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The effect of gamma-irradiation on graphene oxide in a monoglyceride/ethanol solution

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Abstract: Gamma-irradiation effects on graphene oxide (GO) in a monoglyceride/ethanol (MG/EtOH) solution was investigated. GO was dispersed in MG/EtOH solution (GOM) with the GO: MG ratio of 1:10 (w/w). The prepared GOM was irradiated by γ-ray under nitrogen atmosphere in a range of absorbed dose from 0 to 50 kGy. The characteristics and morphology of reduced GOM were analyzed by Ultraviolet-visible (UV-vis) spectroscopy, Fourier transform infrared (FT-IR) spectroscopy, X-ray diffraction (XRD) and Transmission Electron Microscopy (TEM). The results confirmed that the structure of reduced graphene oxide in monoglyceride solution was changed and exfoliated completely after γ-ray irradiation at absorbed dose 50 kGy compared with non-irradiation sample.

Keywords: *reduction, monoglyceride, graphene oxide, γ-ray.*

I. INTRODUCTION

Graphene is a two-dimensional crystalline material, discovered by two scientists Novoselov and Geim in 2004 [1]. The structure of graphene is a flat plate with a thickness of one atom of carbon atoms connected by the Sp^2 bonds forming a hexagonal lattice, due to its interesting properties including excellent thermal conductivity, large theoretical specific surface area, high electron mobility, good optical transmittance, very thin, and harder than diamonds [1,2]. Until now, graphene has been studied for applications in solar cells, electronic or optical devices, sensors, energy storage and nanocomposite [3-6]. In recent years, in addition to the chemical methods used to produce graphene from graphite through an intermediary graphene oxide [7], the physical methods have also been used to reduce graphene oxide to graphene such as thermal reaction, ultrasonic irradiation, gamma ray and/or electron beam irradiation because of their several advantages, including not using reducing agents, the product can be produced a large amount, and being environmentally-friendly one [8-15]. Therefore, γ-ray and/or electron beam irradiation–induced reduction of GO in an ethanol/water and ethylenediamine solution has been reported [11,12].

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GO is synthesized from the oxidation of graphite so the GO structure contains polarizing function groups (hydroxyl, carboxyl, epoxide) [16].

In this paper, the effects of γ-ray irradiation on graphene oxide in a monoglyceride/ethanol solution (GOM) were studied. A monoglyceride (MG) is a fatty ester. Its chemical structure includes two functional groups of hydroxyl and alkyl. So, MG has self-asssembly ability when it contacted with the polarized surface. When GO modified with monoglyceride, monoglyceride could be self - assembly well between the cavity of GO structure and expanded the cavities or exfoliated single-layer graphite oxide by the hydrogen bond formation with polarizing functional groups on the surface of GO. After creating monolayer modified GO, γ-ray irradiation – induced reduction of method is used to restore the aromatic skeletons (Sp^2 bonds) of mono-layer graphite and/or the preeminent properties of the pristine Graphite.

II. EXPERIMENTAL

Chemicals and materials

Graphite flake (purity 99.9%) was purchased from SEC Carbon, Japan. Monoglyceride with M_w = 356.54 g/M (MG) was supplied by Kamdhenu Foods Ltd., Delhi, India. Ethanol (99.5 wt.%) (EtOH) was provided by Chemical and scientific technology JSC, Hochiminh City Branch, Vietnam. Sulfuric acid (H_2SO_4), hydrogen peroxide (H_2O_2), potassium permanganate ($KMnO_4$), sodium nitrate ($NaNO_3$), hydrochloric acid (HCl), acetone were obtained from commercial sources. All chemicals were of reagent grade and used without further purification. Distilled water was used in all experiments. A commercial alpha-cellulose membrane with 8-12 μm pore-size was used for filtration.

Synthesis of Graphite oxide (GO)

The Hummers method was used to synthesize GO from a graphite powder [16]. Accordingly, 10 g graphite, 5 g $NaNO_3$, and 240 ml H_2SO_4 (98 wt.%) were placed in 2,500 ml four-neck flask. The mixture was stirred in an ice bath at 0°C. Then 38 g $KMnO_4$ was added slowly. After 30 min, the temperature of the system was raised to 35 °C and the mixture was continuously stirred in 2 hours for graphite oxidation. Next step, the suspension was diluted slowly by 460 ml of distilled water. The system was raised to 98 °C and stirred for 15 min. Then, the suspension continues to be diluted with 1,400 ml distilled water and to lower temperature of reaction system to 50 °C. To remove residual $KMnO_4$ and other impurities, 500 ml H_2O_2 (30 wt.%) was added to the mixture. After completion of the reaction, the suspension became bright yellow in color. The prepared suspension was centrifuged, washed with HCl (5 wt. %) solution to remove all residual ions of SO_4^{-2}. After that, hydrochloride acid and other impurities in GO suspension were removed by acetone solvent. Finally, the repaired suspension of GO was dried at 60 °C in vacuum and ground into fine powder.

Preparation of GOM and iGOM

GO suspension in a MG/EtOH blend solution (GOM) was prepared by adding an amount of 0.01 g GO, 0.1 g MG into 10 ml ethanol/distilled water solution (25 v/v %) and dispersing under ultrasonic for 1 hour. After deoxygenated by nitrogen bubbling for 10 min, the prepared suspension is sealed and irradiated at a room temperature by γ - ray. The total absorbed doses were ranged from 0 to 50 kGy at a dose rate of 1.0 kGy/h. Afterward, irradiated GOM (iGOM) was obtained by filtration and washed with distilled water several times and dried an oven at 70 °C.

Characterization methods

UV-vis absorption spectra were recorded on a UV-vis spectrophotometer (JASCO V630, Japan) within the wavelength region of 200-800 nm. Distilled water was used as a solvent.

FT-IR spectra were colleted by a FTIR spectrophotometer (8400S, Shimadzu, Japan) using KBr pellets on the transmission module at 4 cm^{-1} resolution and for 20 scans.

The X-ray diffraction (XRD) was carried out using X Pert' Pro, Panalytical model at a scanning rate of 0.04^o/s in a wide angle range (2θ = 0-60^o) with monochromatized Cu K_α radiation (λ= 1.5406Å).

The Transmission electron microscopy (TEM) measurements of all the samples were carried out in the high resolution scanning electron micro-scope JEM 1010, JEOL in a transmission mode

III. RESULTS AND DISCUSSION

Gamma irradiation of GOM

Fig. 1 shows the images of the GO dispersion in MG/EtOH mixture solution and the irradiated dispersion at 50 kGy with a dose rate of 1.0 kGy/h. Evidently, the brown color of initial GO dispersion changed in black after gamma irradiation.

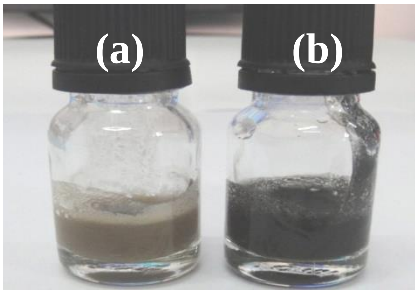

Fig.1. Photographs of non-irradiated GOM dispersion (a), and GOM dispersion irradiated at 50 kGy (b).

UV-Vis spectra

The γ-ray irradiation–induced reduction of GO in MG/EtOH mixture solution was studied by UV-vis as presented in Fig. 2, the UV-Vis spectra of iGOM-23.5kGy was appeared two absorption peak at 242 and 264 nm, and the absorption peak of iGOM-50 kGy was shifted toward a longer wavelength at 264 nm in comparison to that the control GOM (0 kGy) at 240 nm ($\pi\rightarrow\pi^*$ transition of aromatic C–C bonds) and at 300 nm ($n\rightarrow\pi^*$ transition of C=O bonds) [13,17,18]. The overall results were demonstrated that GO was reduced in MG/EtOH solution by γ-ray and GO was reduced completely at an absorbed dose of 50 kGy.

FT-IR analysis

FT-IR spectroscopy was used to characterize the functional groups attached to MG, non-irradiated GOM and irradiated GOM as shown in Fig. 3, the oxygen containing functionalities at 3315 (hydroxyl, -OH), 1731 (carboxyl, C=O), 1392 and 1049 cm^{-1} (epoxide, C–O) were greatly reduced with an increase in the absorbed dose. In comparison to the FT-IR spectra of MG and GOM, the FT-IR spectrum of iGOM-50 kGy was appeared a new peak at 1575 cm^{-1} attributed to the stretching vibration bands C=C. It was suggested that the restoration of aromatic skeletons (Sp^2 bonds) [12,15] on the mono-layer graphite was occurred in the sample of iGOM-50 kGy, which correspond to the results of the UV-Vis spectra analysis.

XRD study

The XRD patterns for graphite, GO, non-irradiated and irradiated GOM samples are described in Fig. 4. In case of pristine graphite, a sharp diffraction peak was observed at 2θ = 26.49^o (d-spacing = 3.36 Å) corresponding to the (002) plane of graphite. GO has damaged crystalline structure with a broad diffraction peak was observed at 2θ = 10.87^o (d-spacing = 8.13 Å) indicating introduction of hydrophilic functional groups (-OH, -COOH, epoxide) to carbon layers.

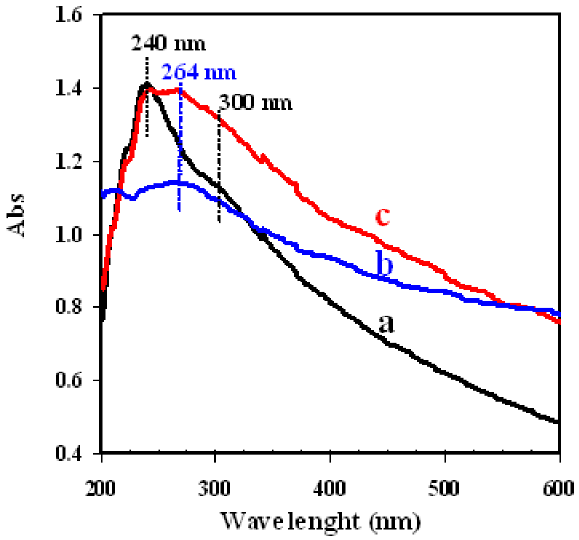

Fig. 2. UV-Vis spectra of non-irradiated GOM (a), GOM irradiated at 23.5 kGy (b) and 50 kGy (c).

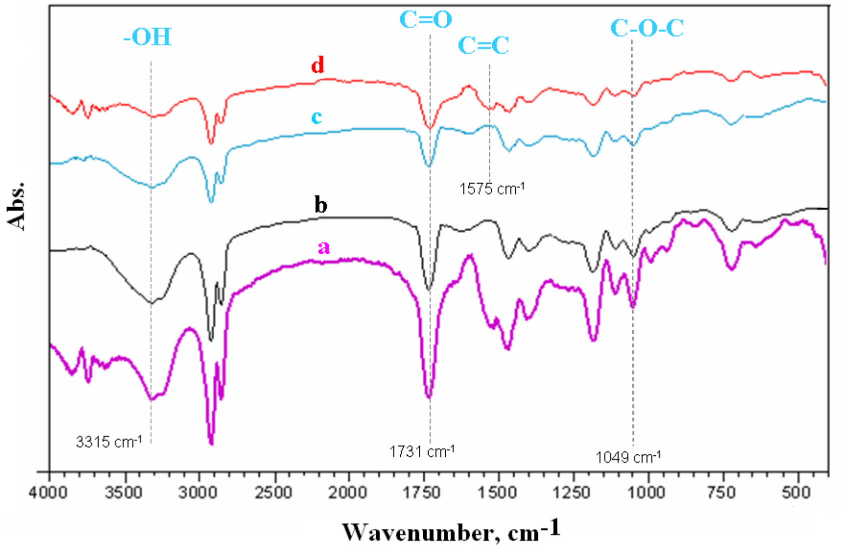

Fig. 3. FT-IR spectra of MG (a), non-irradiated GOM (b), GOM irradiated at 23.5 kGy (c) and 50 kGy (d).

For iGOM-50 kGy sample (Fig. 4d), a new broaden diffraction peak was appeared at 2θ = 21.52° (d-spacing = 4.12 Å) instead of the peak at around 10.87° of GO and GOM (0 kGy) suggesting the efficient reduction and exfoliation [11,15] of GO by γ-gamma irradiation in a MG/EtOH solution.

TEM analysis

Fig. 5 presents TEM images of non-irradiated GOM and GOM irradiated at 50 kGy. The non-irradiated GOM sheets are not perfectly flat. Dark areas in Fig. 5a indicate the thick stacking nanostructure of several graphene oxide and/or graphene layers in MG/EtOH solution.

After γ–ray irradiation, the image of iGOM-50 kGy (Fig. 5b) is observed on highly exfoliated graphite. The higher transparency areas indicate much thinner sheets of a few layers stacked attributed to reduced graphene oxide due to from stacking nanostructure exfoliation [15,18].

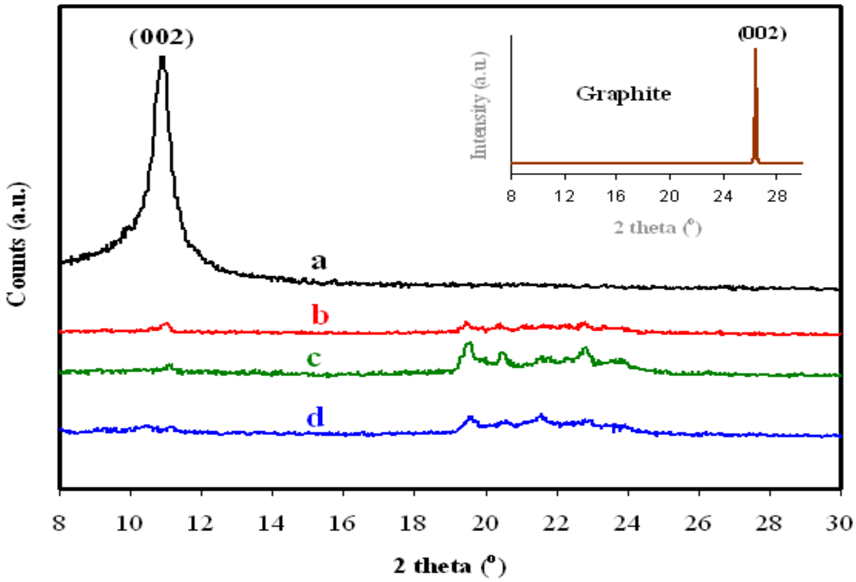

Fig. 4. XRD pattern of Graphite, GO (a), non-irradiated GOM (b), GOM irradiated at 23.5 kGy (c) and 50 kGy (d).

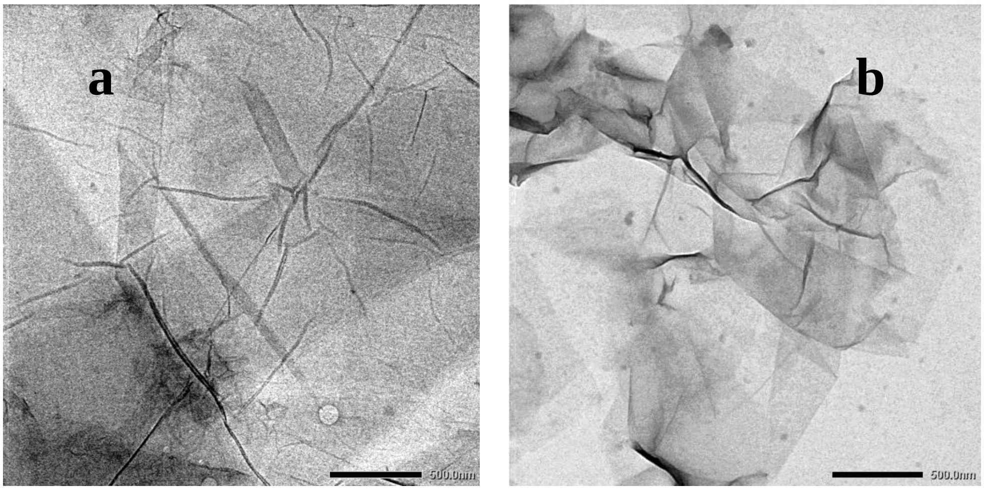

Fig. 5. TEM images of non-irradiated GOM (a) and iGOM-50 kGy (b)

III. CONCLUSIONS

GO was successfully reduced in a monoglyceride/ethanol solution by gamma irradiation under nitrogen gas. The UV-vis, FT-IR, XRD and TEM results confirmed that GOM can be simply and quickly reduced by γ –ray irradiation, and the resulting iGOM exhibited dose-dependent manner. This method is a promising way for the massive production of graphene from GO with a support of the self-assembly agent as MG.

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