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Comparative Study of Solvent-Assisted Exfoliation of Low Rank Coal for Few-Layer Graphene Production via Multi-Stage Ultrasonication

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ABSTRACT

Low rank coal (LRC), due to its abundance and carbon richness, represents a promising and sustainable precursor for graphene synthesis. This study compares the effects of different solvents CTAB (2%), NaOH (1N), H_2SO_4 (1N), and isopropyl alcohol (IPA)—on the exfoliation efficiency of LRC using a multi-stage ultrasonication approach. The resulting materials were characterized by FTIR, SEM, TEM, and XRD to evaluate their structural, morphological, and chemical properties. The findings reveal that IPA provides the most effective exfoliation, yielding few-layer graphene (FLG) with minimal oxidation and structural distortion. FTIR spectra showed reduced hydroxyl and carbonyl peaks in IPA-treated samples, while SEM and TEM confirmed more open and less-stacked layers. XRD analysis indicated decreased crystallinity and larger interlayer spacing. These results demonstrate that solvent selection plays a critical role in determining exfoliation performance, with IPA emerging as the most efficient and environmentally friendly medium for graphene production from LRC.

1. INTRODUCTION

Graphene is a two-dimensional nanocarbon material that exhibits extraordinary electrical conductivity (Gonçalves, 2025; Zheng et al., 2023); high thermal stability (Gonçalves, 2025), and superior mechanical strength. Despite its immense potential in electronics (Menaa et al., 2021; Rustamaji et al., 2025) energy storage, and catalysis (Fan et al., 2021), the commercial-scale production of graphene remains limited due to the reliance on expensive precursors and energy-intensive synthesis routes (Tamuly et al., 2022). Among emerging alternatives, low rank coal (LRC) a carbon-rich, low-cost, and abundantly available material in Indonesia, offers a promising and underutilized source for graphene synthesis (Purwandari et al., 2023).

Unlike highly crystalline graphite, LRC possesses an amorphous structure enriched with oxygen-containing functional groups (Chen et al., 2025), making it chemically reactive and amenable to structural modifications (Das et al., 2022). This study explores the solvent-assisted exfoliation of LRC into graphene using a multi-stage ultrasonication method. Four solvents—CTAB, NaOH, H_2SO_4, and isopropyl alcohol (IPA)—were evaluated based on their chemical interaction with LRC and their efficiency in layer separation and stabilization. The resulting materials were analyzed by FTIR, SEM, TEM, and XRD to assess structural, morphological, and chemical changes.

Most previous studies have focused on graphite as the starting material and used expensive or toxic solvents for graphene exfoliation. Only a few have explored low rank coal (LRC) as a viable carbon precursor, particularly in a comparative solvent framework under controlled ultrasonication conditions (Gao et al., 2024). Furthermore, the

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combined influence of solvent types and multi-stage ultrasonication on the exfoliation efficiency and material quality remains largely unexamined. To address this gap, the present investigates the effect of four different solvents—CTAB, NaOH, H_2SO_4, and isopropyl alcohol (IPA)—on the exfoliation behavior of LRC through multi-stage ultrasonication.

The central objective of this study is to determine the solvent that produces the highest-quality few-layer graphene (FLG) with minimal oxidation and structural distortion. In this context, the term optimization refers to the comparative identification of solvent conditions that provide the best structural integrity and exfoliation efficiency, as verified through spectroscopic and microscopic analyses.

2. METHODS

Materials

The main material utilized in this research was low rank coal (LRC) sourced from Musi Banyuasin, Indonesia. The coal was initially processed by crushing and screening it to achieve a particle size of less than 44 μm with a 200-mesh sieve. The solvents employed for exfoliation included 2% Cetyltrimethylammonium Bromide (CTAB) (Bu et al., 2024), 1N Sodium Hydroxide (NaOH), 1N Sulfuric Acid (H_2SO_4), and Isopropyl Alcohol (IPA) (Dai et al., 2023; Das et al., 2022), all of which were of analytical grade. Deionized water was utilized in all the processes.

Sample Preparation

The raw coal was subjected to pyrolysis at 400–500°C for 3 hours to reduce volatile matter content. Following thermal treatment, the sample was allowed to cool and stored in airtight containers prior to solvent treatment.

Exfoliation Procedure

Around 2 grams of pretreated LRC was mixed into 100 mL of each solvent and stirred with a magnetic stirrer for 15 minutes to achieve homogenization ultrasonication setup. Exfoliation was performed using a probe-type ultrasonic processor (model: MH-020S). The operating frequency was 24 kHz, with amplitude set to 60% (corresponding to an actual power of P = 120 W, measured at the generator readout). Sonication was applied in a multi-stage regimen to limit overheating: n = 10 stages, each t_{stage} = 10 min, with cooling intervals = 15 min between stages (total effective sonication time t_{total} = 10 × 10 = 100 min). The suspension temperature was maintained below ≤ 40 °C using an ice/water bath. After sonication, the dispersion was centrifuged at 4000 rpm for 10 minutes to separate the graphene layers that were suspended. The supernatant was gathered and Dehydrated in an oven at 105°C for a duration of 6 hours. The residue was re-sonicated and centrifuged multiple times until all carbon particles were no longer visible.

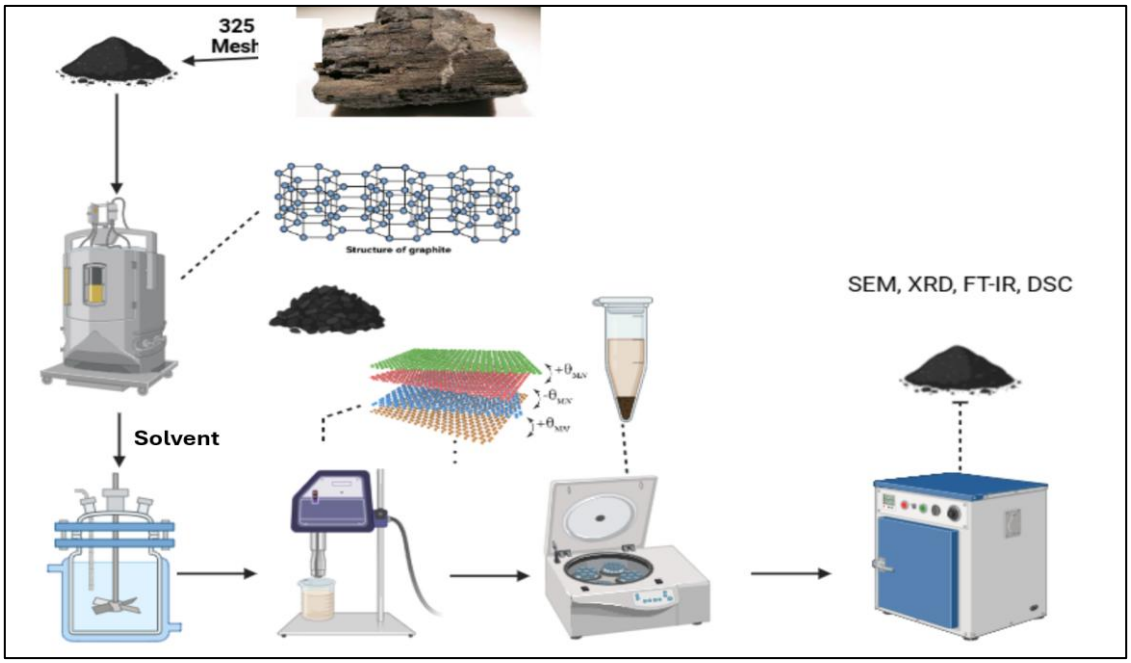


Figure 1. Sub-bituminous coal exfoliation procedure

Methods for Characterization

The structural and chemical characteristics of the exfoliated samples were analyzed using various methods. Fourier Transform Infrared Spectroscopy (FTIR, Shimadzu) was employed to detect surface functional groups within the wavenumber range of 4000 to 400 cm^{-1}. The morphology and particle size distribution were examined using Scanning Electron Microscopy (SEM, JEOL JSM-6510). High-resolution imaging for assessing layer thickness was achieved using Transmission Electron Microscopy (TEM, JEOL JEM-2100). X-ray Diffraction (XRD, PANalytical Empyrean) was performed utilizing Cu Kα radiation (λ = 1.5406 Å) to assess the crystallinity and interlayer spacing.

3. RESULT AND DISCUSSION

Proximate analysis

Table 1. Proximate Analysis of low rank coal (LRC) obtained from Musi Banyuasin, Indonesia

No	Sample	Proximate	
		In.Moist (%, adb)	VM (%, adb)
1	LRC	13.58	44.40

The proximate analysis of the low rank coal (LRC) sample indicates it has promise as a suitable starting material for synthesizing graphene. The sample contains a high percentage of volatile matter (VM) at 44.40%, which suggests strong reactivity and ease of thermal decomposition—beneficial traits for carbon exfoliation (Sun, 2024). The fixed carbon (FC) content of 40.93% reflects a significant proportion of combustible carbon, crucial for forming graphitic structures (Sun, 2024). Meanwhile, the inherent moisture (In.Moist) is relatively moderate at 13.58%, which, although it may lower thermal efficiency, can enhance solvent interaction during liquid-phase exfoliation. Importantly, the ash content is low at 1.09%, minimizing the presence of inorganic residues and thereby supporting the production of purer graphene materials (Bu et al., 2024). Overall, this proximate composition confirms that LRC possesses the fundamental characteristics necessary for conversion into few-layer graphene via physical and chemical exfoliation methods.

FTIR Spectral Analysis

The FTIR spectra (fig. 2) revealed distinct functional group variations between raw LRC and exfoliated samples. A wide absorption band around ~3400 cm^{-1} was associated with O–H stretching vibrations, which suggests the presence of hydroxyl groups.(Dai et al., 2023). This peak was prominent in NaOH-treated samples, suggesting hydroxylation during exfoliation. The band at ~1700 cm^{-1}, associated with C=O stretching (Nandiyanto et al., 2023), appeared in H_2SO_4-treated samples due to partial oxidation. In contrast, IPA-treated samples exhibited minimal oxygen-containing groups, preserving the aromatic C=C stretching around 1600 cm^{-1}(Bouramdane et al., 2022) thereby indicating less disruption to the graphene basal planes. These results confirm that IPA is a milder solvent, favoring structural integrity during exfoliation.

a

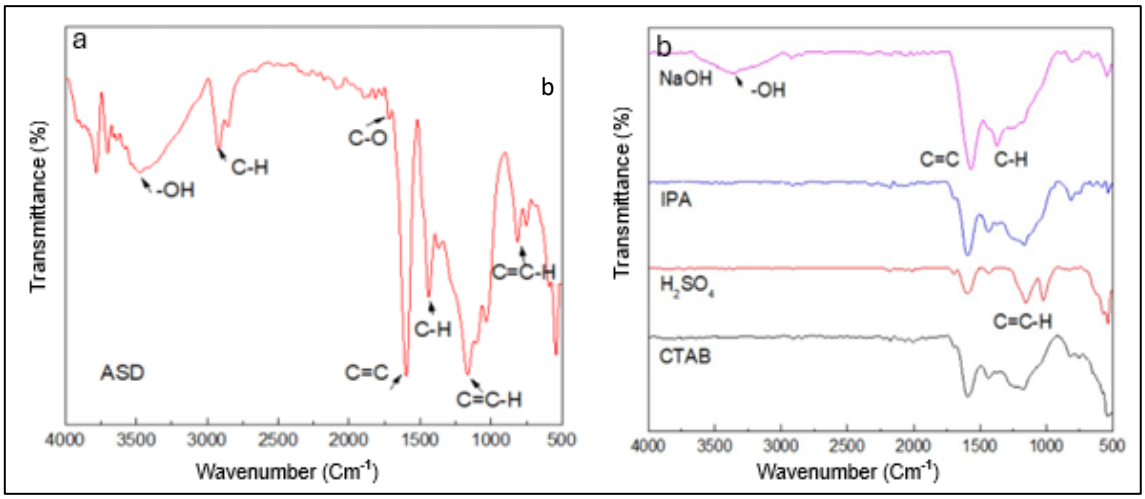
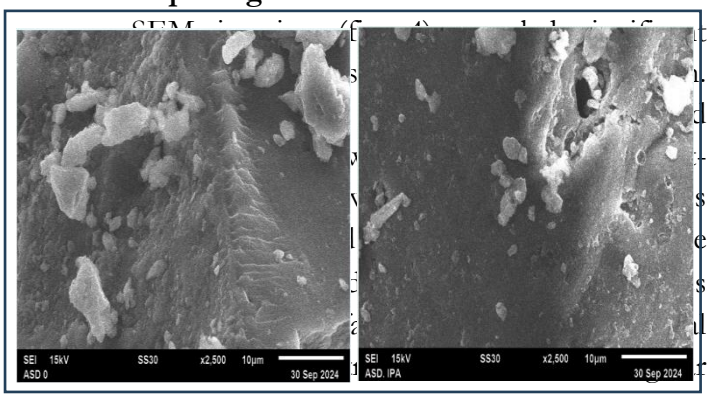


Figure 2. FTIR Spectra of Low Rank Coal and Exfoliated Products

Figure 2. (a) FTIR spectrum of raw low rank coal (ASD) showing characteristic peaks of hydroxyl group (-OH) 3493 cm^{-1}, aliphatic carbon-hydrogen (C–H) 2987 cm^{-1}, carbonyl group (C=O) 1700cm^{-1}, aromatic carbon-carbon double bond (C=C)1648 cm^{-1}, and out-of-plane bending of carbon-hydrogen (C–H) bonds. (b) FTIR spectra of exfoliated coal using different solvents: NaOH, IPA, H_2SO_4, and CTAB.

Based on the FTIR results, the IPA-treated sample exhibited the most favorable spectral characteristics among all solvents. The dominant aromatic C=C peak at ~1595 cm^{-1} was retained with the highest relative area, indicating that IPA effectively exfoliated the carbon layers while preserving the sp^2-hybridized graphene framework. In contrast, the H_2SO_4-treated sample showed the smallest peak area and additional features near 998 cm^{-1}, reflecting stronger oxidation and structural disruption. The consistent yet minor shifts around 1411 cm^{-1} observed in all treated samples suggest mild chemical modification, but in the case of IPA, these changes were minimal. Overall, the broader aromatic intensity and absence of significant oxygenated bands confirm that IPA serves as the most efficient and least aggressive solvent for exfoliating low-rank coal into few-layer graphene.

SEM Morphological Observations



flake morphology with apparent particle fragmentation. The open-layered morphology in IPA samples correlates with effective solvent penetration and cavitation-induced separation.

Figure 3. SEM Coal LRC and LRC-IPA

TEM Imaging

TEM analysis provided high-resolution images of exfoliated structures, confirming the presence of few-layer graphene (FLG) in IPA-treated samples. The images exhibited transparent, thin sheets with moderate stacking, while CTAB and H_2SO_4 treatments led to denser flake regions. The hexagonal lattice fringes were visible in some regions, indicating the preservation of sp^2 bonding networks. NaOH-treated samples showed disordered carbon layers with limited transparency, suggesting partial degradation of the graphene structure.

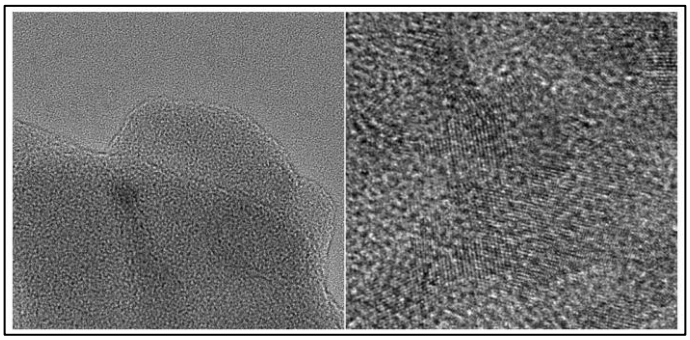


Figure 4. TEM image of LRC in IPA

XRD Analysis

XRD patterns revealed a broad peak at $2\theta \approx 16°$ in raw LRC, characteristic of amorphous carbon (Moseenkov et al., 2023). Post-exfoliation, IPA-treated samples displayed diminished peak intensity and broadened diffraction signals, indicating reduced crystallinity and increased interlayer spacing.

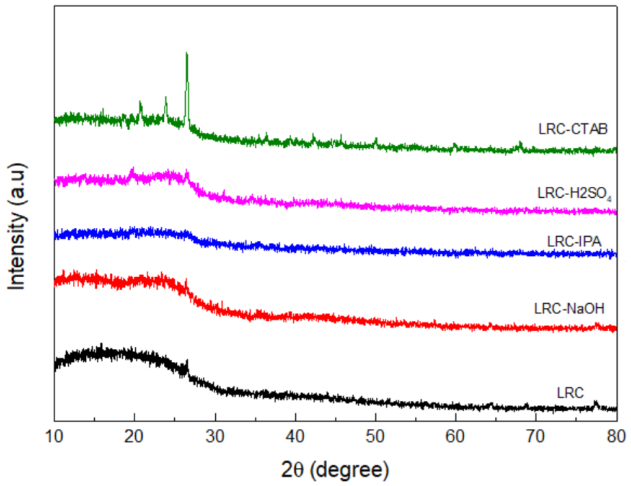


Figure 5. XRD analysis of LRC and LRC-NaOH; LRC-IPA; LRC-H_2SO_4; LRC-CTAB samples

The disappearance of the sharp peak at ~26.5° (graphitic (002) plane) in IPA samples suggests successful delamination into few-layer graphene. Meanwhile, NaOH and H_2SO_4 samples retained partial crystallinity, whereas CTAB-treated samples exhibited intermediate behavior. These results substantiate that IPA enables better exfoliation with minimal disruption to the sp^2 domains.

The XRD profiles (Figure 3) show a broad diffraction band centered at ~16° in raw LRC, corresponding to the amorphous (002) plane of carbon. Upon solvent-assisted exfoliation, all treated samples exhibit diminished peak intensity and increased broadness, reflecting disrupted graphitic stacking. Among the tested solvents, the IPA-treated sample displays the most diffuse pattern and lowest (002) intensity, confirming the effective delamination of LRC into few-layer graphene. Conversely, the CTAB and H_2SO_4 samples show partial retention of the (002) reflection, indicating limited exfoliation.

Table 2. XRD Parameters of Raw and Solvent-Treated LRC Samples

Sample	Main Diffraction Peak (2θ,°)	d002 (nm)	Relative Intensity	FWHM (Qualitative)	Crystallinity / Structural Interpretation
LRC (raw)	~16.0	0.55	Moderate	Narrow	Amorphous carbon with limited graphitic ordering.
LRC–NaOH	~15.0	0.59	Low	Broad	Partial oxidation and layer disruption; hydroxylation increases interlayer spacing.
LRC–IPA	~25.0	0.36	Very low	Very broad	Highly delaminated structure; indicates few-layer graphene with minimal restacking.
LRC–H_2SO_4	~25.2	0.35	Moderate	Moderate	Partial exfoliation and oxidation; residual ordering or restacking
LRC–CTAB	~26.5	0.34	High	Narrow	Retained crystalline domains, possibly due to surfactant residues; limited exfoliation.

Table 2 presents the XRD-derived parameters of raw and solvent-treated LRC samples. The untreated coal (LRC) shows a broad amorphous (002) peak at ~16°, corresponding to a d-spacing of 0.55 nm. Following solvent-assisted exfoliation, all treated samples exhibit broadened and weakened diffraction peaks, confirming disruption of long-range stacking. The IPA-treated sample, in particular, displays the lowest intensity and broadest profile, implying the formation of few-layer graphene. In contrast, NaOH and H_2SO_4 cause partial oxidation and layer distortion, while CTAB retains partial graphitic order due to surfactant adsorption. These findings confirm that IPA achieves the most efficient exfoliation with minimal structural degradation.

4. CONCLUSION

This study demonstrates that low-rank coal (LRC) can be effectively transformed into few-layer graphene (FLG) through solvent-assisted exfoliation using a multi-stage ultrasonication process. Among the tested solvents, isopropyl alcohol (IPA) proved to be the most efficient and environmentally friendly medium, enabling the formation of thin, well-dispersed graphene layers while maintaining the structural integrity of the carbon framework.

Beyond laboratory findings, this work highlights a sustainable pathway for graphene production that utilizes an abundant, low-cost, and underexploited carbon source. The process reduces reliance on high-purity graphite and toxic reagents,

offering compatibility with scalable, green manufacturing approaches. These insights contribute to the growing field of sustainable nanocarbon synthesis and suggest that LRC-based graphene could complement existing industrial routes for energy storage, catalysis, and composite materials.

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