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Original Article

Liquid phase exfoliation of graphite using extracts from coffee grounds and spent coffee grounds

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Abstract

The green exfoliations of graphite by using extracts from Arabica and Robusta coffee grounds (CG), and cold-brew and hot-brew spent coffee ground (SCG) were studied. The reactions were performed in an ultrasonic bath (200 W, 40 kHz) at 55ºC for 2 h. The suspension was left at room temperature for 2 h and the exfoliated graphite was separated from the supernatant. The yield fluctuated when the amount of graphite was increased from 20 to 30 mg/mL, at which point the yield was around 2.8-4%. The total phenolic contents in the extracts were quite consistent with the yield. The Raman spectroscopy results showed that all extracts could exfoliate graphite to different degrees: Robusta extract gave fewer layers than Arabica, cold-brew and hot-brew SCGs. All, except for the hot-brew sample, showed quality comparable to a commercial graphene-based product. The SEM images showed thinner sheets and more wavering edges after exfoliation.

Keywords: graphene, spent coffee ground, phenolic compounds, Arabica, Robusta

1. Introduction

Coffee grounds (CGs) before brewing contain 12.4% cellulose, 39.1% hemicellulose (3.6% arabinose), 19.07% mannose, 43% galactose, 23.9% lignin, 17.44% protein, 2.29% fat, and 60.49% dietary fiber (Cruz, Morais, & Casal, 2015). There are many polyphenolic compounds in CGs, such as chlorogenic, caffein, ellagic, trans-ferulic, gallic, parahydroxybenzoic, p-coumaric, protocatechuic acid, tannic acids, catechin, epicatechin, rutin, and quercetin (Kovalicik, Obrucaa, & Marova, 2018). A study reported that 90% of the phenolic compounds can be extracted from coffee grounds by using ethanol (Zuorro & Lavecchia, 2012).

In 2015, coffee consumption worldwide reached about 9.1 billion kilograms and the production of instant coffee and coffee brewing generates about 6 million tons of spent coffee grounds (SCG) per year (Kovalicik *et al.*, 2018). SCGs have been applied as a supplement in animal feed, as soil fertilizers, and as antioxidants. In the last context, they were investigated as a source providing reducing and stabilizing agents in the synthesis of gold nanoparticles (Manyuan & Danwanichakul, 2021).

Graphene has gained interest in the research community because it has many beneficial properties including high elastic modulus, high thermal conductivity and high electrical conductivity. Therefore, graphene has been investigated increasingly for engineering and biomedical applications (Edwards & Coleman, 2013) leading to more research and development on graphene production. Graphene can be prepared with both bottom-up and top-down methods. The former includes chemical vapor deposition, in which the carbon atoms from the vapor source form a hexagonal structure, producing high-quality graphene but not in an industrial scale. The latter involves the separation of graphite layers to yield single-layer, few-layer and multilayer graphene, which are collectively called graphene-based materials. Yi and Chen (2014) applied liquid phase exfoliation to produce graphene in an industrial scale with a simple process of low cost and high production rate. However, the quality of produced graphene was relatively low, depending on the techniques and substances used to separate the layers.

The exfoliation of graphite can be achieved chemically or mechanically. In chemical exfoliation, graphite is turned to graphene oxide by reacting with many strong

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oxidizing agents that form polar functional groups on the surfaces, leading to a higher degree of the dispersion in water and resulting in a high yield. However, graphene oxide has a low electrical conductivity and the process takes a long time and involves a lot of highly toxic chemicals (Alzari *et al.*, 2011). In contrast, the mechanical exfoliation using ultrasound or shear force does not change the chemical structure of graphene so it still has high conductivity. It was found that sonication of graphite layers in ethanol could yield thinner graphene depending on the frequency of the applied ultrasonic waves (Telkhozhayeva *et al.*, 2021). In addition, Paton *et al.* (2014**)** reported that high-shear rotor–stator mixers yielded 200-700 nm thick graphene with 4-8 layers. Varrla *et al.* (2014) applied household blenders in exfoliation, resulting in a non-defective graphene with 6 layers and total thickness of 630 nm. However, liquid phase exfoliation usually gives a low yield, so the yield needs to be increased either by varying the process parameters or changing the liquid to manipulate the interactions with graphite layers.

Various organic solvents have been investigated in liquid phase exfoliation. The most widely used solvents are N-methyl-2- pyrrolidone (NMP) and N,N-dimethylformamide (DMF), but they are toxic. Moreover, their removal from graphene sheets is difficult due to the high boiling points. As a result, the residual solvents tend to reduce the properties of graphene. Zhang, Small, Pontius, & Kim (2005) adopted low-boiling point ethanol as an alternative to other solvents. However, the low surface energy of ethanol limits its ability to disperse graphene and sustain stability, resulting in a small amount of graphene.

Recently, natural substances have been in focus in the green preparation of nanomaterials. In the literature, curcumin in ethanol solution was used to exfoliate graphite (Navik, Gai, Wang, & Zhao, 2018). It was hypothesized that phenyl rings in curcumin structure had π-π interactions with graphitic structure and facilitated the dispersion of layers of graphite during ultrasonication. The hydrophobic effects of natural organic substances from cinnamon and red pepper, and curcumin, were thought to help exfoliate graphite (Koutsioukis, Florakis, Sakellis, & Georgakilas, 2022). In addition, extracts from butterfly pea flowers (Abdullah & Ismail, 2023), those from *Oxalis corniculata* (Puliyarai Keerai) (Perumal, Atchudan, Ramalingam, Nesakumar, & Edison, 2022) and gallnut extracts (Bu, Kushwaha, Goswami, & Kim, 2022), all containing polyphenols, were investigated in exfoliation of graphite in various conditions.

In this work, the liquid phase ultrasound-assisted exfoliation of graphite was performed using extracts from the spent coffee grounds because they still contain some phenolic compounds, which are expected to play a role in facilitating exfoliation. Therefore, they are of low cost and environmentally friendly. The SCGs used in this study were obtained after hot and cold brewing of coffee. The experiments were done with comparison of using Arabica and Robusta CG extracts.

2. Materials and Methods

2.1 Materials

Extra pure fine powder graphite with particle size less than 50 μm was purchased from MERCK. Ethanol with 96% purity was purchased from MERCK. The commercial graphene-based powder with apparent thickness of 6-8 nm and width of 5 μm was purchased from TCI, Ltd. The spent coffee grounds (SCGs) both from cold brewing and hot brewing were provided by Starbucks, Thammasat University branch, Pathumthani, Thailand. The Arabica and Robusta coffee grounds (CGs) were purchased from a local store in Bangkok, Thailand. Sodium carbonate was purchased from Kemaus Pty Ltd. Folin-Ciocalteus Phenol (FCP) reagent and gallic acid were supplied by Sisco Research Laboratories Pvt. Ltd., India. Sodium dodecyl sulfate (SDS) was purchased from Ajax Finechem.

2.2 Preparation of the extracts

The samples of SCGs were dried in an oven at 60ºC until constant weight before use. 5 g of cold-brew SCG, hot-brew SCG, Arabica CG and Robusta CG were each added to 75.5 mL of water. The extraction was done at 50ºC for 1 h using a water bath. The extracts were then filtered with a piece of filter paper to remove the remaining coffee grounds.

2.3 Determination of total phenolic content (TPC) of the extracts

Gallic acid is commonly chosen as a reference substance for checking TPC (Mussatto, Machado, Martins, & Teixeira, 2011). It is a trihydroxybenzoic acid and is classified as a phenolic acid. A calibration curve was constructed using gallic acid solutions at concentrations from 93.75 to 750 mg/L. 0.3 mL of the gallic acid solution was added to 0.3 mL of Folin–Ciocalteu reagent. This was followed by the additions of 100 μL of 75%w/v Na_2CO_3 solution and 10 mL of water. The mixtures were checked for their absorbances by UV-Vis spectrophotometry (model UV-5100, Metash) at 720 nm. After that, the TPC of the CGs and SCGs were measured by replacing gallic acid solutions with the extracts. The results are reported as gallic acid equivalents in mg per gram of solid.

2.4 Exfoliation of graphite with the CG and SCG extracts

200, 250 and 300 mg of graphite powder were each mixed with 10 mL of the CG and SCG extracts. The suspensions were placed in an ultrasonic bath (model GT-2227QTS, GT SONIC) at 40 kHz and 200 W and at about 55ºC for 2 h for exfoliation. After that, they were left standing at room temperature for 2 h. Subsequently, about 7 mL of the top part of the settling suspension was taken and centrifuged at 10,500 rpm for 30 min using a microcentrifuge (model CF-10, Wise spin) to obtain the sediment of the exfoliated graphite. It was washed twice, first with ethanol and later with water.

2.5 Yield and characterization of the exfoliated graphite samples

Each sample from Section 2.4 was dried in an oven at 60ºC until the weight was constant. The process yield can be calculated as the percentage of the mass of the dispersed sample obtained after 2 h sedimentation in the total mass of the precursor graphite used. The experiments were repeated

for each amount of graphite so that at least three samples were used to obtain the average yield.

Raman spectroscopy results of the dried samples along with the precursor graphite and the commercial graphene-based product were obtained using a dispersive Raman microscope (Model Senterra, Bruker Optics) 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 ASTM: E1693 and ASTM: E1840. In addition, the morphology of the exfoliated samples was investigated by Scanning Electron Microscopy (SEM, JSM-7800F) with an accelerating voltage of 15 kV. The magnification was adjusted to 10,000X.

It was observed in our prior work that settling behavior of the exfoliated graphite was different from the precursor graphite. Therefore, sedimentation was also done in this study. 0.052 g of dried sample was dispersed in 5 mL of 1% w/v sodium dodecyl sulfate solution. Each suspension was sampled into 1.5 mL microcentrifuge tube and centrifuged at 10,000 rpm. The absorbance of the supernatant was measured from 400-700 nm using the UV-Visible spectrometer at various centrifugation times up to 8 min.

3. Results and Discussion

3.1 The total phenolic content (TPC) of the extracts

Upon using the ratio of liquid volume to solid mass in the extraction equal to 15.1 mL/g solid, the TPCs of the extracts from Arabica and Robusta CGs and cold-brew and hot-brew SCGs were about 25, 24, 16 and 11 mg GAE/g solid, respectively, as shown in Figure 1. It has been reported that there was about 7% chlorogenic acid, a type of phenolic compound, in Arabica coffee and 5.5% in Robusta coffee and it could be transformed to ɣ-quinolactones during the roasting processes (Farah & Donangelo, 2006). In addition, the TPCs in Arabica CGs from various countries were reported to be around 5-7.5 mg GAE/g CG for both unroasted and roasted coffee beans (Muzykiewicz-Szymańska, Nowak, Wira, & Klimowicz, 2021). Therefore, TPCs are dependent on the type of coffee grounds and the extraction conditions.

As expected, the TPCs of the SCG extracts are lower than those of the CG extracts. The TPC in SCG can be lowered by even more than one half after hot brewing. Unlike cold brewing using cold water, hot brewing using hot water can extract more of the phenolic compounds into the coffee beverages, thereby lowering the amounts of substances left in the hot-brew SCG. It was reported that using 10-40 mL hot water/g SCG could yield the extracts with TPCs around 6-7.4 mg GAE/g SCG and it increased up to 18.2 mg GAE/g SCG when using 60% methanol solution (Mussatto, Machado, Martins, & Teixeira, 2011). In another work, when CGs and SCGs were extracted with water and 25% ethanol solution with the same ratio of 83 mL/g of solids, the TPCs were 26-27 mg GAE/g CG and 9-29 mg GAE/g SCG, respectively (Ramón-Gonçalves, Gómez-Mejía, Rosales-Conrado, León-González, & Madrid, 2019).

3.2 The yield of the exfoliation of graphite

Figure 2(b) shows the process yield, defined as the weight of dispersed solid obtained (shown in Figure 2(a)) from 100 g of the precursor graphite, in the experiments using the initial concentrations of graphite in the extracts set at 20, 25 and 30 mg/mL. It seems that for all extracts, the amount of graphite used need to be optimum; the yield decreases when the amount increases from 20 to 25 mg/mL and then increases when it increases from 25 to 30 mg/mL. The yields when using 20 mg/mL of the extract solutions of Arabica and Robusta CGs, and cold-brew and hot-brew SCGs are 1.60 ± 0.80%, 1.3 ± 0.38%, 2.35 ± 0.035% and 0.95 ± 0.245%, respectively. The standard deviations are quite large for all extracts, implying that the precursor graphite is not homogeneous while the extracts used were from the same lots.

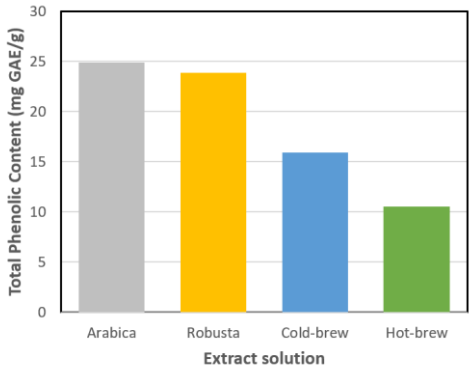

Figure 1. The total phenolic contents (TPC) of the extract solutions were measured as mg equivalents of gallic acid per gram of solid

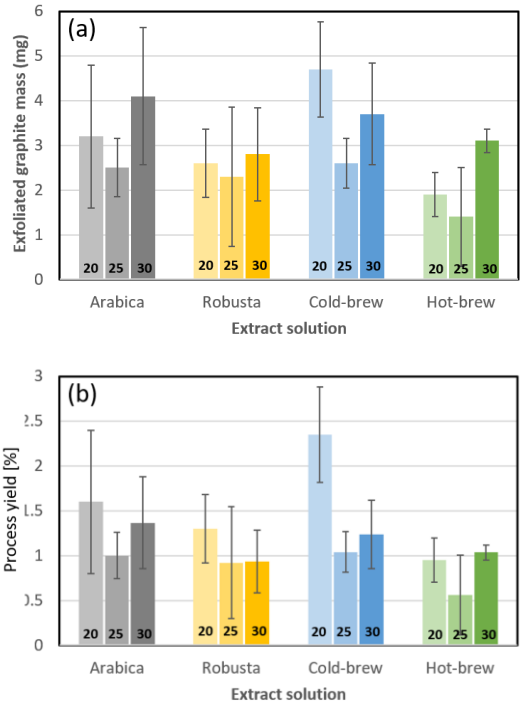

Figure 2. The masses of exfoliated graphite obtained and the process yields are displayed when varying the amount of graphite and using different extract solutions

These yields were greater than 0.20-0.35% when using the serum from skim natural rubber latex with leftover ammonia in an ultrasound-assisted exfoliation (Jirasitthanit & Danwanichakul, 2023) and some were greater than 2.0% when using curcumin in ethanol and ultrasound (Navik *et al.*, 2018) at the same ratio of initial graphite concentration. It was also reported that using the graphite concentration at 0.5 mg/mL increased the yield to 25% when using red pepper extract in an ultrasound-assisted exfoliation (Koutsioukis, Florakis, Sakellis, & Georgakilas, 2022).

Theoretically, in an ultrasonic system, the sound wave generates fine bubbles. The resulting pressure from the collapse of bubbles can move the layers of graphite whose inter-layer interactions are partly reduced by the attractive forces among phenolic compounds and graphite layers. Therefore, it can be hypothesized that the yield should depend on the power of the ultrasonic bath and time of sonication (Navik *et al*., 2018) as well as the TPC. Because the power and time were fixed in this experiment, the TPCs of the extract solution can be obviously compared. For all graphite concentrations, Arabica CG extract gives higher yields than Robusta CG extract, and cold-brew SCG extract gives higher yields than hot-brew SCG extract. These findings are consistent with the TPC results. However, when comparing the four extract solutions in all experiments, the cold-brew SCG extract solution seems to give the highest yields, implying that not only is the TPC significant, but the types of remaining phenolic compounds in the extracts should also play an important role. The extractability of each phenolic compound from the CGs and SCGs seems to be different. In other words, it depends on the temperature, the medium and the time of extraction. For instance, the extraction of CGs with water yielded some different types of phenolic compounds from those in SCGs with ethanol solution, because of different solubilities of the phenolic compounds (Ramon-Gonclaves *et al.*, 2019).

3.3 The morphology of the exfoliated graphite

The exfoliated samples using different extract solutions in the exfoliation, when fixing the graphite concentration at 20 mg/mL, were investigated for their morphologies. The micrographs are shown in Figure 3 along with those of the precursor graphite and the commercial graphene-based materials.

In Figure 3, the morphologies of the exfoliated graphite samples using different extract solutions (Figure 3(c), (d), (e), and (f)) look similar. They show a lot of small sheets distributed among large sheets which are different from that of the precursor graphite sample (Figure 3(a)) whose structure shows thick stacks of a lot of sheets before exfoliation. Upon exfoliation, the stacks look thinner and the edges of the exfoliated sheets look wavering as marked by the red circles. However, the sheets of the commercial graphene-based product (Figure 3(b)) are obviously larger. Each sheet seen in every micrograph is actually composed of many graphene layers so the exfoliated sheet might be called multi-layer graphene-based material.

3.4 Raman spectroscopy results

Results from Raman scattering can be used to indicate the number of layers and structural defects in graphitic materials. In the measurement, the laser photons excite the electron-hole pair causing 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 electrone-hole pair. Both inelastic and elastic scatterings yield characteristic peaks of graphitic structures including D-peak at ~1,350 cm^{-1}, G-peak at ~1,580 cm^{-1} and 2D-peak at ~2,700 cm^{-1}. Secondary peaks such as D'-peak at 1,620 cm^{-1} are also reported (Wu, Lin, Cong, Liu, & Tan, 2018). The results of the chosen exfoliated samples are shown in Figure 4 and Table 1.

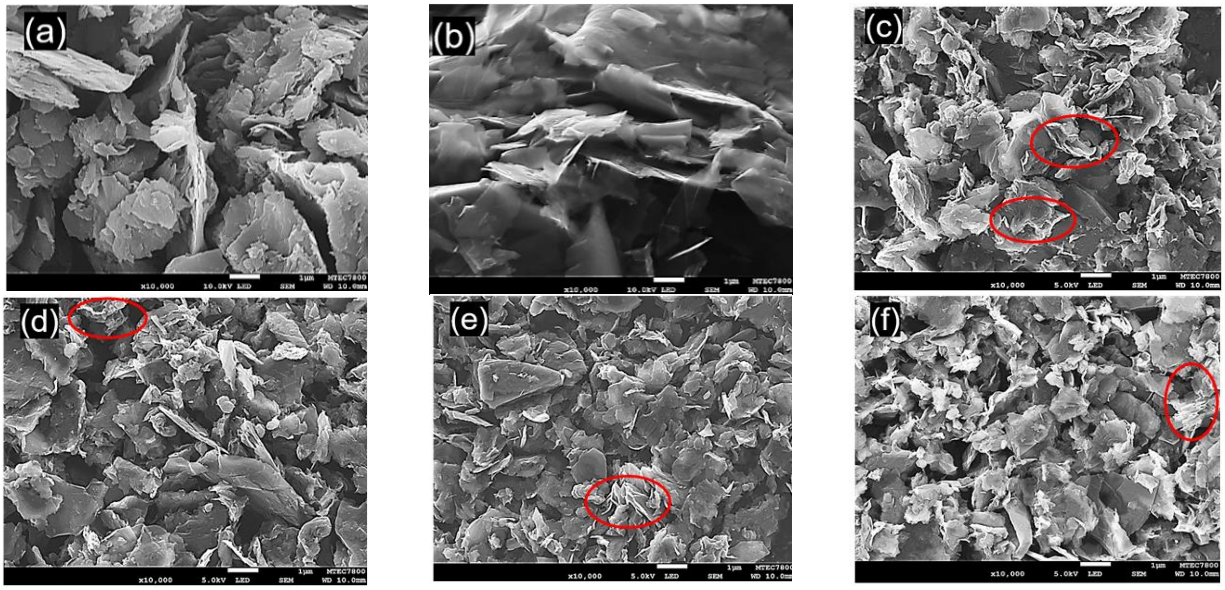

Figure 3. SEM micrographs of (a) the precursor graphite, (b) the commercial graphene-based product, and those of exfoliated graphite using extracts from (c) Arabica CG, (d) Robusta CG, (e) cold-brew SCG, and (f) hot-brew SCG

Table 1. The positions of D, G, and 2D peaks and their intensities together with the intensity ratios of I_D/I_G and I_{2D}/I_G are shown.

Sample	D (cm^{-1})	I_D	G (cm^{-1})	I_G	2D (cm^{-1})	I_{2D}	I_D/I_G	I_{2D}/I_G	FWHM of 2D
Graphite	1347.8	3265.2	1575.1	12580.1	2706.0	4404.7	0.26	0.35	75.56
Commercial graphene-based	1344.2	1817.0	1572.9	7739.2	2686.0	3967.6	0.23	0.51	69.39
Arabica	1348.0	10428.1	1577.0	23452.4	2694.5	13420.0	0.44	0.57	71.24
Robusta	1347.5	15310.4	1579.0	25913.5	2694.0	17132.3	0.59	0.66	71.45
Cold-brew	1345.5	10121.7	1572.0	21303.1	2688.0	12936.0	0.48	0.61	73.73
Hot-brew	1340.5	4048.8	1564.5	18152.0	2680.0	8806.4	0.22	0.49	73.73

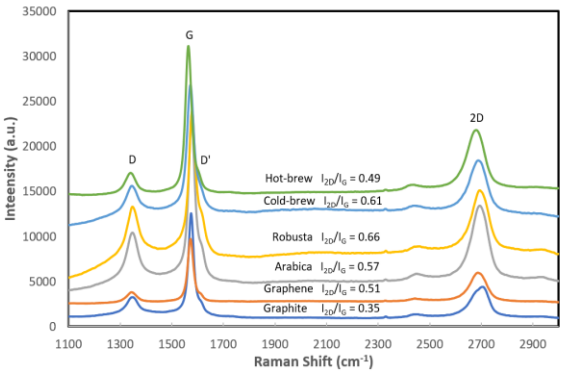

Figure 4. Raman spectroscopy results of the exfolaited graphite samples using different extract solutions, compared with those of the precursor graphite and the commercial graphene-based product.

D-peak appears from the scattering at the defects, which can be from the edge disorders, the vacancies, and the functionalization of carbon atoms. Additionally, D'-peak adjacent to G-peak also indicates the type of defect when the ratio $I_D/I_{D'}$ is considered. It was reported that this ratio could reach about 3.5 for edge disorders, 7 for vacancies, and 13 for sp^3-type defects (Wu *et al.*, 2018). Those of our samples (1.0-1.3) are all smaller than 3.5, implying that the defects are mainly from edge disorders, which are seen as wavering edges, as marked by red circles for examples seen in the SEM micrographs. They were subjected to the pressure forces from bubble collapses during the exfoliation.

In addition, G-peak represents C-C stretching mode in sp^2 hybridized carbon bonds, which can be caused by chemical doping, structural defects, elongation, and temperature. Therefore, the I_D/I_G ratio is commonly used to normalize the effect of spatial dimension when studying the edge defects (Wu *et al.*, 2018). It was indicated that for graphene with very few layers, when defect density increased, the value of I_D/I_G increased to a maximum of 3.5 and then decreased (Childres *et al.*, 2013). In our study, no chemical reactions occurred and the values of I_D/I_G for all samples were much less than 3.5, so the results are classified as low defect density. The values of I_D/I_G for all exfoliated samples using various extract solutions (0.22-0.59) are greater than 0.26 of the precursor graphite, indicating that edge disorders were generated during ultrasound-assisted exfoliation in all samples, except for the hot-brew sample. When compared with the reduced-graphene oxide sheets undergoing the oxidation-reduction method, whose I_D/I_G ratio was within 1.1-1.5 (Wu *et al.*, 2018), those of our samples are much lower. The I_D/I_G ratios reported in literature on using natural substances in exfoliation were less than 0.23 for the extracts of cinnamon and red pepper (Koutsioukis, Florakis, Sakellis, & Georgakilas, 2022), 0.25 for the extract of butterfly pea flower (Abdullah & Ismail, 2023), 0.855-0.857 for the extracts of *Oxalis corniculate* (Perumal, Atchudan, Ramalingam, Nesakumar, & Edison, 2022), and 0.220-0.391 for gallnut extracts (Bu, Kushwaha, Goswami, & Kim, 2022).

2D-peak appears because of the scattering of the electrons by two phonons before it recombines with the hole. If there are more defects and electrons are mediated with the defects, fewer electrons will recombine with the holes, resulting in a lower 2D-peak (Li, Deng, Kinloch, & Young, 2023). So, this ratio is closely related to the number of graphene layers. Theoretically, an increase in I_{2D}/I_G reflects fewer layers, but it is not enough to determine the exact number. It was stated that value of I_{2D}/I_G increased upon exfoliation from 0.35 for graphite to 0.50 for multilayer graphene, and to 0.69 for bilayer graphene (Li *et al.*, 2023). Table 1 confirms that the exfoliations of graphite in the ultrasonic bath using all extract solutions occurred because I_{2D}/I_G (0.49-0.66) is greater than 0.35 for graphite. The I_{2D}/I_G of Robusta sample is the greatest, followed by cold-brew, Arabica, and hot-brew ones in the same order as already seen for the I_D/I_G. All, except for the hot-brew sample, are greater than that of the commercial graphene-based product, implying that the quality of the exfoliated graphite is comparable to the commercial one. Figure 4 and Table 1 also show that the positions of the 2D-peaks of all the exfoliated graphite samples shift to lower wavenumbers than that of the precursor graphite, closer to that of the commercial graphene-based product.

Furthermore, the full width at half maximum (FWHM) of 2D-peak can also be used to relate to the number of layers. Because 2D-peak resulted from constructive interferences of all peaks generated by each layer of graphitic materials, the FWHM of 2D-peak was wider for a sample with more layers (Childres *et al.*, 2013). In this study, all samples have narrower FWHM (71.24-73.73 cm^{-1}) than 75.56 of the precursor graphite. The Arabica and Robusta samples (71.24 and 71.45 cm^{-1}) have FWHMs closest to 69.39 for the commercial graphene-based sample. These are greater than 70 cm^{-1} reported when using the extracts of cinnamon and red pepper (Koutsioukis, Florakis, Sakellis, & Georgakilas, 2022). It was reported that the FWHM of 2D-peak for five-layer graphene was between 65 and 67.5 cm^{-1} (Li et *al.*, 2023). Therefore, all the obtained samples, as well as the commercial graphene-based product, should be composed of more than five layers.

3.5 Sedimentation of the exfoliated graphite

Upon exfoliation the number of layers decreased, resulting in a powder with a lighter weight. The sedimentation was performed to justify this assumption. The results at the initial time and at 8 min are shown in Figure 5(a) and (b), respectively. To represent how the absorbances change over settling time, the average absorbances were determined over the wavelengths from 400 to 700 nm and plotted against the settling time as shown in Figure 5(c).

Figure 5(a) shows that at the beginning, due to the good dispersion, the absorbances of the Arabica, Robusta, cold-brew samples are about the same as the commercial graphene-based product. Those of the hot-brew sample are lower, and those of the precursor graphite were the lowest, because some powders were seen settling down immediately at the beginning. The centrifuge-expedited settling was seen pronounced over time and the absorbances are evidently seen decreased at 8 min in Figure 5(b). All the precursor graphite had settled since the first minute. The overall settling behavior can be seen in Figure 5(c). The settling behaviors of all samples, except for the hot-brew sample, are seen similar to that of the commercial graphene-based product. So, the degree of dispersion of the exfoliated sample can be compared. It is the best for Robusta sample, followed by cold-brew, Arabica and hot-brew samples. This trend is consistent with the I_{2D}/I_G ratio in Table 1. This correlation implies that the exfoliated graphite sample with fewer layers can disperse better in the SDS solution. Therefore, this macroscopic property can be used to relate to the microscopic structures of the exfoliated graphite.

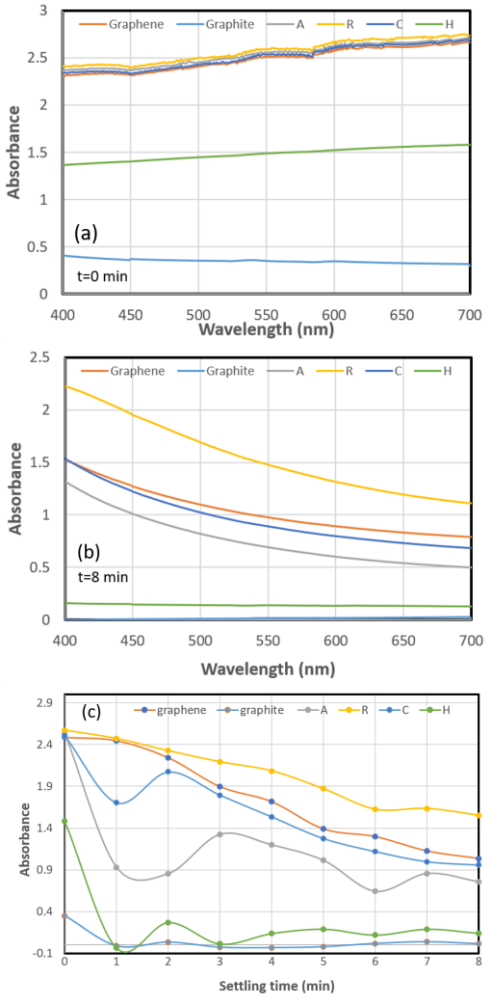

Figure 5. The absorbances of the supernatants of the samples using Arabica CG (A), Robusta CG (R), cold brew SCG (C), and hot brew SCG (H), compared with those of graphite and commercial graphene material samples at (a) the initial time, and (b) after 8 min of centrifugation. The changes in average absorbances along the centrifugation time up to 8 min are shown in (c).

4. Conclusions

Liquid phase exfoliation of graphite was performed in this research by using extracts from Arabica and Robusta CGs, and those from cold-brew and hot-brew SCGs. It was seen that graphite could be exfoliated using any of these extracts in the ultrasonic bath at 55ºC for 2 h. The phenolic compounds in these extracts were believed to assist the exfoliation because it was found that the yield when using Arabica CG extract was greater than with Robusta CG extract, and that when using cold-brew SCG extract was greater than with hot-brew SCG extract, consistent with their TPCs. However, Raman spectroscopy showed that the I_{2D}/I_G ratio of the Robusta sample was the highest implying the fewest graphene layers; this was followed in rank order by Arabica, cold-brew and hot-brew SCG samples. These, except for the hot-brew sample, were either comparable to or greater than the commercial graphene-based product. Moreover, the settling behavior in the SDS solution was found consistent with the I_{2D}/I_G ratios, indicating that with fewer layers of graphene, the exfoliated graphite could be dispersed better in the solution. This experiment could be helpful in differentiating the exfoliated graphite from the precursor graphite. According to the yield and I_{2D}/I_G ratio, cold brew SCG extract showed potential for use as the liquid assisting exfoliation, in this study. In addition, because cold-brew SCG is waste from cold brewing of coffee grounds, and this practice is gaining popularity among coffee drinkers, it could be used as a sustainable source of phenolic compounds in liquid phase exfoliation of graphite. To study the effects of phenolic compounds in exfoliation, FTIR results can be helpful to directly confirm the adsorption of those molecules on exfoliated graphite layers, which can lower the inter-layer interactions during the exfoliation and can stabilize the layers afterwards.

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