

Article

Utilising Curcumin, Turmeric Extract and Cinnamon Extract Solutions for Graphite Liquid Phase Exfoliation

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Abstract. This work aimed at utilizing extracts from inexpensive plants including turmeric and cinnamon to exfoliate graphite compared with the expensive curcumin which was commercially derived from turmeric. The extract solutions were prepared from 5 g of powder immersed in 100 mL ethanol, which yielded the extract concentration of 10.4 and 16.8 mg/mL for turmeric and cinnamon, respectively, whereas curcumin solution at 0.5 mg/mL ethanol was used. The loading of graphite was fixed at 25 mg/mL. The mixture was sonicated at 50°C for 2 h at 200 W and 40 kHz, followed by settling at room temperature for 2 h. Finally, the exfoliated graphite was collected by centrifugation of the supernatant at 10500 rpm for 30 min. The yields obtained from using curcumin, turmeric extract and cinnamon extract were $0.28 \pm 0.07\%$, $0.32 \pm 0.096\%$ and $0.36 \pm 0.126\%$, respectively, comparable to those previously reported for liquid phase exfoliation. However, Raman spectroscopy results considering I_{2D}/I_G and FWHM of 2D peak showed that the quality of the sample using curcumin was the highest and comparable to the commercial graphene-based product, followed by turmeric and cinnamon. SEM micrographs did not show significant differences among all samples. A TEM figure showed that the exfoliated product was multi-layer graphene-based material.

Keywords: Graphene, liquid phase exfoliation, curcumin, turmeric, cinnamon

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1. Introduction

Graphene is a two-dimensional material composed of sp^2 -hybridized carbon bonds in a connecting hexagonal structure. Graphene composites are used to produce sport equipment, a bending touch screen, electrodes in batteries, and drug delivery materials because graphene has high strength, high toughness, high electrical and thermal conductivities at room temperature [1]. It could be applied together with magnetic quantum dots for sensing, water treatment and drug delivery [2]. In addition, it could be used as a template for the synthesis of ultrathin graphdiyne, which is another carbon allotrope with sp - and sp^2 -hybridizations [3]. Strictly defined, graphene is composed of only one layer, which is so called single-layer graphene and the extension has been used for few-layer graphene. Those with many layers could be called multilayer graphene or graphene-based materials.

Graphene is obtained from bottom-up methods, in which the molecules of carbon atoms are arranged to form graphene, such as pyrolysis and chemical vapor coating, and top-down methods, in which graphite layers held together by weak van der Waals forces are separated, such as mechanical and chemical exfoliations [1]. Among top-down options, Hummers method is commonly applied by using many strong oxidizing agents to form carbonyl and carboxylic groups on graphite surfaces, making their polarity so high that they could be separated and dispersed more easily [4]. The resulting product is called graphene oxide (GO). The yield of exfoliation is high with the expenses of lower electrical conductivity of the product and using a lot of water and hazardous chemicals. To regain the electrical conductivity, GO has to be reduced by a strong reducing agent to obtain reduced graphene oxide (rGO) [5-9]. However, for the application involving electrical conductivity of graphene, it closely relies on the extent of the reduction of graphene oxide.

An alternative to reduce the usage of dangerous chemicals and a large amount of water, and still maintain electrical properties as high as possible is liquid-phase exfoliation with various substances. For example, the exfoliation of graphite using 10 mg graphite powder in 1 mL N-methyl-2-pyrrolidone (NMP) with sonication for 48 h yielded 0.2% of exfoliated product and it was successfully applied as sensors for highly-sensitive detection of diethylstilbestrol and estradiol [10]. In addition, the direct exfoliation of graphite using NMP together with Fe_3O_4 nanoparticles in combination with ultra-sonication for 30–120 min was studied. It was found that using 0.5 mg graphite in 1 mL NMP gave the highest exfoliation yield of 19%. The presence of Fe_3O_4 nanoparticles also increased the exfoliation efficiency [11]. Therefore, the concentration of graphite could play a role in exfoliation. Furthermore, the exfoliation was improved by using NMP together with organic salts such as sodium citrate, sodium tartrate, and potassium citrate. The exfoliation of 10 mg graphite in 1 mL NMP with K-Citrate resulted in a graphene yield of 10% and up to 50% after 8 cycles of exfoliation [12].

Many interests have been directed to using aqueous solutions or other solutions with more environmentally-friendly solvents. Different organic surfactants and alkaline were used with water in the system of ultrasonication to obtain a suspension of single-layer or few-layer graphene. The exfoliation of graphite using 60 mg graphite in 150 ml 50:50 water: ethanol mixture was found more effective than using water or ethanol alone when treated by a sonotrode [13]. An aqueous solution of ammonia at pH 9 was used to exfoliate graphite in an ultrasonic bath. At the graphite concentration of 25 mg/mL, the yield of the exfoliated graphite was 0.29% which was 20 times higher than using water alone [14]. Besides, 1 w% urea solution was used to disperse graphite at 10 mg/mL together with ultrasonication. The yield was reported to be 0.12%. The authors hypothesized that urea could act as wedge molecules to attack the edges of graphite layers, leading to separation of layers [15].

Moreover, the use of natural substances has been investigated in the exfoliation of graphite. Curcumin was dissolved in ethanol to prepare the solution at 0.5 mg/mL and used in exfoliation with ultrasonication. It was found that the greater the power of the ultrasound machine, the greater the graphene concentration. But if it was more than about 1 kW, it would cause graphene to break, and when the sonication time exceeded 15 h, it could give only a little more amount of graphene. The maximum yield obtained was 1.38% when using the graphite concentration at 60 mg/mL and treating with the ultrasonic power at 1 kW for 4 h. It was hypothesized that curcumin could help exfoliated graphite layers because of π - π interactions among curcumin molecules and graphite [16].

Therefore, it is expected that π - π interactions between the phenolic compounds such as curcumin and graphite layers could facilitate the separation of graphite layers. Many natural substances were reported to contain various phenolic compounds including turmeric and cinnamon. Turmeric powder was composed of curcuminoids such as bisdemethoxycurcumin, demethoxycurcumin, and curcumin [17], whereas cinnamaldehyde, cinnamyl acetate, cinnamyl alcohol, and eugenol were found in cinnamon powder [18]. These natural substances are environmentally-friendly and safe to human beings. In addition, they are cost effective unlike pure curcumin, so they are more attractive in the practical use for a large-scale exfoliation. In this work, these substances were extracted by using ethanol and the potential of using the extracts in ethanol solutions to exfoliate graphite was explored along with curcumin. Both the yield of the exfoliation and the quality of the exfoliated graphite based on Raman spectroscopy were reported.

2. Materials and Method

2.1. Materials

Graphite powder (extra pure powder with particle size less than 50 μm) and ethanol (96%, C_2H_5OH) were

purchased from Merck, Ltd. Curcumin was purchased from Toronto Research Chemicals Inc. Turmeric powder (Sabai Sabai brand, produced from turmeric stem underneath the soil) and cinnamon powder (McGarrett brand, produced from inner shell of Cinnamomum branch) were purchased from a local supermarket. Commercial graphene-based powder (powder with the thickness of 6-8 nm and the width of 5 μm) was purchased from Tokyo Chemical Industry Co., Ltd. Sodium carbonate was purchased from Kemaus Pty Ltd. Folin & Ciocalteu Reagent and gallic acid were supplied by Sisco Research Laboratories Pvt. Ltd., India.

2.2. Preparation of dispersing media for exfoliating graphite

Because water solubility of curcumin is very low, the solution of curcumin was prepared by using ethanol instead. Subsequently, for comparison, ethanol was used to prepare turmeric and cinnamon extracts. 0.5 mg/mL curcumin solution was prepared by dissolving 0.0375 g curcumin in 75 mL ethanol. The solution was mixed vigorously with a vortex mixer.

Turmeric and cinnamon extracts were prepared by dispersing 5 g turmeric or cinnamon powder in 75 mL ethanol at about 45°C for 1 h. The mixtures were then filtered with a piece of filter paper and a yellowish-orange extract of turmeric and a dark brown extract of cinnamon were obtained. The liquids were measured for absorbance intensities in the range of 340-940 nm. The concentrations of the turmeric and cinnamon extracts in ethanol were determined by sampling 1 mL extract, placing on a piece of aluminum foil, and drying at 80°C until the weight was constant.

2.3. Determination of the total phenolic content (TPC) of the extract solutions

To determine TPC, 0.75 mL of Folin-Ciocalteu reagent solution was pipetted and diluted with 7.5 mL of water. Subsequently, 0.1 mL of the undiluted extract solutions were added. The mixture was thoroughly blended using a vortex mixer and allowed to stand for 5 min. Next, 0.75 mL of 10% sodium carbonate solution were added to the mixture, which was then mixed again using a vortex mixer. The mixture was left at room temperature for 90 min. The absorbance was measured at a wavelength of 725 nm. The absorbance values were compared with the standard curve prepared from gallic acid at various concentrations.

2.4. The exfoliation of graphite in curcumin, turmeric extract and cinnamon extract solutions

The exfoliation process in this study followed steps explained in our previous work [19] where left-over ammonia in the serum of skim natural rubber latex was used in the exfoliation of graphite as shown in Fig. 1. The graphite powder was dispersed in a liquid at different concentrations of 15, 20, 25 and 30 mg/mL. For example,

the sample of 25 mg/mL was prepared by mixing 250 mg graphite powder with 10 mL liquid. The suspensions in closed bottles were placed in an ultrasonic bath operating at 200 W and 40 kHz and the temperature was controlled at 50°C for 2 h. After that they were left standing at room temperature for another 2 h. At this step, the scheme was separated into 2 paths, Path#1 with two times of centrifugation (full line) and Path#2 with one time of centrifugation (dash line).

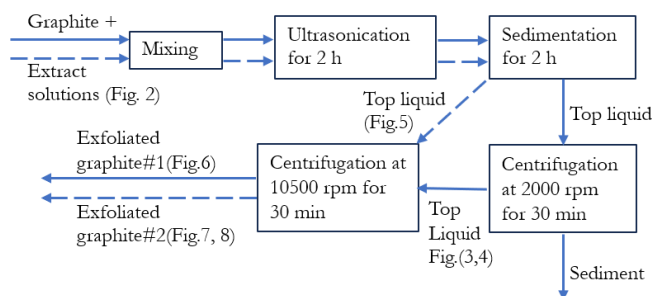


Fig. 1. The experimental scheme of the exfoliation of graphite in solutions of natural substance extracts.

For two-time centrifugation, 7 mL of the top liquid of the settling suspension was centrifuged at 2000 rpm for 30 min, the supernatant of which was measured for its absorbance intensities by a visible spectrophotometer at wavelengths of 340-940 nm. Finally, it was centrifuged at 10500 rpm for 30 min to collect the sediment of the exfoliated graphite. For one-time centrifugation, 7 mL of the top liquid was characterized with the visible spectrophotometer and centrifuged at 10500 rpm for 30 min to collect the sediment. The collected solid was washed with ethanol and subsequently with water. It was dried at 80°C to a constant weight so that the process yield could be determined from Eq. (1). The reported %yields were the average from three replicates. The samples were then characterized by Raman spectroscopy, scanning electron microscopy and transmission electron microscopy.

$$\% \text{ yield} = \frac{\text{weight of suspended solid (g)}}{\text{weight of graphite used (g)}} \times 100 \quad (1)$$

Initially, the extract solutions were used undiluted. Later, they were diluted with ethanol using the dilution ratio of the extract solution volume to the solvent volume at 1:1 and 1:3 and used in the exfoliation step via Path#1 in Fig. 1.

2.5. Characterization of the exfoliated graphite samples

2.5.1. Raman spectroscopy

Raman spectroscopy results of the chosen exfoliated graphite samples were obtained using a dispersive Raman microscope (Bruker Optics, Model: Senterra) with the

laser wavelength at 532 nm, the laser power at 5 mW and the spectral range of 4500 - 70 cm^{-1} . The measurement followed the standard methods, ASTM: E1693 and ASTM: E1840.

2.5.2. Scanning electron microscopy (SEM) and transmission electron microscope (TEM)

The structures and surface morphology of the chosen uncoated exfoliated-graphite samples were investigated using a scanning electron microscope (SEM, JSM-7800F) with an accelerating voltage of 15 kV. The magnification was adjusted to 10,000X. In addition, a chosen sample was checked with transmission electron microscope (TEM, JEM-2010) with voltage of 200 kV and the magnification of 500,000X.

2.6. Determination of activity coefficients using Universal Quasi-Chemical Functional Group Activity Coefficient (UNIFAC) model.

In this work, the analysis of activity coefficient is based on UNIFAC model. In the model, the activity coefficient is composed of two parts, a combinatorial term to account for the molecular size and shape differences and a residual term to account for molecular interactions. Each part contains many parameters calculated step by step according to the many equations provided in [20].

In this work, the activity coefficients of the substance and graphite (which is assumed to consist of 10,000 carbon atoms) at 323 K (50°C) were calculated. The substances including curcumin (as a constituent in turmeric), cinnamaldehyde (as a constituent in cinnamon), ethanol, NMP (extensively investigated in the literature of exfoliation of graphite) and water were compared in this work.

3. Results and Discussion

3.1. Characterization of the extract solutions

Curcumin, turmeric and cinnamon extracts in ethanol solutions were measured for their absorbance intensities which are shown in Fig. 2 along with the colors of the solutions. The broad peaks were seen at 500, 510 and 550 nm for curcumin, undiluted turmeric extract and undiluted cinnamon extract solutions, respectively. Close to 428 nm reported in a literature [21], the characteristic peak of curcumin was seen at 440 nm when the solution was diluted twofold and fourfold with ethanol. The intensity of the characteristic peak dropped when the curcumin concentration decreased while the absorbance intensities at wavelengths greater than 650 nm were higher when the amount of ethanol was greater. The peak positions also changed for turmeric extract and cinnamon extract solutions when they were diluted with ethanol. For both extracts, the absorbance intensities at wavelength greater than 650 nm were higher when the amount of ethanol was greater like the case of curcumin solution. For the extracts,

Fig. 2(a) and Fig. 2(b) show that when diluted, there was no prominent peak to be seen unlike the characteristic peak of curcumin, because different molecules in the extracts contributed to the absorbance intensity differently and might be combined as broad spectrum.

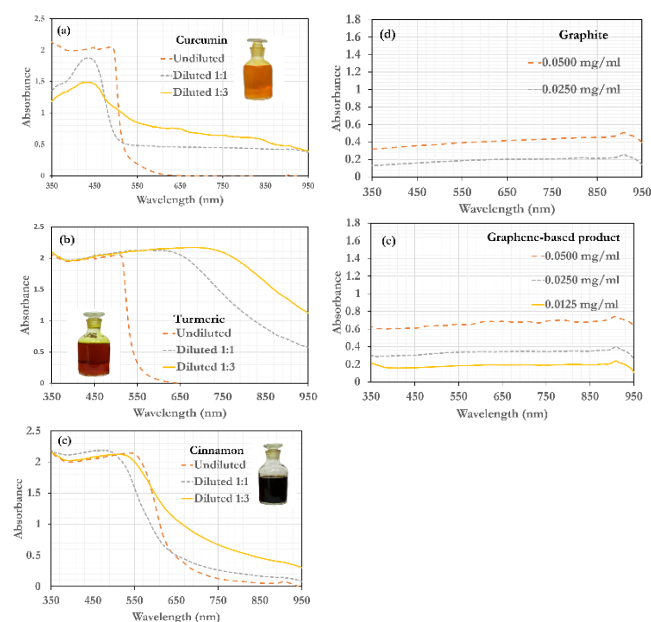


Fig. 2. UV-Vis Absorbance Spectra of the solutions of (a) curcumin, (b) turmeric extract and (c) cinnamon extract and the spectra of the suspensions of (d) the precursor graphite and (e) the commercial graphene-based powder.

As expected, the peak of undiluted turmeric extract solution was similar to that of undiluted curcumin solution because curcumin is a constituent in turmeric extract. Based on HPLC results [17], it was reported that in one liter of the extract, there were 15.02 mg curcumin, 25.50 mg demethoxycurcumin and 22.19 mg bisdemethoxycurcumin. Therefore, the total concentration of the three curcuminoids was about 62.71 mg/L, which was only 0.063 mg/mL, much less than 0.5 mg/mL, the concentration of pure curcumin used in this study.

In this work, based on the weight of the dried extracts, the concentrations of turmeric and cinnamon extracts were 10.4 mg/mL and 16.8 mg/mL, respectively. In addition, the total phenolic contents (IPC) of the obtained extracts were found to be 182.5 and 112.7 mg GAE/g extract, respectively, confirming the presence of polyphenols in both extracts that could help facilitate the exfoliation of graphite.

Based on proximate analysis in a report [22], the percentages of carbohydrate, fiber, fat, proteins and ash in turmeric powder were 66.55, 4.80, 7.27, 7.23 and 5.72, respectively. The rest was moisture and other substances. Therefore, a larger quantity of soluble components other than curcuminoids were present in the extracts. For cinnamon extract, it was composed of 4.16% carbohydrate, 16.67% fat, 16.67% protein, 20% ash and the rest were moisture and other substances [18]. Based

on GCMS results [18], among other phenolic compounds, cinnamaldehyde was reported to be the major active compounds with the concentration as high as 60-80%. Like turmeric extracts, other soluble components with a larger quantity than cinnamaldehyde should be present in the extracts.

Figure 2 also shows spectra of the precursor graphite powder and the commercial graphene-based powder suspended in ethanol. The suspension was left standing for 10 min and the upper liquid was sampled and checked with spectrophotometer. It is evident that the absorbance intensities of the suspension of the commercial graphene-based powder are greater than those of graphite powder at the same concentration because graphene-based powder could disperse better in the solvent and suspended for a longer time before settling down. The characteristic peaks at 910 nm were seen in both Fig. 2 (d) and Fig. 2(e) indicating the interaction of ethanol and graphitic structures. In addition, it was also seen for the commercial graphene-based powder that the line was curved up around 350 nm, which actually was part of the absorbance peak observed with graphene in the UV range. The technique of spectrophotometry was also applied to the suspension of the exfoliated graphite suspended in the solutions of curcumin and extracts as discussed in the next section.

3.2. UV-Visible spectrophotometry results of the exfoliated graphite suspensions

Following Path#1 of the experiments described in Section 2.3, the absorbance intensities of the mixtures after two-time centrifugation were measured and displayed in Fig. 3(a)–(c) for curcumin-treated, turmeric extract-treated and cinnamon extract-treated samples, respectively. Fig. 3(a)–(c) show that the peaks at 910 nm are observed for all samples, representing the interaction of ethanol and graphitic structures so all samples had graphitic products suspending. It is seen that the absorbance intensities of mixtures using the graphite concentration at 25 mg/mL are the highest among all concentrations, implying that the yield is the highest. The absorbance intensities should be related to the yield as it has been recently reported that the absorbance at 660 nm of the exfoliated graphite in ethanol and water was the highest when compared with using ethanol or water alone, corresponding to yield twice that of using water [13]. However, the amounts of the obtained products from the experiments of using curcumin solution, turmeric extract solution and cinnamon extract solution were 0.06 ± 0.009 , 0.08 ± 0.016 and 0.04 ± 0.01 mg, respectively. According to Eq. (1), the yields were $0.024 \pm 0.0004\%$, $0.032 \pm 0.0064\%$ and $0.016 \pm 0.0004\%$, respectively, which were very low.

Considering the interactions of natural substances and the graphite powder, it could be seen from the spectra that after the samples went through centrifugation at 2000 rpm for 30 min in the exfoliation procedure in Path#1, curcumin molecules were not significantly adsorbed on the graphite powder while molecules in turmeric and

cinnamon extracts were greatly adsorbed as the absorbance intensities around the peaks of the supernatant of the turmeric extract-treated and cinnamon extract-treated samples decreased at all graphite concentrations.

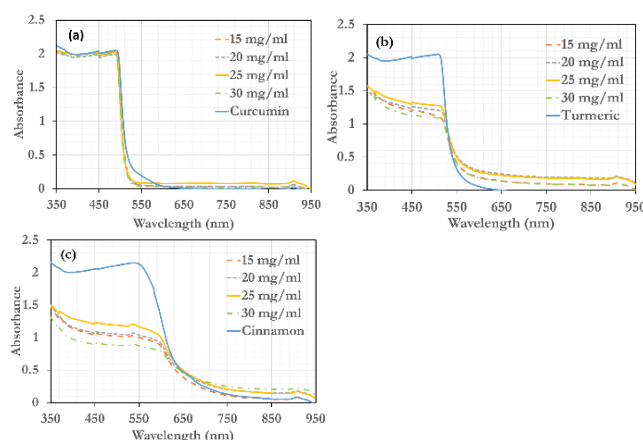


Fig. 3. UV-Vis absorbance spectra are shown of the mixtures with (a) curcumin solution, (b) turmeric extract solution and (c) cinnamon extract solution, which were obtained after centrifugation at 2000 for 30 min (Path#1).

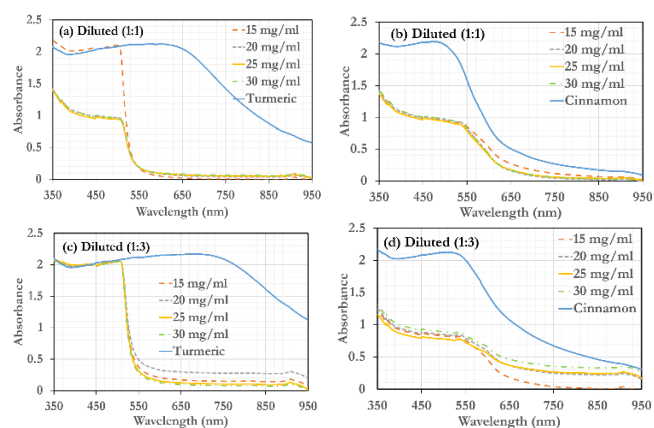


Fig. 4. UV-Vis absorbance spectra are shown of the mixtures with (a) diluted turmeric extract solution (1:1), (b) diluted cinnamon extract solution (1:1), (c) diluted turmeric extract solution (1:3) and (d) diluted cinnamon extract solution (1:3), which were obtained after centrifugation at 2000 rpm for 30 min (Path# 1).

In later experiments using the diluted extract solutions to disperse the graphite powder, the UV-absorbance spectra changed as shown in Fig. 4 because the adsorption of substances on solid surfaces depends on many factors including the amount of adsorbent (the graphite powder) and the concentration of adsorbate (molecules in turmeric and cinnamon extracts). Fig. 4 apparently shows these effects. As the concentration of turmeric extract was lowered by dilution with ethanol, the change was observed from that adsorption occurred at all graphite concentrations in Fig. 3(b), to that adsorption occurred in all, except for the case of the lowest graphite

concentration in Fig. 4(a), and to that no adsorption occurred at all graphite concentrations in Fig. 4(c). This was consistent with the theory that lowering the concentrations of adsorbate and adsorbent causes the chemical potential, the driving force in mass transfer, to decrease. For the case of cinnamon extract, adsorption occurred in all cases of different graphite and extract concentrations.

Nonetheless, considering the low yields of the exfoliated graphite obtained from the exfoliation procedure in Path#1, which were also reflected by low values of absorbance intensities (less than 0.5) in Fig. 3 and Fig. 4 at wavelengths greater than 650 nm, Path#1 was not practical. Therefore, the exfoliation procedure was adjusted to Path#2 by removing the step of centrifugation at 2000 rpm for 30 min. Even though the absorbance intensities at wavelengths greater than 650 nm for the diluted extract cases were higher than those of the undiluted extract cases, it might be attributable to the absorbance intensities of the extract solutions which were higher for the diluted cases. Therefore, Path#2 was investigated by using the graphite concentration at 25 mg/mL in undiluted extract solutions, which gave the highest yield in the previous study. The supernatant after settling at room temperature for 2 h was checked with the spectrophotometer and was centrifuged at 10500 rpm for 30 min to collect the exfoliated graphite. The absorbance spectra are shown in Fig. 5.

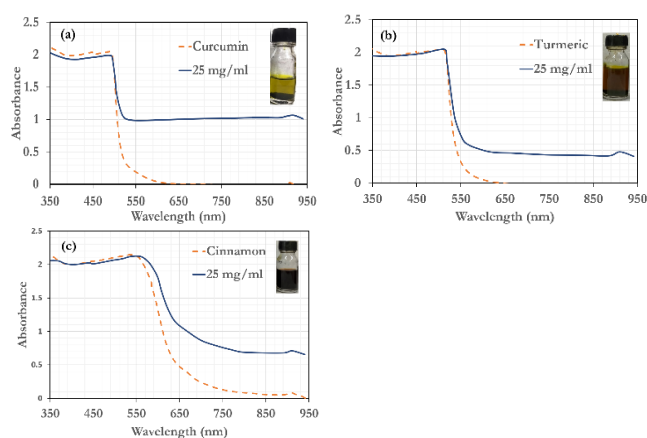


Fig. 5. UV-Vis absorbance spectra are shown of the mixtures with (a) curcumin solution, (b) turmeric extract solution and (c) cinnamon extract solution, which were obtained after settling for 2 h (Path#2).

Figure 5(a)-(c) show that the absorbance intensities at the wavelengths greater than 650 nm were all greater than 0.5, implying that higher yields of the exfoliated graphite were obtained and adsorption was not significant. It seemed that centrifugation at 2000 rpm for 30 min facilitated the adsorption of extract molecules on the surface of the graphite powder because the effect of centrifugal force was combined with the effect of concentration gradient for the extract molecules to reach the surface of the graphite powder. In other words, only settling by gravity did not help facilitate the adsorption.

The weights of the dried suspended solids were 0.7 ± 0.175 , 0.8 ± 0.24 and 0.9 ± 0.315 mg from 10 mL curcumin-treated, turmeric extract-treated and cinnamon extract-treated samples, respectively, corresponding to the yields of $0.28 \pm 0.07\%$, $0.32 \pm 0.096\%$ and $0.36 \pm 0.126\%$, which were not consistent with the order of the absorbance intensities at wavelengths greater than 650 nm in Fig. 5. This could be due to the quality of the exfoliated graphite, i.e. fewer number of layers. The dispersion of solids suspended uniformly throughout the liquid volume can play an important role in light absorption as shown earlier in Fig. 2(d) and Fig. 2(e), where the amounts of the graphite powder and the commercial graphene-based product were the same.

These yields were comparable to or greater than those reported in some literatures for liquid phase exfoliation [14,15]. Specifically, the yield reported in the literature [16] using curcumin solution at 0.5 mg/mL and graphite concentration at 50 mg/mL in the ultrasonic system at 180 W for 4 h was 0.02 mg/mL exfoliated graphite or 0.04%, which was lower than our case. In our case, the same concentration of curcumin was used with the graphite concentration at 25 mg/mL in the ultrasonic bath at 200 W at 50°C for 2 h and the yield was $0.28 \pm 0.07\%$. Therefore, in our case, the higher temperature could compensate for a shorter sonication time.

A literature comprehensively explained the effect of sonication on liquid phase exfoliation of graphite with video recording and pressure measurement [13]. The mechanism begins with the generation of primary cavitation cloud, the large bubble cluster formed directly under the sonotrode tip, and secondary cavitation cloud, the bubbles separating from the primary cavitation cloud. Both clouds contribute to the moving bubbles and bubble clusters in the overall space of cavitation zone. After the cavitation cloud collapses, many shock waves with large pressure spikes are generated and assist the exfoliation of graphite. The collapse of a large cloud often initiates the collapse of bubbles in the secondary cloud, yielding a higher intensity of pressure spikes. In addition, a higher temperature increases the number of bubbles, thereby increasing the shock wave generation.

In our study, it could be hypothesized that initially, the ultrasound bath generated fine bubbles inside the suspension in the bottle. Once the bubbles collapsed, generating shock waves with pressure high enough to push the edges of adjacent layers of graphite apart from each other, which can accommodate the molecules of phenolic compounds such as curcumin into this space. The interactions between phenolic groups in curcumin molecules and carbon atoms in sp^2 -hybridized bonds in hexagonal structures of graphite layers via π - π interactions would weaken the van der Waals forces between two layers, yielding greater separation distance between two layers. Both the attacks from shock waves and weakening the interlayer interactions continued until the two layers were completely separated. It was also possible that when the interlayer interactions were weaker, one layer of graphite could be shifted spatially far from another one,

leading to even lower interactions and the exfoliation could be easier.

3.3. Morphology of the exfoliated graphite

The micrographs of the exfoliated graphite samples using graphite concentration at 25 mg/mL and different liquids were obtained by scanning electron microscopy. Firstly, the exfoliated samples from Path#1 were investigated. Fig. 6(a)-(c) are micrographs of curcumin-treated, turmeric extract-treated, cinnamon extract-treated samples after washing with ethanol and water while Fig. 6 (d) is the micrograph of cinnamon-treated sample before washing. It is obviously seen that there are small particles deposited on the surface of graphite powder. These particles should be the undissolved part in the turmeric extract which cannot be observed with naked eyes. Clearly, the washing step is necessary.

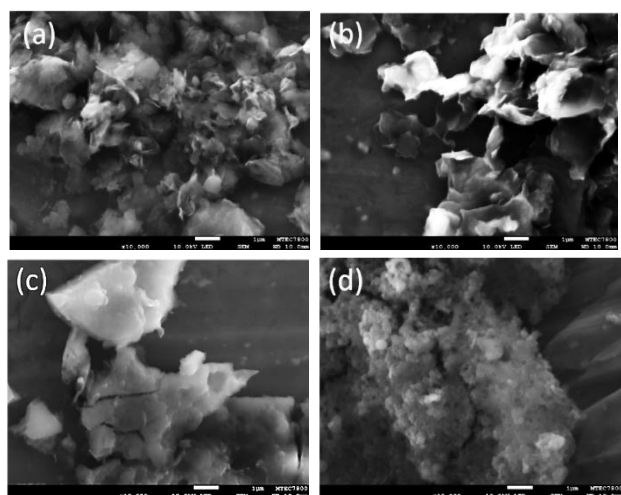


Fig. 6. SEM micrographs with the scale bar of 1 µm of the exfoliated graphite samples treated with (a) curcumin, (b) turmeric extract and (c) cinnamon extract, from Path#1 after washing, are displayed along with (d) the cinnamon-extract treated sample before washing.

The micrographs of the exfoliated graphite samples from Path#2 along with those of the precursor graphite and the commercial graphene-based product are displayed in Fig. 7. It was evidently seen that the sizes of the exfoliated graphite samples in Fig. 7 were larger than those in Fig. 6, confirming that the step of centrifugation at 2000 rpm for 30 min in Path#1 removed a lot of powders with larger sizes.

Overall, the figures of the exfoliated samples were similar. Many plates were seen stacking and each plate might actually be the graphene-based product, which could be confirmed by TEM in Fig. 7(f) showing many layers (e.g. 8 and 10 layers in the figure) of graphene-based material was obtained when using the turmeric extract solution in exfoliation. The quality of the exfoliated graphite had to be checked by Raman spectroscopy in the next section.

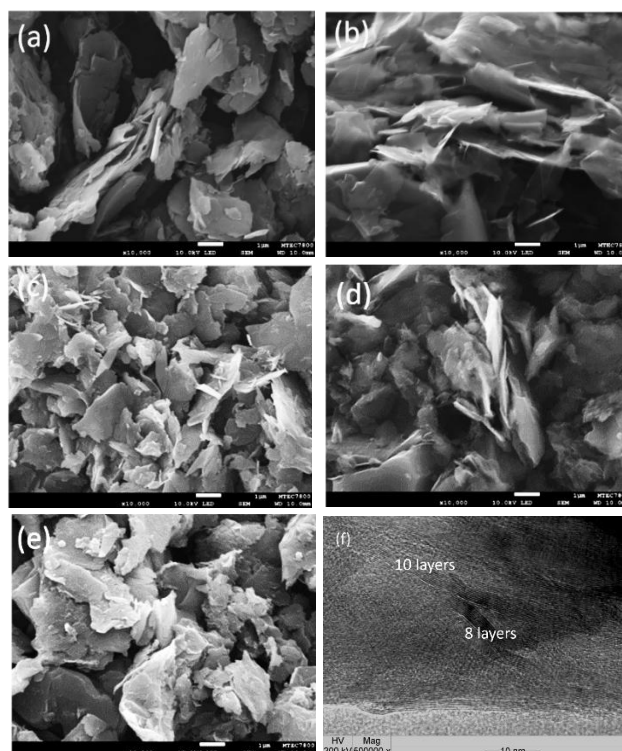


Fig. 7. SEM micrographs with the scale bar of 1 µm of (a) the precursor graphite, (b) the commercial graphene-based powder and the exfoliated graphite samples treated with (c) curcumin, (d) turmeric extract and (e) cinnamon extract, from Path#2 after washing are displayed. TEM figure of an exfoliated sample is also shown in (f).

3.4. Raman spectroscopy results

The characteristics of the exfoliated graphite samples, the precursor graphite, and the commercial graphene-based product can be investigated through Raman scattering. It can be used to indicate crystal flaws and the number of layers in graphitic materials. In the measurement, the laser photons excite the electron-hole pair causing the inelastic scattering of the electrons and holes by phonon emission/absorption and elastic scattering of the electrons mediated by the defect and recombination of the electron-hole pair. Both inelastic and elastic scatterings yielded characteristic peaks of graphitic structures including D-peak at ~1350 cm⁻¹, G-peak at ~1580 cm⁻¹ and 2D-peak at ~2700 cm⁻¹. The secondary peak such as D'-peak at 1620 cm⁻¹ was also reported [23]. The Raman spectra of the chosen exfoliated samples are shown in Fig. 8 and some important values are tabulated in Table 1.

When the elastic scattering of the electron by one phonon is mediated by the defect, D-peak is generated. Defects in graphitic structures could be from three sources, the first of which is the disorders at the edges of all layers, the second is from vacancies in the structures and the third is from the functionalization of carbon atoms. The last one is most found in structures of graphene oxide, where the carbonyl and carboxylic groups are formed with sp³-type bonding. Besides, D'-peak which

is adjacent to G-peak also indicates the type of defect when the ratio of $I_{D'}/I_D$ is considered. It was reported that the value of the ratio could reach about 3.5 for edge disorders, 7 for vacancies in crystal structures, and 13 for sp^3 -type defect [23].

Table 1 shows that the values of this ratio of our samples were 1.15-1.37 implying that the defects were mainly from the edge disorders which could imply the multilayers of the exfoliated products. They were larger than that of the commercial graphene-based product but smaller than that of the graphite precursor.

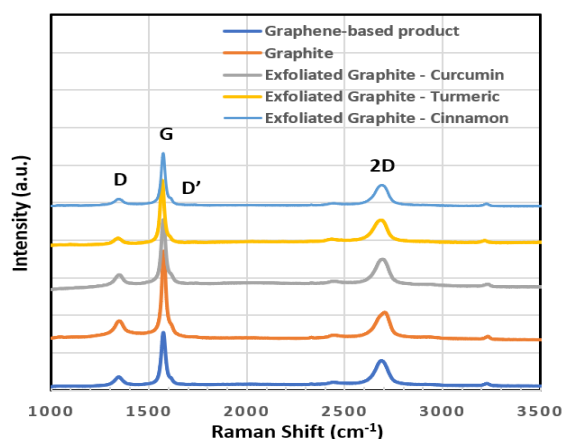


Fig. 8. Raman spectra of the exfoliated graphite samples were compared with those of the precursor graphite and the commercial graphene-based powder.

Table 1. Results from Raman spectra, including primary peak positions according to [23], their intensities, and the corresponding peak ratios of the exfoliated samples.

Sample	D-peak (cm ⁻¹)	I_D	G-peak (cm ⁻¹)	I_G	G- FWHM (cm ⁻¹)	2D-peak (cm ⁻¹)	I_{2D}	2D- FWHM (cm ⁻¹)
Graphene	1344.2	1817.00	1572.9	7739.20	25.7	2686.2	3967.60	73.8
Graphite	1347.8	3265.20	1575.1	12580.10	24.1	2706.0	4404.70	78.3
Curcumin	1347.0	3412.30	1573.5	10748.06	25.9	2695.5	5481.71	74.4
Turmeric	1341.0	2285.84	1566.5	9992.31	23.7	2687.0	4689.12	76.5
Cinnamon	1343.5	1477.25	1572.5	7582.81	24.5	2690.0	3345.07	76.3

Sample	D'-Peak (cm ⁻¹)	$I_{D'}$	$I_D/I_{D'}$	$I_{D'}/I_G$	I_D/I_G	I_{2D}/I_G
Graphene	1622.0	2331.39	0.78	0.30	0.23	0.51
Graphite	1626.0	1196.97	2.73	0.10	0.26	0.35
Curcumin	1625.0	2954.74	1.15	0.27	0.32	0.51
Turmeric	1625.0	1950.71	1.17	0.20	0.23	0.47
Cinnamon	1624.5	1079.71	1.37	0.14	0.19	0.44

G-peak represents C-C stretching mode in sp^2 -hybridized carbon bonds in the graphitic structures. Many factors could influence this bond stretching include defects, doping, strain and temperature. Its shift towards higher wavenumbers indicates both disorder in regular structure and an increase in sheet thickness [13]. It could

be seen in Table 1 that G-peak shifted from 1575.1 cm^{-1} for the precursor graphite to 1573.5, 1566.5 and 1572.5 cm^{-1} for curcumin-treated, turmeric extract-treated and cinnamon extract-treated samples, respectively, indicating that the exfoliated graphite should be thinner than the precursor graphite. These values were close to that of the commercial graphene-based product, which was 1572.9 cm^{-1} . In addition, broader G-peak corresponds to higher degree of disorder [13]. Therefore, the full width at half maximum (FWHM) of G-peak should be compared in Table 1. The values of G-FWHM of curcumin-treated, turmeric extract-treated and cinnamon extract-treated samples were 25.9, 23.7 and 24.5 cm^{-1} , respectively, which were around those of the precursor graphite (24.1 cm^{-1}) and the commercial graphene-based product (25.7 cm^{-1}).

It is common to compare I_D/I_G ratios of the samples in order to attenuate the effect of spatial dimension when studying the defects [23]. For graphene with few layers, it was reported that when defect density increased, the value of I_D/I_G increased to a maximum of 3.5 and then decreased [24]. In our case, there were no chemical reactions involved and the values of I_D/I_G of all samples were much less than 3.5 so they should be in the low-defect density region, where the values of the ratio increased with the defect density. The values of I_D/I_G of curcumin-treated, turmeric extract-treated and cinnamon extract-treated samples were 0.27, 0.20 and 0.14, respectively. Similar to values of G-FWHM, these values were around those of the commercial graphene-based powder (0.23) and the precursor graphite (0.26) indicating that edge disorders were not significantly occurred during ultrasonication-assisted exfoliations of all samples. In the exfoliation of graphite using water and ethanol solution together with the sonotrode, it was reported that the value of I_D/I_G was about 0.25-0.50 [13]. In contrast to the reduced-graphene oxide sheets undergoing the oxidation-reduction reaction, whose I_D/I_G ratio was between 1.1-1.5 [23], those of our samples were still much lower.

While D-peak represents the defects, 2D-peak is related to the number of graphene layers. Generally, the values of I_{2D}/I_G decreased as the number of layers increased but it is not enough to determine the exact number of layers due to complication from doping and laser polarization effect [25]. 2D-peak appears because of the scattering of electron by two phonons before it recombines with the hole. If there are more defects and electrons are mediated with the defects, fewer numbers of electrons will be recombined with the holes, resulting in lower 2D-peak [22]. For multilayer graphene, the edge defects could decrease with a decrease in the number of layers. It was stated that values of I_{2D}/I_G could be increased upon exfoliation from 0.35 of graphite to 0.50 of multilayer graphene, and to 0.69 of bilayer graphene [24].

In Table 1, the value of I_{2D}/I_G of curcumin-treated sample was the largest (0.51), greater than that of turmeric extract-treated sample (0.47), which was larger than cinnamon extract-treated sample (0.44). They were greater than that of the precursor graphite (0.35), indicating that

the exfoliation of graphite truly occurred by using ultrasonication with any of these substances. The value of the curcumin-treated sample was comparable to that of the commercial graphene-based powder (0.51), indicating that both of them should be multi-layer graphene [25].

Considering the amount of curcuminoids in the turmeric extract which was only 0.063 mg/mL as discussed earlier in Section 3.1, it showed a good potential in exfoliation of graphite. Cinnamon extract solution was the least effective in exfoliation of graphite probably because the phenolic compounds such as cinnamaldehyde in cinnamon caused weaker π - π interactions than those in turmeric. Unfortunately, the concentration of these phenolic compounds in cinnamon seemed to reach the maximum as small particles were found undissolved and already deposited on the surface of the graphite powder as observed in Fig. 6(d). Considering all exfoliated samples, it could be seen that when the value of I_{2D}/I_G increased, the value of I_D/I_G and $I_{D'}/I_G$ also increased, implying that when the precursor graphite was exfoliated to yield exfoliated graphite with fewer layers, the number of defects were increased, probably because more edges could be exposed and ultrasonic treatment could cause defects in the structures [13].

Moreover, another parameter, the full width at half maximum of 2D-peak (2D-FWHM) is used to relate to the number of layers of graphitic materials too. Because 2D-peak is resulted from constructive interferences of all peaks generated by each layer, the 2D-FWHM is larger for the sample with more numbers of layers [24]. Obviously, the 2D-FWHM of curcumin-treated sample was the smallest (74.4 cm^{-1}), which was smaller than those of turmeric extract-treated (76.5 cm^{-1}) and cinnamon extract-treated (76.3 cm^{-1}). They were smaller than that of the precursor graphite (78.3 cm^{-1}), indicating that the exfoliated graphite samples possessed fewer layers than the precursor graphite. That of curcumin-treated sample was comparable to that of the commercial graphene-based product (73.8 cm^{-1}), confirming again that curcumin-treated sample had the quality of the commercial graphene-based product used in this study. 2D-FWHM was reported for single-layer, bilayer, trilayer, four-layer and five-layer graphene [25]. The 2D-FWHM of five-layer graphene was between 65 and 67.5 cm^{-1} . Therefore, all the obtained samples as well as the commercial graphene-based powder should be multilayer graphene composed of many more than 5 layers.

3.5. Thermodynamic analysis based on Universal Quasi-Chemical Functional Group Activity Coefficient (UNIFAC) model.

The results above show that exfoliation of graphite really occurred in all experiments with different degrees of exfoliation when using different natural substances, indicating the role of natural substances in assisting the exfoliation of graphite when ethanol was used as the solvent and the condition of ultrasonication was set the same in all experiments. The effect of curcumin, turmeric

extracts and cinnamon extracts was thought to relate with the intermolecular interactions between the extract molecules and graphite layers. The investigation of intermolecular interaction could be performed directly with molecular simulation with various models of molecular potentials and indirectly with some thermodynamic properties such as activity coefficient.

Activity coefficient is defined as the ratio of the fugacity of a substance in the real mixture to that of the same substance in the ideal mixture. The ideal mixture or the ideal solution is the solution with no change of interactions between the different species when compared with the interactions in the pure component. Therefore, activity coefficient indicates the change of interactions between different substances. Basically, the activity of a substance in the mixture is calculated from the derivative of the excess Gibbs free energy of the solution respect to the number of moles of that species and could be modeled with various equations based on experimental results such as Margules and Wilson model. There are also the predictive models of activity coefficient including Universal Quasi-Chemical Activity Coefficient (UNIQUAC) and Universal Quasi-Chemical Functional Group Activity Coefficient (UNIFAC) models, the former of which is based on the interactions among all real functional groups of the molecules and the latter is based on the interactions among different structural units (part of functional groups) from which the molecules are formed.

Usually, activity coefficients were used in the study of liquid-vapor and liquid-liquid equilibrium. They were used in the study of liquid-solid equilibrium as well. Little was known in the investigation of activity coefficients in case of solid-solid substances [26]. However, its definition is based on Gibbs free energy no matter what state of the two materials.

As shown in Fig. 9, the activity coefficients of all substances when they were mixed with graphite were less than one while the activity coefficients of graphite when it was mixed with each substance were greater than one. When the activity coefficient is greater than one, that material is likely to have less attraction among their molecules, implying that all substances could help decrease interactions among graphite powders (or graphite layers) at different degrees. The highest was observed for curcumin, followed by NMP, water, ethanol and cinnamaldehyde and the effect was stronger at higher concentrations of the substances. This was consistent with the Raman spectroscopy results in Section 3.4, where the turmeric extract-treated sample had fewer number of layers than cinnamon extract-treated sample. In Fig. 9(a), when the activity coefficient of the substance was less than one, implying that attractions among substance molecules are stronger with the presence of another substance but the effect was weaker (the activity coefficient is closer to one), when the concentrations of substances are higher. The activity coefficient in this study could confirm the assumption of the attractive interactions between the phenolic compounds and graphite layers.

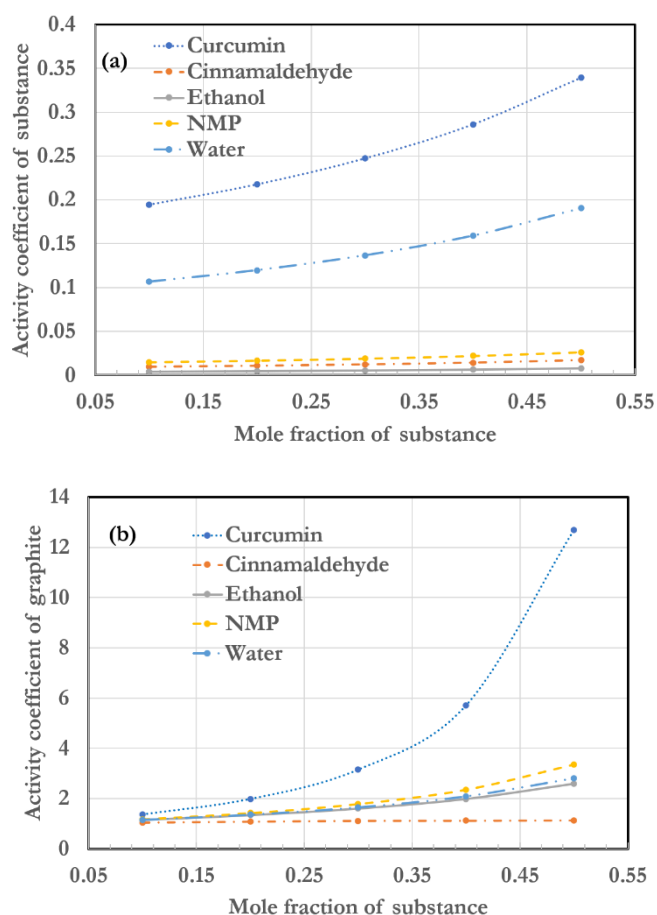


Fig. 9: Activity coefficients of (a) substances and (b) graphite based on UNIFAC model when they are mixed at different mole fractions.

Ethanol itself can exfoliate graphite at some degree but together with curcumin, they can greatly facilitate the exfoliation of graphite as shown in this study. It seemed that using higher concentration of curcumin could help facilitate the exfoliation of graphite at a higher degree. However, in practice, the solubility of curcumin is limited to 1 mg in 1 mL of 100% ethanol or only 0.6 μg in 1 mL of water. Therefore, to increase the interaction between curcumin and graphite, there are two possible ways, firstly changing the solvent to increase solubility of curcumin and secondly using curcumin powder directly in solid-phase exfoliation of graphite. According to molecular structures of substances in turmeric and cinnamon, there are many nonpolar functional groups. Therefore, the selected solvents to be used in extraction should have a fewer degree of polarity. In addition, less polar solvent should help dispersing the graphite powder more effectively. However, the selected solvents have to be environmentally friendly.

Because curcumin is a promising material to be used in exfoliation of graphite, turmeric should be of interest too since it contains curcumin. As for cinnamon, some other factors to increase the solubility of their phenolic substances should be explored.

4. Conclusions

The ultrasonication-assisted exfoliation of graphite was performed using turmeric extract and cinnamon extract solutions along with curcumin solution which has been used in exfoliation before in a literature. Based on Raman spectroscopy results, all samples were truly exfoliated but at different degrees of exfoliation and all including the commercial graphene-based powder should be multilayer graphene-based product with many more than 5 layers. With the condition used in this work, the quality (reflected by I_{2D}/I_G and FWHM of 2D peak) of curcumin-treated sample was comparable to that of the commercial graphene-based powder but those of the extract-treated samples were still lower. However, the turmeric extract-treated sample had potential to be used in exfoliation of graphite with modification of the condition to obtain the quality of the exfoliated graphite comparable to that of the commercial graphene-based powder used in this study. This should be very interesting because turmeric is cheap while curcumin is expensive. A thorough investigation should be done in the future for more trials on extraction of the turmeric extract.

Even though the essence of checking the quality of the exfoliated graphite is strictly based on Raman spectroscopy, this work also showed the results from visible spectrophotometry. They were useful in measuring the absorbances of the suspensions of the exfoliated graphite. The higher values in the range greater than 650 nm corresponded to higher yields. However, there might be the effect of the color of the extracts on the measurement. The extract solutions should be used as control samples whose absorbances had to be subtracted from those of the exfoliated graphite in the extracts.

It is of interest to study the settling behaviors of the exfoliated graphite powder together with those of the precursor graphite and the commercial graphene-based powders in the future because this could be used as guidance to approximately quantify the yields of the exfoliated graphite products.

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