zataria multiflora is the main type of local Thymus in Iran. It is situated in Shiraz (Mount Sivand, mountains neglecting the Maharlou Lake, between the backwoods, among Jahrom and Mansurabad, near Tarom Darab), Zataria multiflora is contained around 40% thymol, the primary successful fixing with a specific end goal to restore coriander and thyme because of the extravagance of this substance is the best decision for biological recovery (43-45). There are mixes, for example, bicyclic acid, propionic acid, acrylic acid, unpredictable lalalum, and germabol. Mixes of the plant incorporate lanthanide acetic acid derivation, coumarin, flavonoids, and phytosterols. Studies have demonstrated that the blend of green graphene oxide reduced by the utilization of Zataria multiflorais remove is less known in the present examination. The amalgamation of green-graphene oxide from Zataria multiflora species was considered (43).

2 Materials and methods

2.1 Preparation of Graphene oxide powder

Graphene oxide prepared by oxidation of regular graphite powder by Modified Hummers techniqu (1, 16, 23). In the ordinary strategy for preparation 5 gr graphite and 5 gr of $NaNO_3$ (sodium nitrate) were added to 200 ml of H_2SO_4 (in Ice and water) and bit by bit 30 gr of $KMnO_4$ was added to this blend and amid this procedure, temperature kept on 30°C (since the response is exothermic). The reaction mixture at that point mixed for 18 hours at room temperature with the goal that thick glue is shaped. The glue at that point filled 400 ml of distilled water and following 20 minutes another 1 liter of distilled water added to the blend. 30 ml of 30% H_2O_2 arrangement added to the reaction mixture drop insightful to reduce the abundance $KMnO_4$. The arrangement transformed into light dark-colored in the wake of including H_2O_2. The solution was expelled in a decanter and centrifuged and after that washed out with distilled water to achieve the pH of 7 and afterward dried under vacuum for 24 hours to frame GO powder (15-18).

2.2 Preparation of Zataria multiflora leaf extracts

Leaves of the Zataria multiflora plant gathered from different regions of Shiraz, Fars, Iran, washed independently with water at that point dried for 7 days at room temperature45. The plant leaf separates set up by the expansion of 10 g of altogether washed and finely grounded leaves (utilizing a residential blender) to 200 ml of deionized (DI) water in a 500 ml Erlenmeyer flask. The mixture boiled for 60 min before sifted under encompassing conditions. The concentrates put away at 4 °C and utilized inside seven days (37-46).

2..3 Reduction of graphene oxide

In an ordinary methodology for the reduction of graphene oxide, 50 mL of Zataria multiflora leaf extract added to 200 mL of a homogeneous scattering of graphene oxide (1 mg/mL). In the wake of energetically shaking and sonicated for 20 minutes, the mixture warmed in an oil shower at 98 °C under reflux for 24 hrs. The subsequent dark scattering centrifuged at 15,000 rpm for 60 min. The pellet then washed three times with DI water to evacuate the undesirable materials. Finally, the lessened graphene oxide (RGO) dried at 70 °C for 24 hrs (46, 47).

3 Results and Discussion

Optimization of reaction conditions for the reduction of graphene oxide an unmistakable change in the shade of the suspension on reduction can be an undeniable sign of the reaction (48-50). The chemical reduction of the yellow-dark colored colloidal suspension more often than not results in a dark accelerate, which likely outcomes from an expansion in the hydrophobicity of the graphene oxide sheets caused by a decline in polar functionality on the surface of the sheets (49-51). Consequently, the reduction of graphene oxide on introduction to Zataria multiflora extricates checked by an adjustment in shading and an expansion in absorbance at 600 nm (optimized after scan) and furthermore it tends to be observed by optical thickness estimation at 600 nm (4, 9, 53, 60). Serial dilution checked the straight connection between absorbance at 600 nm (52) and the concentration of graphene suspension. We examined the impact of Zataria multiflora concentrates of various fixations on the reduction of graphene oxide. Figure 1 demonstrates the adjustment in absorbance at 600 nm with time, for the response between various concentrations of Zataria multiflora extracts (5, 10, 15, 20, 25 and 30 % v/v) and 0.5 mg/mL of the graphene oxide suspension at 96 °C for 30 hrs.

The level of reduction seen to increment with a concentration of the Zataria multiflora removes from 5% to 20%. No further increment in absorbance discovered when the concentration of the Zataria multiflora extracts expanded to 25%. Thus, 20% of Zataria multiflora extracts utilized for future tests. Figure 2 demonstrates the adjustment in absorbance at 600 nm for the reaction between 20% Zataria multiflora extracts and 0.5 mg/mL of graphene oxide at three distinct temperatures (25, 30, 45, 60, 75 and 96 °C). As the temperature expanded from 25 to 96 °C, the level of reduction likewise expanded of course (53). Both the reaction rate and values of final absorbance were much higher at 96 ºC than those at 25, 30, 45, 60 were and 75 °C.

3.1 Infrared spectra

Graphite powder utilized to make graphene oxide, which acquired from Merck Inc. The aftereffects of the graphite infrared spectroscopy test are watched, the graphite contains no carbonyl and epoxide group, and the adsorption in the locale of around 3422 cm-1 is identified with the hydroxyl group of the atom water consumed by graphite (3, 13, 17) (Figure 3).

Graphene oxide arranged by changed Hummer strategy at that point refined. The FT-IR spectroscopy of graphene oxide demonstrated an absorption frequency of 1720 cm-1relative to the tensile vibrations of the carbonyl group (C = O) and additionally strong absorption in the 3421 cm-1 of the hydroxyl group, (OH) 1384 cm^{-1} group of the flexural vibrations of the hydroxyl group (OH) and absorption in the 1031 cm-1 locale of the epoxide tensile frequencies (Figure 4). Likewise, the pinnacle contained in 1621 cm^{-1} is identified with the functional groups (C = C) staying on graphene plates, which have not experienced any progressions amid the oxidation procedure (3, 11, 13, 17). . At that point, the concentrate of the leaf extract of Zataria multiflora was added to arranged GO, refluxed then sonicated, and at the last stage, the suspension was centrifuged and the RGO dark shaded washed with water twice and went away in the stove.

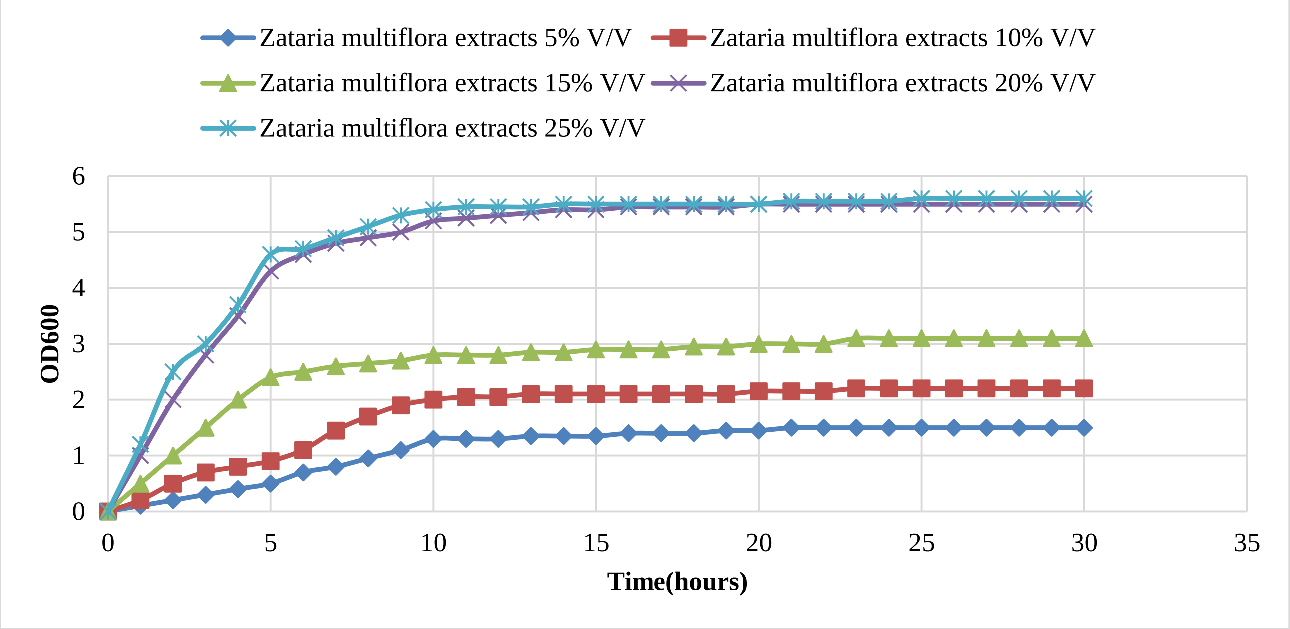

Figure 1: Effect of the concentration of Zataria multiflora extracts on the reduction of graphene oxide (0.5 mg/mL) at 96 °C

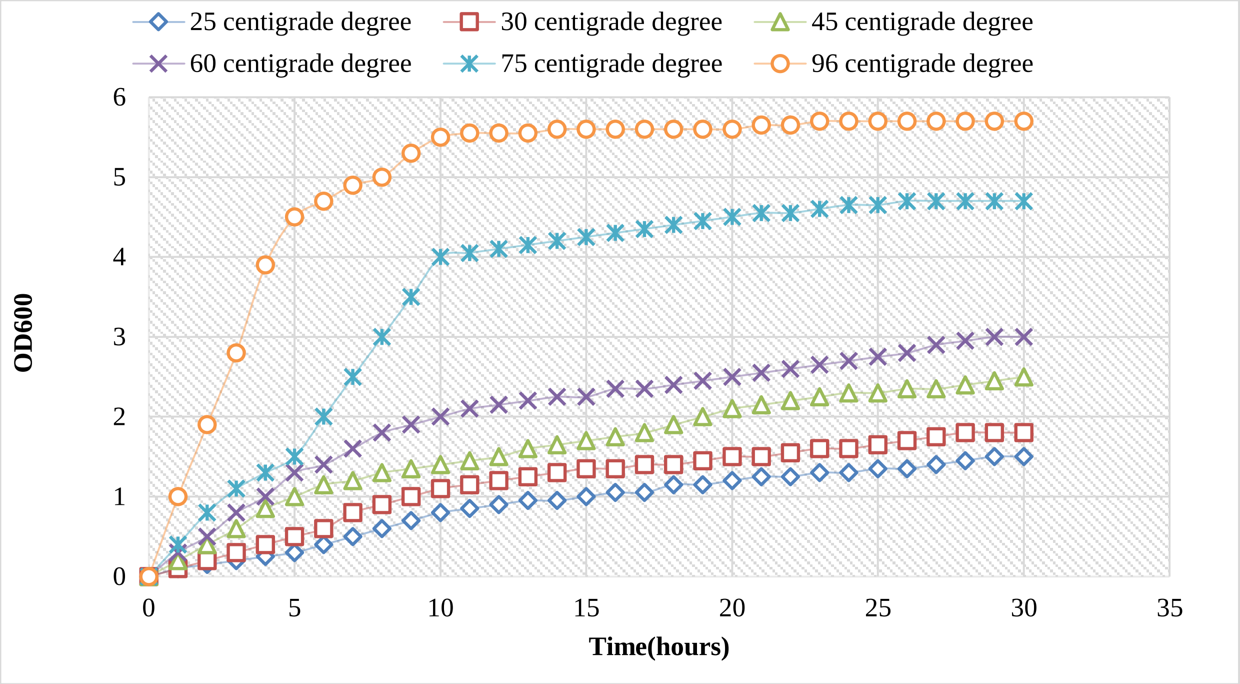

Figure 2: Effect of reaction temperature on the reduction of graphene oxide (0.5 mg/mL) using 20 % v/v Zataria multiflora extracts

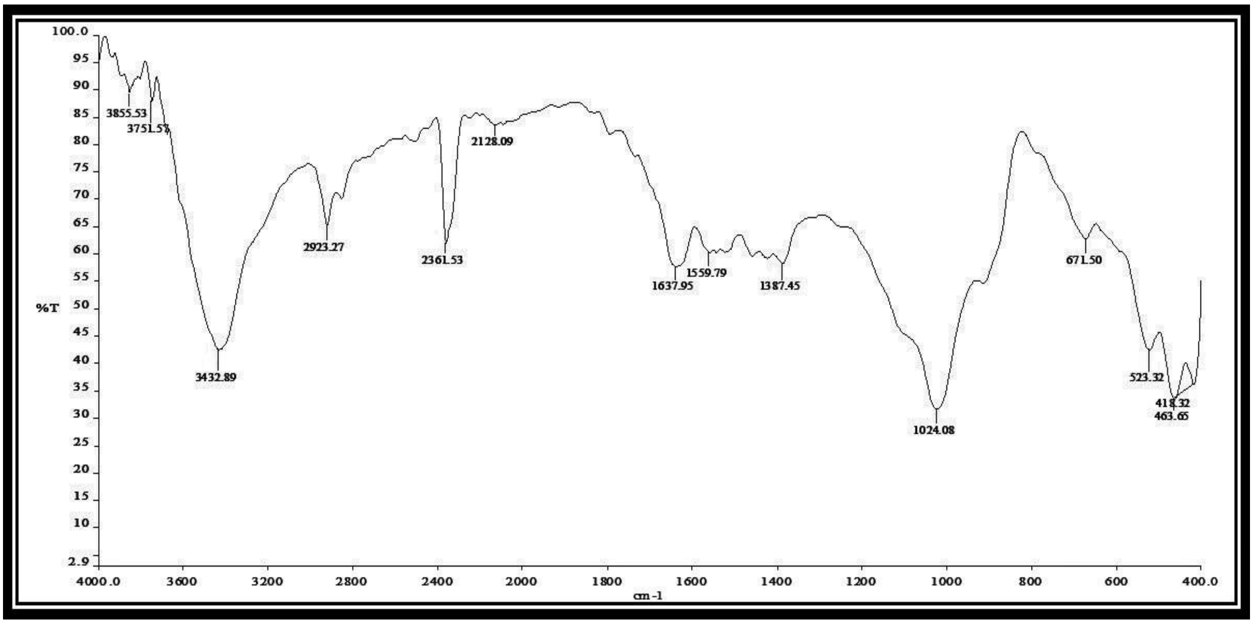

Figure 3: Infrared spectra of graphite

The examination of the FT-IR spectra in GO and RGO demonstrates that in GO there is a tensile vibration of C = O in cm-1 of 1720 cm^{-1}, yet in RGO the ductile vibration of C = O isn't so serious and moved to 1797 cm^{-1}, and also, the power of the vibration retention of OH in 3421 cm^{-1} and 1384 cm^{-1} which at RGO diminished fundamentally after reduction and achieved the estimations of 3350 cm^{-1} and 1100 cm^{-1} (Figure 5), so it tends to be presumed that amid the recovery procedure (planning of RGO), the current oxygen species on the surface of graphene sheets, the concentrate of Zataria multiflora plant has been decreased and graphene oxide has been reduced to graphene (3, 11–13, 21, 54).

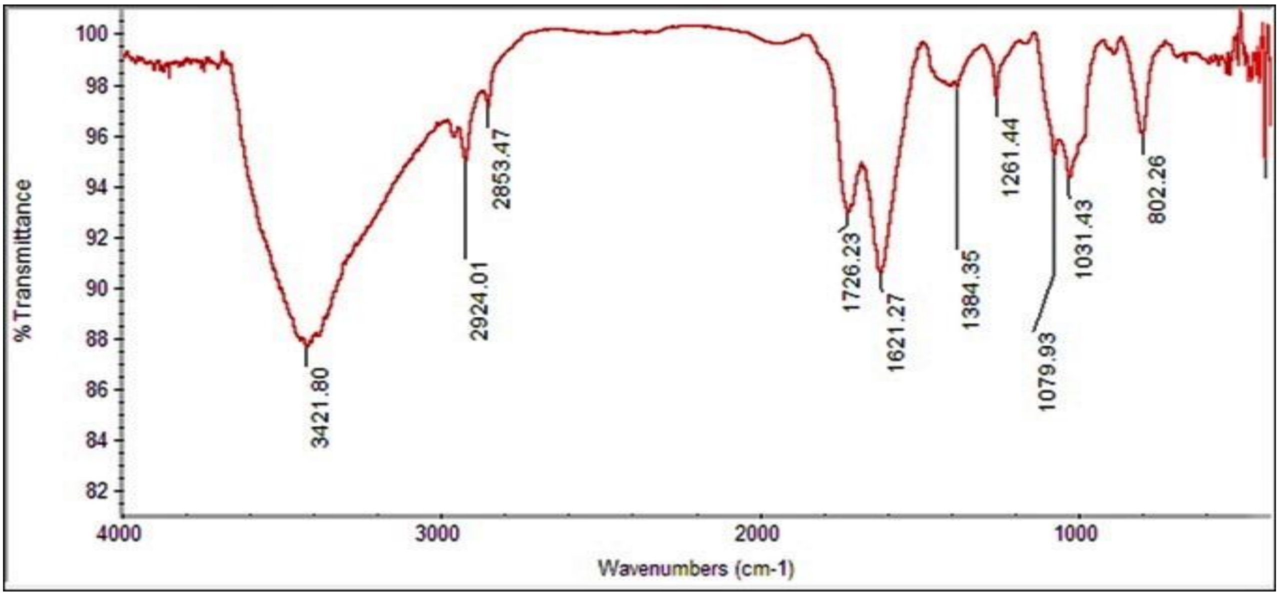

Figure 4: Infrared spectra of graphene oxide

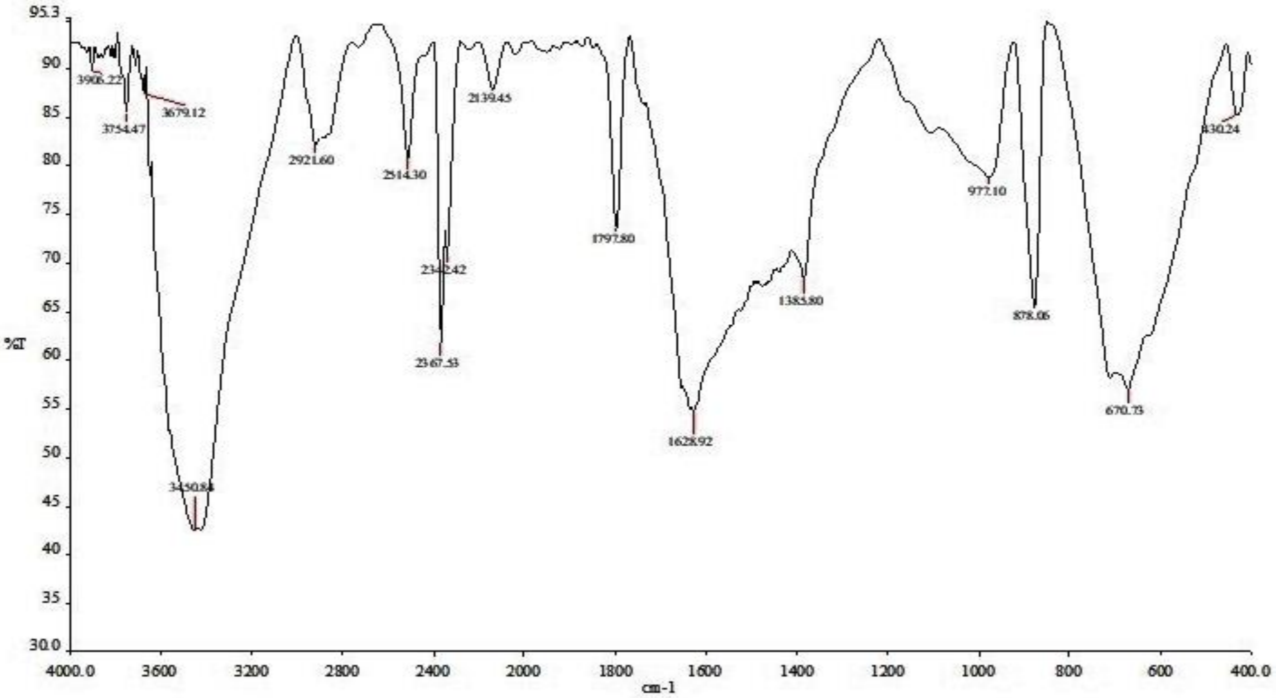

Figure 5: Infra-red spectra of reduced graphene oxide

3.2 XRD analysis

The X-ray beam diffraction test utilized to check the structure of graphene oxide, graphene, and oxidized graphene. As appeared in figure 7, X-rays beam diffraction (XRD) for graphene oxide demonstrates a strong peak at 2θ = 10.5, which shows the nearness of oxygen-containing functional groups in void spaces between graphene oxide plates. As found in Figure 6, the graphene X-ray beam diffraction example of this peak has been exchanged to the range 2θ = 13 and speaks to the graphene cover after the way toward recovering and emptying water particles and oxygen bunches from the spaces between the layers of graphene oxide (11, 12, 54).

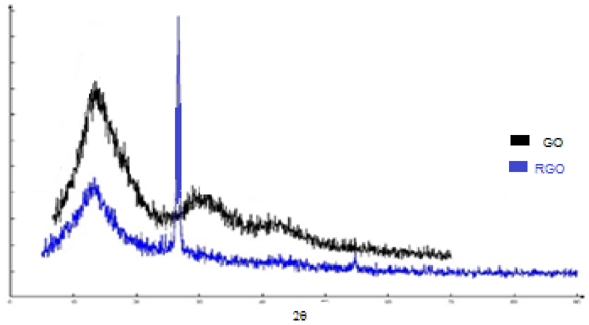

Figure 6: XRD spectra of reduced graphene oxide and graphene oxide

3.3 Raman study

Raman scattering is a valuable device to portray graphite and graphene materials as this dissipating emphatically relies upon the electronic structure. Raman range of GO was observed to be altogether changed after the reduction (Figures 7 and 8). In the spectra of GO and RGO, two crucial vibration bands were seen in the scope of 1250–1750 cm^{-1}. The G vibration mode, inferable from the first-order scattering of E2 g phonons by sp2 carbon of GO and RGO were discovered 1592 and 1587 cm-1 separately, while the D vibration band acquired from a breathing method of j-point photons of A1g symmetry of GO and RGO showed up at 1350 and 1345 cm-1 separately. .After the reduction of GO, the force proportion of the D band to the G band (ID/IG) was expanded essentially. As D band emerges due to the sp2 carbon group, the higher power of D band proposed the nearness of more detached graphene space in RGO in contrast with GO and expulsion of oxygen moieties from the former. Notably, the two-phonon (2D) Raman dissipating of graphene-based materials is an important band to separate the monolayer graphene from a twofold layer/multi-layer graphene as it is profoundly discerning to stacking of graphene layers. For the most part, a Lorentzian peak for the 2D band of the multi-layer graphene sheets seen at 2700 and 2900 cm-1 (11, 12, 54). This demonstrates RGO has a multilayer structure, after the reduction of GO, the 2D band is extraordinary, which proposed about stacking of graphene layers. As GO has diverse kinds of functional groups that may avoid stacking of graphene layers yet after reduction because of an abatement of such functional groups a couple of graphene layers are stacking and framed multilayer RGO (54).

3.4 Transmission Electron Microscopy (TEM) analysis

Figure 9 shows TEM images of the RGO sheet reduced with Zataria multiflora leaf extract. The appearance of transparent and silky sheets of RGO in TEM images verifies its stability under a high-energy electron bar. The high-resolution TEM images are utilized to get to the number of layers in numerous areas. The edges of the suspended graphene films tend to crease back, permitting the cross-sectional perspective of the films. The collapse of a couple of layers at the edges of the films shows up as a couple of dim lines, individually. The arrangement of a few layers RGO is unmistakably obvious. It has likewise been discovered that the forces of all the diffraction spots are not equivalent and sharp enough. Additionally, the diffraction spots are related to some uncertain spots. Every one of these perceptions additionally supporting the development of a few layers RGO (3, 13, 17, 21, 54).

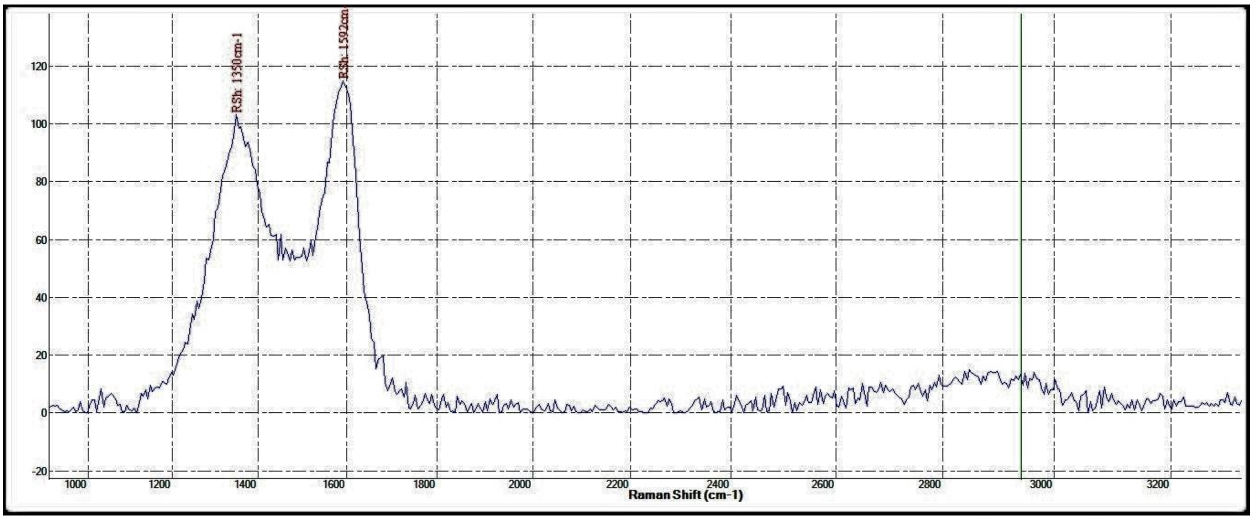

Figure 7: Raman spectra of GO

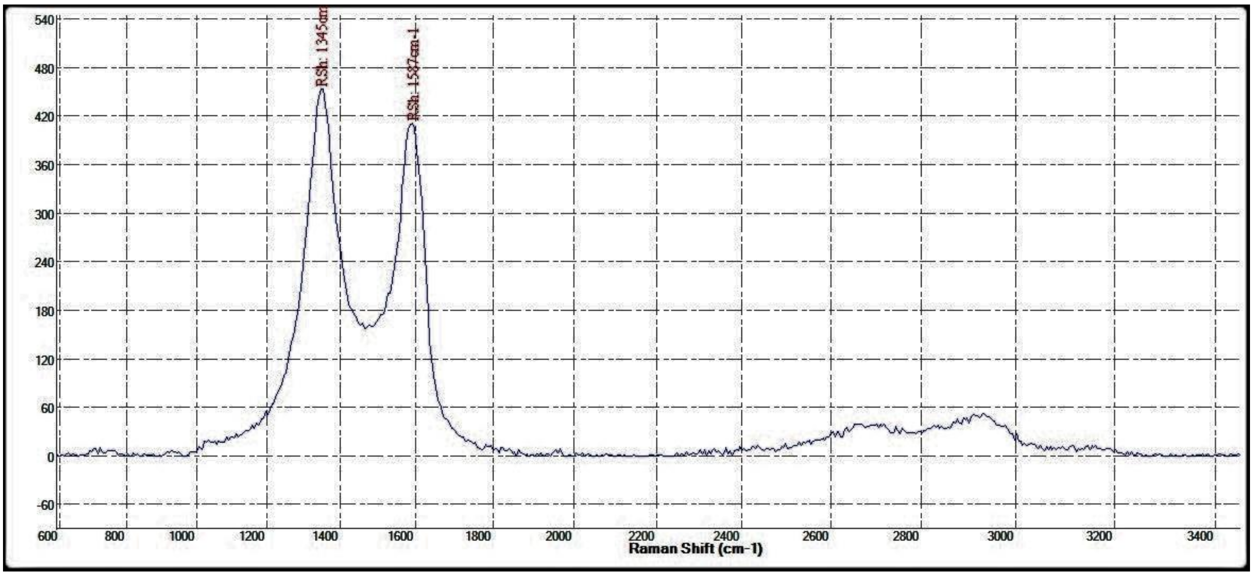

Figure 8: Raman spectra of RGO

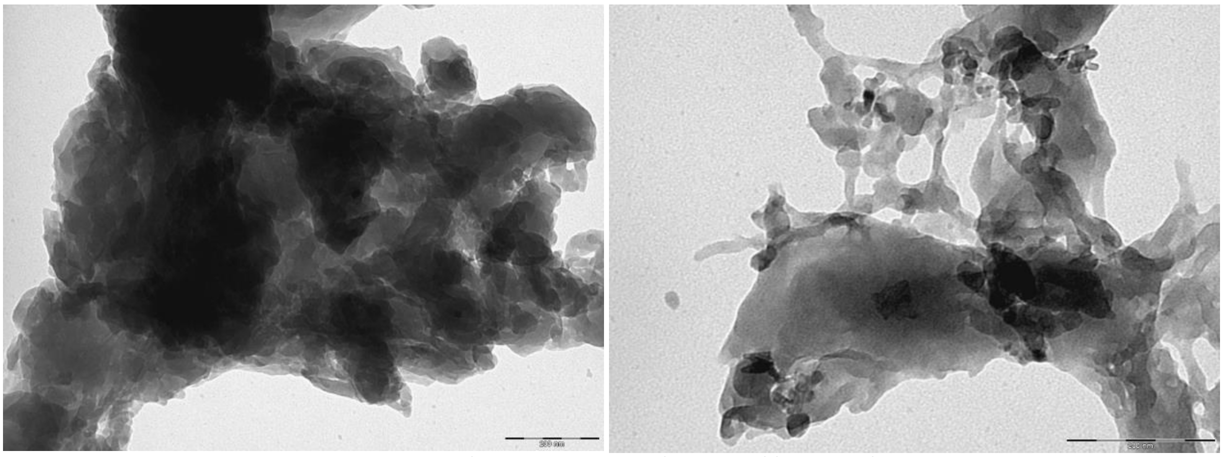

Figure 9: TEM images at 200 nm magnifications, showing the formation of few layer of reduced graphene oxide

4 Conclusion

We demonstrated that the studied phyto extracts have tremendous potential to be used as reducing agents for the reduction of GO with an environmentally benign synthetic protocol. Graphene oxide was prepared from graphite powder using a modified Hummers' method followed by ultra-sonication. The optimized reaction condition for the reduction of graphene oxide was determined to be a reaction time of 20 hours and a temperature of 96 C using 20% v/v of Zataria multiflora extracts. We showed that the considered phyto extracts can possibly be utilized as diminishing operators for the reduction of GO with an ecological amiable engineered convention. Graphene oxide was set up from graphite powder utilizing an altered Hummers' strategy trailed by ultra-sonication. The upgraded response conditions for the reduction of graphene oxide was resolved to be a reaction time of 20 hours and a temperature of 96 °C utilizing 20% v/v of Zataria multiflora extracts. The most imperative favorable circumstances of the phytochemicals are their wealth in nature, cost-viability, and simple item segregation after reduction as they extricated from non-palatable or squander plant items. The estimations of particular capacitance, high electrical conductivity, and high carbon to oxygen proportion of the phyto extract RGO are adequate. In this way, this green strategy can be utilized for vast scale generation of RGO. An eco-accommodating and naturally considerate reduction framework by utilizing Zataria multiflora removes as a biocatalyst for the reduction of graphene oxide is portrayed. The reduction was done in a watery medium at an alternate temperature. TEM uncovers the development of few-layer of reduced graphene oxide. FTIR and X-ray examination give proof to the end of labile oxygen usefulness from the surface of GO. De-oxygenation and the development of deformities in the RGOs have been affirmed by Raman spectroscopy. The primary points of interest of this system over the conventional synthetic reduction are the cost-adequacy, ecologically agreeable methodology, and basic item confinement process. This ecologically amicable reduction of graphene oxide can possibly be utilized in different fields, for example, biomedical applications.

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Ethical issue

Authors are aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

Authors' contribution

All authors of this study have a complete contribution for data collection, data analyses and manuscript writing.

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