Research Paper

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Facile and Environmentally Friendly Synthesis of Low-defect Few-Layers Graphene (FLG) Using Combined Shear Exfoliation Methods

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**Abstract**

This study presents an environmentally friendly and scalable method for synthesizing high-quality few-layer graphene (FLG) through a combination of turbulence-assisted shear exfoliation (TASE) and high shear exfoliation (HSE) techniques. By systematically varying the high-shear mixer (HSM) speed (3000–5000 rpm) and processing time (1–3 hours), we precisely controlled key material attributes, including the number of graphene layers, crystallinity, lateral size, and defect density. Optimal conditions (5000 rpm, 3 hours) resulted in FLG with ~2–3 layers, confirmed by a symmetric 2D peak with a full-width at half-maximum (FWHM) of ~35 cm⁻¹ and a high I2D/IG ratio (~0.6), indicating excellent structural integrity. The ID/IG ratio (~0.1) further verified the presence of minimal defects, predominantly edge vacancies rather than oxidative disruptions. Raman imaging revealed a dominance of zigzag edge chirality, while TEM and PSA analyses demonstrated control over lateral size (~396.5 nm) and particle uniformity. The application of household dishwashing liquid as a green surfactant innovatively enabled selective and pure exfoliation. This work highlights how precise modulation of shear parameters can directly influence graphene quality, paving the way for sustainable large-scale production of low-defect FLG.

**Keywords:** *few-layer graphene (FLG); high shear exfoliation (HSE); mixer rotation; process duration; turbulence-assisted shear exfoliation (TASE).*

**Introduction**

In recent decades, significant advancements have been made in the field of nanotechnology, especially in the area of carbon-based nanomaterials, such as graphene. This advancement is closely linked to graphene's remarkable properties, which include a tensile strength of up to 15 GPa, a Young’s modulus of 1 ± 0.1 TPa, electron mobility reaching 35 × 104 cm²/Vs, high thermal conductivity, ranging from 3000 to 5000 W/mK at room temperature, as well as electrical conductivity approximately ~6×10⁸ S·m⁻¹, and a large specific surface area of 2600 m²/g (Choudhary et al., 2014; Murdiya et al., 2022; Qiao et al., 2019; Shahnaz et al., 2024; Zhen & Zhu, 2018). Structurally, graphene consists of a hexagonal honeycomb lattice with a sp² hybridization configuration, which is characterized by sigma (σ) and pi (π) bonds (Rao et al., 2009). The fewer layers and defects present in graphene, the higher its quality, closely approaching the ideal properties of single-layer graphene (Allen et al., 2010; Bazylewski & Fanchini, 2019; Yi & Shen, 2015). Potential applications of graphene include electronic devices, energy storage systems, nanocomposites to speed charging in lithium-ion batteries, filler agents for polymer composites, photovoltaic materials, membranes for gas and water separation and purification processes, and coatings characteristics (Agudosi et al., 2020; Amri et al., 2021; Godoy et al., 2019; Shahnaz et al., 2024). Recent studies have demonstrated superior electrical properties, mechanical strength, and chemical stability of few-layer graphene (FLG) compared to multilayer graphene by applying FLG as a lithium battery electrode, showing higher lithium storage capacity than that in multilayer graphene (Tong et al., 2011). Several works report that the presence of graphene significantly increases the surface area, adsorption capacity, and also the photocatalytic process (TiO2) (Heltina et al., 2023). Graphene can be synthesized through various methods, including mechanical cleavage, epitaxial growth, chemical vapor deposition (CVD), reduction of graphene oxide (rGO), and liquid-phase exfoliation (Amri et al., 2021; Hou et al., 2018; Wang et al., 2018). Over the years, several top-down methods have been developed, including Liquid-phase exfoliation (Amri et al., 2021; Varrla et al., 2014), shear exfoliation (Paton et al., 2014; Zhang et al., 2018), Taylor-Couette, and thin-film reactors (Chen & Xue, 2014; Stafford et al., 2018), intermediate-assisted grinding exfoliation (Zhang et al., 2020), oxidation-reduction reactions (Luong et al., 2020), ultrasonic exfoliation (Kaur et al., 2022; Tyurnina et al., 2020), electrochemical exfoliation (Regasa & Nyokong, 2024), molten salt methods (Atakoohi et al., 2024), most of which can produce low-defect graphene without oxidation. Liquid-phase exfoliation offers advantages in terms of mass production of graphene, simplicity, and low cost as a top-down approach (Ma & Shen, 2020).

Mechanical liquid exfoliation (MLE) is a common approach to preparing low-defect graphene, utilizing graphite powder as the primary raw material and specific solvents and surfactants as working fluids (Allen et al., 2010; Yi & Shen, 2015). Graphene was produced from graphite by Varrla, et al., (2014) using a purely mechanical TASE method in a domestic kitchen blender with surfactant-enriched water as the working fluid. This paper also successfully produces a full-thickness and large-area defect-free graphene up to an average lateral size of around 630 nm with approximately six layers (Varrla et al., 2014). Nonetheless, the number of graphene layers created is still quite substantial. Paton, et al., (2014) also used a high-shear rotor-stator mixer for the exfoliation of graphite powder into graphene after adding further dispersion agents, including N-methyl-2-pyrrolidone (NMP) and two surfactants (NaC & polyvinyl alcohol). The Graphene obtained was 4 to 8 layers thick and had a lateral size of 200–700 nm (Paton et al., 2014). The solvent and surfactants employed are highly expensive, which in turn renders the process non-economic and also produces a large number of graphene layers (Dimov et al., 2018).

Graphene nanosheets were prepared from graphite via a liquid shear exfoliation route using a rotor-stator mixer, as described by Liang, et al., (2021). An organic salt was used as a surfactant in conjunction with N-methyl-2-pyrrolidone (NMP) as a working fluid. At higher rotor rates and extended exfoliation times, this leads to results in thin graphene nanosheets. Unfortunately, the problem of residual solvent was the most challenging to resolve in the overall elimination process with graphene material, and it reflected negatively on the quality of graphene nanosheets (Liang et al., 2021). Very few-layer graphene (VFLG) was synthesized using a two-step sheared exfoliation process by Amri, et al., (2021). Blending started with a kitchen blender and progressed to a high-shear mixer (HSM). Aqueous solution of sodium lauryl sulfate (SLS)-anionic surfactant was employed as the working fluid in this investigation. The morphology of the produced graphene was confirmed to be 1 to 3 layers with an average lateral size of ~375.4 nm (Amri et al., 2021). In contrast, both operational parameters and factors that influence the quality of graphene produced in HSM are still inadequately understood.

Graphene was synthesised by a method TASE in an in-kitchen blending rotating-blade mixer, followed by HSE in a high-shear mixer. We examined the effects of HSM rotor speed (rpm) and HSE processing time on the quality attributes of graphene. The experimental scale of graphene produced mainly consists, as expected, of 2-3 layer graphene with relatively few defects. The method promises to be a powerful technique for producing high-quality, low-cost, and eco-friendly few-layer graphene (FLG).

**Materials and Methods**

**Preparation of Graphene**

Both the TASE and HSE procedures were used to synthesize graphene. TASE was performed using a rotating-blade mixer (kitchen blender) and followed by a high-shear mixing process with an L5M high-shear mixer. Afterward, the extrudates were processed as described previously (Amri et al., 2021). The 18% anionic surfactant (sodium lauryl sulfate, SLS) aqueous solution was used as the stabilizer and process fluid. The effects of the rotation speed of HSM (3000, 4000, and 5000 rpm) as well as the processing time for HSE (1, 2, and 3 hours) were investigated during the experiments. The homogenized solution was kept for 24 hours before being decanted from the precipitate by Varrla, et al., (2014). The solution was then dried using a hair dryer.

**Characterization**

The characterization of synthesized raw graphite and graphene was performed using Raman spectroscopy (RS), Raman imaging (RI), particle size analysis (PSA), transmission electron microscopy (TEM), and Fourier-transform infrared spectroscopy (FTIR) techniques to determine their properties. Raman studies were performed using a WITec Alpha 300R Raman spectrometer to acquire images, exciting molecular vibrations at a wavelength of 532 nm. Each sample was measured at 10 different location landmarks. OriginPro 8.5.1.315 was utilized to remove or reduce any noise in test results. Analyses of the band peak in the 2D spectrum were performed using Lorentzian fitting with these tools. The morphology and size of the graphene were identified using a TEM HT7700 at an acceleration voltage of 120 kV, with visual data processing conducted in ImageJ version 1.47. Particle size analysis (PSA) was performed to measure particle size distribution and median diameter using the HORIBA Laser Scattering Particle Size Distribution Analyzer LA-960. The FTIR analysis confirmed that no oxidation defects were introduced to the graphene using a Perkin Elmer Spectrum IR Version 10.6.

**Result**

**Raman Spectroscopy (RS) Analysis**

Figure 1 presents the Raman spectra of the initial graphite (particle size of 5 µm) and the graphene synthesized via the combined TASE-HSE process, with varying HSE rotation speeds (3000-5000 rpm) and HSE processing times of (a) 1 hour, (b) 2 hours, and (c) 3 hours. Figure 1 (a-b) illustrates the TASE-HSE samples processed at an HSE rotation speed of 3000 rpm for 1-2 hours, which exhibited a branched 2D peak and an additional broad peak around ~1750 cm⁻¹ with relatively high intensity.

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1. Raman spectra of the initial graphite and TASE-HSE graphene at varying HSE processing times: (a) 1 hour, (b) 2 hours, and (c) 3 hours.

Table 1 presents the ID/IG and I2D/IG ratio values for graphene samples produced via the TASE-HSE process. For TASE-HSE samples, the ID/IG ratio ranged from 0.1-0.6, which is consistent with the presence of few-layer graphene (FLG). Table 1 also shows that as the HSE rotation speed and processing time increased, the ID/IG ratio decreased.

1. Comparison of the ID/IG and I2D/IG ratios.

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| --- | --- | --- | --- | --- |
| **HSE Process Time (Hours)** | **HSM Rotation Speed**  **(rpm)** | **Sample Number** | **ID/IG** | **I2D/IG** |
| 1 | 3000 | Sample 1 | 0.6 | 0.5 |
| 4000 | Sample 2 | 0.3 | 0.6 |
| 5000 | Sample 3 | 0.4 | 0.6 |
| 2 | 3000 | Sample 4 | 0.2 | 0.6 |
| 4000 | Sample 5 | 0.6 | 0.5 |
| 5000 | Sample 6 | 0.1 | 0.6 |
| 3 | 3000 | Sample 7 | 0.5 | 0.5 |
| 4000 | Sample 8 | 0.1 | 0.5 |
| 5000 | Sample 9 | 0.1 | 0.6 |

Further analysis, including Lorentzian peak fitting for the 2D band in the Raman spectrum, is illustrated in Figure 2. Figure 2 illustrates the application of Lorentzian peak fitting to the 2D band in the Raman spectrum. This was conducted using a 532 nm laser excitation and analyzed with OriginPro 18.1.2 software on FLG and graphite samples. The most favorable result was observed in the TASE-HSE (3 hours, 5000 rpm) sample, which exhibited a relatively symmetric Lorentzian component at 2716.7 cm⁻¹ with a full-width at half-maximum (FWHM) of ~35 cm⁻¹, confirming the presence of FLG predominantly consisting of 2-3 layers.

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| --- | --- |
| **Graphite** | **TASE-HSE (1h, 5000 rpm)** |
|  |  |
| **TASE-HSE (1h, 4000 rpm)** | **TASE-HSE (1h, 3000 rpm)** |
|  |  |

1. The application of Lorentzian peak fitting to the 2D band in a Raman spectrum using a 532 nm laser excitation source.

|  |  |
| --- | --- |
| **TASE-HSE (2h, 5000 rpm)** | **TASE-HSE (2h, 4000 rpm)** |
|  |  |
| **TASE-HSE (2h, 3000 rpm)** | **TASE-HSE (3h, 5000 rpm)** |
|  |  |
| **TASE-HSE (3h, 4000 rpm)** | **TASE-HSE (3h, 3000 rpm)** |
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**Figure 2 Continued.** The application of Lorentzian peak fitting to the 2D band in a Raman spectrum using a 532 nm laser excitation source.

**Raman Imaging (RI) Analysis**

Figure 3 presents the micro confocal hyperspectral 3D imaging analysis results for raw graphite (a) and FLG samples produced via the TASE-HSE process with HSE rotation speeds of 5000 rpm and HSE processing times of 3 hours (b). Figure 3(a) depicts the 3D confocal imaging of the initial graphite sample, which exhibited a pure crystalline form with a 97.4% graphite composition. Figure 3(b) displays the 3D confocal imaging of the FLG TASE-HSE sample (3 hours, 5000 rpm), wherein the D band intensity manifested a hexagonal yellow pattern forming a 90° and 150° angle. This pattern exhibited two distinct edge types: armchair and zigzag.

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| A close up of a rock  AI-generated content may be incorrect. | A screenshot of a cell phone  AI-generated content may be incorrect. |

1. 3D hyperspectral Raman imaging confocal data for (a) graphite and (b) FLG TASE-HSE (3 hours, 5000 rpm).

**Transmission Electron Microscopy (TEM) Analysis**

Figure 4 presents TEM morphological analysis of the following samples: (a) graphite, (b) FLG TASE-HSE (3 hours, 5000 rpm), and (c) FLG TASE-HSE (1 hour, 3000 rpm). The graphite sample was observed to consist of large stacks of multilayer graphene with relatively large particle sizes, as shown in Figure 4(a). Figure 4(b) depicts the FLG TASE-HSE (3 hours, 5000 rpm), which exhibited the presence of two graphene layers (2 LG). As shown in Figure 4(c), the FLG TASE-HSE (1 hour, 3000 rpm) was primarily composed of five layers of graphene (5 LG).



1. TEM images of (a) graphite; (b) FLG TASE-HSE (3 hours, 5000 rpm); (c) FLG TASE-HSE (1 hour, 3000 rpm).

Figure 5 presents the histogram of flake length for (a) FLG TASE-HSE (3 hours, 5000 rpm) and (b) FLG TASE-HSE (1 hour, 3000 rpm). The average flake lengths for FLG TASE-HSE (3 hours, 5000 rpm) and FLG TASE-HSE (1 hour, 3000 rpm) were 139.7 nm and 396.5 nm, respectively.

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1. Flake length histogram for (a) FLG TASE-HSE (3 hours, 5000 rpm) and (b) FLG TASE-HSE (1 hour, 3000 rpm).

**Particle Size Analyzer (PSA) Analysis**

Figure 6 illustrates the particle size pattern graphs for raw graphite (a), FLG samples produced via the TASE process (b), and FLG samples produced via the TASE-HSE process (3 hours, 5000 rpm) (c). The average particle sizes in Figure 6(a), (b), and (c) were reported to be 6.1 μm, 5.9 μm, and 4 μm, respectively.

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1. Particle size pattern for (a) graphite; (b) FLG TASE; and (c) FLG TASE-HSE (3 hours, 5000 rpm).

**Fourier Transform Infrared Spectroscopy (FTIR) Analysis**

Figure 7 presents the FTIR spectra of the FLG TASE-HSE (2 hours, 5000 rpm), FLG TASE-HSE (2 hours, 3000 rpm), and graphite samples. This technique helps identify the chemical bonds present in the samples. It is commonly used to assess material composition and detect functional groups. The spectra of the different samples appear quite similar, indicating that the chemical structure has not undergone significant changes.



1. FTIR spectra of graphite and FLG TASE-HSE.

**Discussion**

As illustrated in Figure 1, the graphite spectrum exhibited the presence of the G band peak (1581.2 cm⁻¹) and the 2D band peak (2718.9 cm⁻¹), with either no D band peak or a negligible D band peak, which are characteristic fingerprints of graphite (Zhang et al., 2018). The absence (or diminution) of the D band peak indicates that most of the graphene precursor was defect-free. The typical Raman spectra of graphene are composed of the D band (∼1350 cm⁻¹), G band (∼1580 cm⁻¹), and 2D band (∼2720cm⁻¹) peaks. The 2D band of the TASE-HSE procedure shifts slightly to a lower wavenumber compared to that of the graphite. This is an initial step in the exfoliation of graphite to graphene (Malard et al., 2009).

Both peaks appeared on the left side of the G-band in the form of a strong D band peak and a weak D′ band peak (~1620 cm⁻¹), indicating that graphene had some defects (Biroju & Giri, 2014). Additionally, mechanical dispersion is supported by the count ratio for the band intensity comparison of D and G ports, which decreased with higher mixing rotation speeds. This is consistent with the longer processing time in HSE, which may indicate a lower incidence of defects. It has been demonstrated that the HSE processing to some extent defects in order to reduce graphite size to smaller lateral-size particles and few-layered carbon nanosheets via turbulence, collisions, shredding mechanisms as aforementioned studies (Amri et al., 2021; Paton et al., 2014; Varrla et al., 2014; Yi & Shen, 2014). The defects or scratch propagation of the graphite degrade the graphene sheets, leading to splitting between two opposing graphenes with a somewhat planar crystalline structure (Ganesan et al., 2016). In the HSE process, graphene does not fragment into smaller domains, as observed in rotation at lower rpm (3000–4000 rpm); thus, defect vacancies dominate, supported by high D band intensities. However, at the higher HSE speeds (5000 rpm) and longer processing durations (3 hours), defect vacancies are reduced or even eliminated when graphene splits completely into small fractions (Ganesan et al., 2016).

This peak originates from the tangential out-of-plane optical phonon (oTO) mode, which describes the vibrational motion of carbon atoms perpendicular to the graphene plane (Denison, 2016; Sato et al., 2011). The existence of this peak reflects the van der Waals attraction between carbon atoms in graphene layers that are immediately above and below each other. Even in the case of graphene, modifications to the Raman spectrum and appearance of peaks due to defects in the graphene structure will shift the oTO peak location (Ferralis, 2010). Shift of the oTO peak due to changes in carbon-carbon bonds related to defect vacancies (Leong et al., 2018). The fact that oxidation-induced impact induces vacancies in these defects, causing the oTO peak in the Raman spectrum to disappear, becomes more pronounced with larger vacancies and a higher density of them (Leong et al., 2018). The tangential oTO optical phonon peak is a second-order overtone mode that only appears in few-layer graphene, but not in single-layer or bilayer graphene (Sato et al., 2011). Figure 1 (a-b) shows that the oTO peak's intensity typically decreases as the rotation speed increases and the HSE processing time increases. Figure 1 (c) shows that this peak is no longer present at higher HSE rotation speeds (5000 rpm) and longer HSE processing times (3 hours). This means that the number of graphene layers has decreased, and the number of defects has also decreased.

Figure 1 also illustrated that the G band peak has been displaced, indicating an exfoliation between the layers in graphene processing (Malard et al., 2009). The G peak is associated with the E₂g optical phonon at the Brillouin zone centre, which is a result of stretching sp² carbon bonds within the rings and chains of an ideal graphene structure (Kumar et al., 2019). The position of the G band provides insight into the number of layers in graphene. The number of layers (n) can be calculated quantitatively using Equation 1 (Wall, 2011).

(1)

In this context, *ωG* represents the Raman shift for the G band peak, with *n* denoting the number of graphene layers. According to Equation 1, the *n* value for TASE-HSE (3 hours, 5000 rpm) is approximately ~2.1, which corresponds to bilayer graphene. In contrast, the TASE-HSE graphene sample processed at the lowest rotation speed and shortest duration yielded an *n* value of ~4, indicating that it was composed of few-layer graphene. As the processing time and rotation speed increased, the ωG value also increased, indicating a reduction in the number of layers (*n*). However, the application of Equation 1 for calculating graphene layer numbers is only valid for wavenumbers above 1581.6 cm⁻¹ (Wall, 2011). An increase in layer thickness results in a shift of the G band position to a lower Raman shift region, reflecting the absorption of laser photon energy reflected by the plane and the subsequent transfer of valence electrons (Wall, 2011).

The graphene defect can be quantified by examining the intensity ratio of the D band to the G band (ID/IG) (Bhaskaram & Govindaraj, 2018). The ID/IG ratio is an index of it, and from the value calculated in Table 1, we show that few-layer graphene (FLG) may be present.  Zhang, et al., (2018) highlighted this, as FLG usually possesses a 0.1–0.8 ID/IG ratio. A decrease in the ratio ID/IG means a thinner thickness of graphene layers (Jorio et al., 2010). The relations between processing duration, rotational velocity, and the I2D/IG ratio are shown in Table 1. With the increase of rotation speed and processing time, the I2D/IG ratio increased accordingly. An increased I2D/IG ratio of the sample indicates an improved graphene structure with fewer layers and less defect density. Together with the corresponding decrease in the G-band intensity, this I2D/IG increase results in a broadening of the 2D peak profile, as observed experimentally by Vyshkvorkina, et al., (2021). Material with a high I2D/IG ratio of around ~2, of around 2, sharp and symmetric 2D band, monolayer graphene generally has low defects (Moon et al., 2010). Furthermore, the 2D band peak also suggests the presence of graphene layers. The 2D band (second-order overtone of the D band) arises from a two-phonon process due to lattice vibration, which was described by Wall (Wall, 2011). Another explanation for the increase in intensity with layer thickness is that, as the 2D band red shifts to a lower wavelength (Moon et al., 2010). Broader and more asymmetric 2D band peaks correspond to increased graphene layers (Wall, 2011). The 2D band appears less intense in each spectrum than the G band, indicating that the samples are predominantly few-layer graphene (FLG) (Malard et al., 2009).

As shown in Figure 2, the graphite sample displayed the presence of multiple Lorentzian components at the 2D band peak (Amri et al., 2021). The 2D band peak is associated with double resonance transitions, which result in the production of two phonons with opposite momentum (Jibrael & Mohammed, 2016). For single-layer, bilayer, and few-layer graphene, the 2D band has an FWHM of more than 25 cm⁻¹ but less than 45 cm⁻¹, while multilayer graphene exhibits a broader 2D band with an FWHM of approximately 45-60 cm⁻¹ (Malard et al., 2009). The exfoliation of graphene was significantly enhanced by high shear forces localized around the rotor-stator regions, generated by flow velocities at 3000, 4000, and 5000 rpm, resulting in Reynolds numbers of approximately ~5.1×10⁴, ~6.8×10⁴, and ~8.5×10⁴, respectively. Consequently, inertial forces exerted a dominant influence over viscous forces, thereby promoting collisions between graphite particles (Yi & Shen, 2014, 2016). This finding aligns with the work by Liang, et al., (2021), who demonstrated that the exfoliation of graphene can be accelerated by high rotation speeds, resulting in the formation of thinner graphene layers (Liang et al., 2021). Prolonged exfoliation durations result in a higher number of graphite layers delaminating into thinner multilayer graphene, leading to a lower thickness and a higher concentration of graphene particles in dispersion, which ultimately results in an increased concentration of final graphene (Liang et al., 2021; Phiri et al., 2017).

From Figure 3(a), it appears that purity is associated with higher quality graphene production (Matsumoto et al., 2015). Figure 3(b) displays pattern of graphene exhibited two distinct edge types: armchair and zigzag. When the angles between adjacent sides are 30°, 90°, and 150°, the sides will exhibit different chiralities, as indicated by the presence of zigzag and armchair edges (You et al., 2008). As posited by You, et al., (2018) and Malard, et al., (2009), an increased presence of zigzag structures is indicative of higher graphene quality (low-defect), whereas an abundance of armchair structures indicates a higher incidence of defects within the graphene sample (Malard et al., 2009; You et al., 2008). The D band intensity is significantly stronger at armchair edges than zig-zag edges, suggesting that the armchair edge is the primary contributor to the D band peak in the Raman spectrum (Amri et al., 2021; Malard et al., 2009; You et al., 2008). As illustrated in Figure 3(b), the edges of the resulting FLG were predominantly zigzag structures, with minimal presence of armchair edges. This finding was in agreement with the Raman spectrum analysis of the FLG TASE-HSE sample (for 3 hours, 5000 rpm), which showed very little D band peak intensity. In addition, a white film with high brightness was found around the FLG in Fig. 3(b), showing the intensity of the G-band peak that is higher than the D-band, and Figure 3 points to a massive exfoliation of the graphite to small FLG and an overall pronounced growth from the crystal zigzag faces (Li et al., 2020).

Transmission electron microscopy (TEM) images of the individual layers of FLG. From the bright to dark contrast colours in Fig. 4(b-c) refer to a laminated form of FLG (Amri et al., 2021; Gürünlü et al., 2020; Paton et al., 2014). The following exfoliation step reduced the number of layers, and hence particle thickness and sizes (Amri et al., 2021; Varrla et al., 2014). This is in agreement with the results of higher rotation speeds and longer processing times, which was reported by Gai, et al., (2018) to produce fewer graphene layers (FLG) (Gai et al., 2018)**.** The longer graphene flakes were discovered as the number of graphene layers was further reduced. This study is in agreement with prior work that has shown TASE-HSE results in longer, thicker graphene sheets (Amri et al., 2021). In our studies, the FLG produced has a flake length of <1 μm on average, which is generally similar to what was reported for liquid-phase exfoliation (Amri et al., 2021; Turner et al., 2019).

Figure 6 presents the percentage distribution of particle sizes as a function of q, represented by a differential curve (bell curve). Likewise, the full under unit curve demonstrates the number of particles under a specified size. The *x*-axis (diameter, μm) represents the diameter of the particles in micrometers (μm) (Wang et al., 2022). Figure 6(a) demonstrates a more expansive particle size distribution, indicating a significant variation in graphite particle sizes (non-uniform), with an average particle size of 6.1 μm. Figure 6(b) depicts a narrower particle size distribution for FLG TASE than that observed for raw graphite, suggesting a greater degree of uniformity in particle size, with an average diameter of 5.9 μm. In Figure 6(c), the narrow and sharp particle size distribution suggests that FLG TASE-HSE particles are more uniform, with an average size of 4 μm (Bertran et al., 2020; Burlakov & Goriely, 2017).

This can be rationalized by the test results in Figure 6, which demonstrate that both the rotating blade mixer and the high shear mixer have a limited ability to cut off the graphene frontally (and hence laterally), as they only slightly reduce its size compared to the initial graphite size. These approaches are quicker for exfoliating interlayer graphene by cracking the van der Waals bonds between layers (Amri et al., 2021; Varrla et al., 2014). However, FLG TASE-HSE has a smaller particle size than FLG-TASE. This discovery is consistent with the findings of Liu, et al., (2013), who reported that longer exfoliation times result in smaller mean particle sizes and larger lateral sizes of halloysites (Liu et al., 2013). This is also validated by Raman spectroscopy, where we see that a lower ID/IG ratio corresponds to smaller particle diameters, which translates into larger sizes. The ID/IG decreases when the lateral size is changed (Amri et al., 2021). This can be correlated with the particle sizes of graphene nanosheets synthesized by Shokrieh, et al., (2014) and Ho, et al., (2020), which were 3.3 μm and 5 μm, respectively (Ho et al., 2020; Shokrieh et al., 2014). Thus, it can be inferred that particle size distribution depends significantly on the synthesis methods.

The bands appearing at 690.9–655.2 cm⁻¹ were due to C-H bending vibration modes and are in conformity with the characteristic band observed at 657.3 cm⁻¹, as shown in Figure 7. The absorption peaks at ∼988.4 cm⁻¹ and ∼984.4 cm⁻¹ were assigned to the broad C-H absorption bands associated with graphene (Hayyan et al., 2015), which are particularly dominant in zigzag structures. Additionally, it is also considered that the peaks at 2107.9 cm-1 and 2103.9 cm-1 are due to CO2 in air absorption (Shen et al., 2012). The well-known peaks of 2915.7 cm⁻¹ and 2917.1 cm⁻¹ were related to the asymmetric and symmetric stretching vibrations of CH₂ bonds, respectively. No oxidation or functional defects were observed after the synthesis of graphene by TASE-HSE, as evidenced by the absence of the C=C bond group from the non-oxidized sp² bonding (near 1618–cm⁻¹) (Guo et al., 2016).

**Conclusions**

Through the current process, a simple, green, and low-cost TASE-HSE method was used to reductively prepare few-layer graphene (FLG) with fewer defects. Under optimal conditions (5000 rpm for 3 hours), flake number was stacked in 2–3 layers, which were confirmed by the symmetric 2D peak with FWHM of 35 cm⁻¹ and a high I2D/IG ratio (0.6) thus reflecting good structural stability. The presence of the defects, edge vacancies instead of oxidative disruptions, was further confirmed by the ID/IG ratio (0.1). Additionally, a high defect density was observed at lightly to moderately processed conditions, as determined by Raman spectroscopy analysis. However, the defects decreased with further intense processing of HSM, so that it can be considered that the degradation of FLG started from existing vacancy defects or tiny scratches and later matured to the point of completely cleaving graphite layers. Raman imaging revealed a dominance of zigzag edge chirality, while TEM and PSA analyses demonstrated control over lateral size (~396.5 nm) and particle uniformity. Furthermore, the minimal reduction in graphene size was attributed to this method, which was primarily effective at exfoliating interlayer graphene by breaking the van der Waals bonds between layers rather than breaking the sheets frontally or laterally. Moreover, FTIR analysis revealed that the defects observed in FLG were not due to oxidative defects, but rather to edge and vacancy defects that were formed during the exfoliation process. This study demonstrates that precise control of shear exfoliation significantly impacts the quality of graphene, thereby enabling the sustainable mass production of FLG with minimal defects.

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**Compliance with ethics guidelines**

The authors declare they have no conflict of interest or financial conflicts to disclose.

This article contains no studies with human or animal subjects performed by authors.

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