



# A Simple Microwave Synthesis of Exfoliated Graphite for Spilled Heavy Oils Removal

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## KEYWORDS

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**ABSTRACT:** In recent times, there has been a growing interest in the development of carbon-based absorbent materials for handling oil spills. This attention is primarily due to their advantageous characteristics such as low cost, high surface area, porous structure, natural abundance, and environmental friendliness. In this study, we introduce a straightforward and cost-effective method for producing exfoliated graphite (EG) as an absorbent material. This method involves the utilization of diluted perchloric acid and hydrogen peroxide, followed by microwave irradiation, resulting in a rapid synthesis process. The EG material obtained through this approach demonstrates exceptional adsorption capabilities across a wide range of oil types, including kerosene and engine oil. The adsorption efficiency for kerosene and engine oil was found to be 96% and 99%, respectively, using just 1 gram of EG. Moreover, we discovered that a combination of exfoliated graphite with a weight ratio of 1:2:1 resulted in an increased maximum volume of 570 mL/g. These findings highlight the high yield achieved in this present study compared to previous research efforts.

## 1. Introduction

In recent years, oil contamination has led to significant and widespread consequences. Oil spill incidents have released hundreds of tons of oil into the sea, such as the oil spill disaster of the oil tanker "Prestige" in 2002, which spilled around 77,000 tons of crude oil [1]. Such as the recent Orange County oil spill in 2021 [2]. M V Wakashio (Mauritius) oil spill in 2020 [3]. MT Dawn Kanchipuram accident in 2017 [4]. While this is happening each year, a massive volume of oil spills into the ocean through transporting oil containers, coastal refineries, and urban runoff and discharges. The oil leak not only significantly pollutes the seas, but also puts them at risk of marine life. Additionally, the loss of millions of tons of energy resources is important for humans. As a result, developing a system to recover and recycle the spilled oil is critical [5-10]. Here Exfoliated graphite (EG) discusses graphite upon expansion along the C-axis up to several hundred times, making it a promising candidate for a wide range of potential applications [11-15]. There are many advantages to exfoliated graphite (EG) over other inorganic materials,

including excellent compression, elasticity, and thermal stability [16]. Among its many uses are gaskets, insulation, fire-resistant composites, resin composites, and electrodes [17]. An EG material has a low density, and high porosity, is easy to dispose of, and is environmentally friendly [18]. The porous structure of the sorbent allows it to have a high sorption capacity for cleaning up oil spills [19]. It has been reported by Toyoda and Inagaki that EG has a heavy oil sorption capacity of more than 80 grams per gram [20]. It's time to make EG using a traditional heating complex process requiring more energy (12-hours mixing) (The oven operates at 1000 °C and the drying is dry) [21-25]. With large amounts of distilled water for cleaning. Switch Technologies for rapid Exfoliated Graphite generation include coupled plasma, laser, and Microwave irradiation, which includes microwave irradiation Suitable for EG production due to low cost and Minimal performance followed by ease of use [26]. In, the intermediate Preparation process of intercalated graphite (IG) Washing and drying takes time and effort [27]. In this study, EG was produced directly by microwave irradiation Wash and dry in less than a



minute (10 seconds for mixing, 80 seconds for microwave irradiation). Exfoliated graphite (EG) is a new kind of functional Nano-carbon material, which has attracted global attention due to the increasing number of potential applications. Exfoliated Graphite is a kind of graphitic carbon that is formed by chemically intercalating the carbon layers of graphite with an intercalating agent and then quickly the heating rate. Heating causes vaporization and decomposition of the intercalate, which, in turn, supplies the force required for sliding the carbon walls relative to each other, thereby allowing the expansion of the material [28]. However, the production of EG has several disadvantages such as the consumption of huge quantities of chemicals, time, water energy, etc. In the present study, a mesoporous EG with a high surface area was prepared through the chemical intercalation process followed by microwave heating within 1 minute. As a result of the exfoliation process, the specific surface area of the material increases significantly making it very valuable for application as an adsorbent material for pollutants. Describes the comparison synthesis and properties of previously reported EG with the present study [29-31]

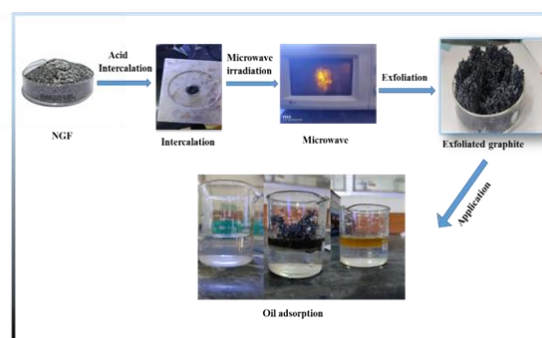
## 2. Experimental Method.

### 2.1. Materials

Natural flake graphite (NGF) (Carbon content 99.88%, normal flake size of 425  $\mu\text{m}$ , India) and perchloric acid (70%) analytical Grade Hydrogen peroxide (30%).

### 2.2. Synthesis of exfoliated graphite (EG):

Natural flake graphite, perchloric acid (70%), and Hydrogen peroxide (30%) it was mixed with various weight ratios (1:2:1) for 10 sec using a glass rod in a Glass beaker to obtain IG. An analysis was conducted on the different ratios of intercalated graphite. The sample was positioned in a glass beaker and subjected to microwaving for 80 seconds at a power setting of 700 watts using an LG microwave oven. EG was generated using various ratios and fumes within the micro oven during the irradiation process. The synthesized EG was stored for further characterization. The graphite structure, intercalation and exfoliation, and oil adsorption performance of EG were represented schematically in the schematic. Figure 1.



**Figure 1. The formation of EG and oil sorption**

### 2.3. Characterization.

The surface morphology of the graphite intercalation compound where recorded using Scanning Electron Microscopy (SEM). Energy Dispersive X-ray Analysis (EDAX) was used to find the elemental composition of the exfoliated graphite (EG). The X-ray diffraction (XRD) is carried out by using structural analysis a beaker with a diameter of 50 mm and a capacity of 500 ml was used to determine the expanded volume UV-VIS absorption spectra in the Wavelength range of 200–800 nm were obtained UV-3600 Plus Series An average of separate batches were created by microwave irradiation to enlarge the volume. After microwave irradiation, the EG's BET surfaces and pore volumes were determined (which displayed a bulky expanded volume). An identified amount of EG material remained placed in a volume measuring jar with no gaps and the total volume occupied by the EG was calculated. The assessed value is the arithmetic average of five experimental studies' calculated values using the methods developed above.

## 3. Results and discussion.

The exfoliated volume of EG: The characteristics of exfoliated volume (EV) were obtained for different weight ratios for exfoliated graphite (EG). Figure 2 shows a plot of Natural Graphite Flaks (NGF), Intercalates, and Oxidants at various ratios. Oxidizing chemicals such as perchloric acid ( $\text{HClO}_4$ ) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) were employed to extract EG from NGF. The weight content of intercalation and oxidizing agents was found to have a significant impact on the Exfoliating volume (EV) of EG, as shown in Figure 2. Maintaining constant the rest of the ratio while changing the oxidation agent, EV was found to have the



maximum value of 570 mL/g for sample 1:2:1 (sample no. 6 Figure 2) which is higher than that of other ratios Figure 2. A 1:2:1 sample was chosen to study for different microwave powers, 700 W, 500 W, and 350 W, because of the higher value of EV=570 mL/g compared to other ratios and the variation of EV was shown in Figure 3. As shown in Figure 4. Microwave power alters EV at 80 s exfoliation time accordingly, it is shown in Figure 3 and 4 that 80 s of microwave irradiation at 700W is sufficient to achieve high-yield exfoliation of EG.

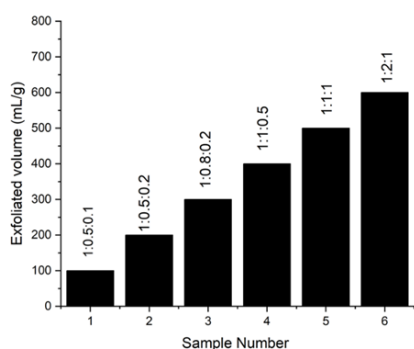


Figure 2. Exfoliating volume of various ratios of EG.

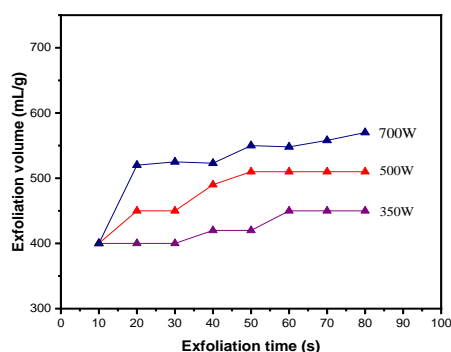


Figure 3. Volume of exfoliation at different time and different watts.

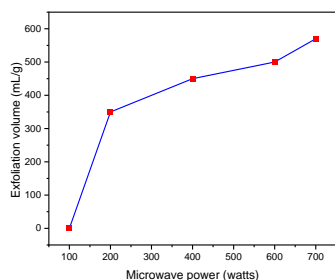


Figure 4. Exfoliation volume at various power levels.

### 3.1. SEM analysis

Microstructural of Exfoliated graphite Figure 5(a-d) shows the SEM micrographs for commercially received natural flake graphite Figure 5(a), corresponds to the flat flake-like morphology of exfoliated graphite and Figure 5(b) shows the lower magnification of exfoliated graphite for intercalation compound. In Figure 5(c) the surface morphology of EG high magnification of the worm-like structure under microwave irradiation. Figure 5(d) shows the EDAX spectrum of the exfoliated graphite indicated by the high percentage of atomic of carbon (>81%) and oxygen (18%) and a trace amount of Cl (1%) the high purity of EG samples present in the quick method.

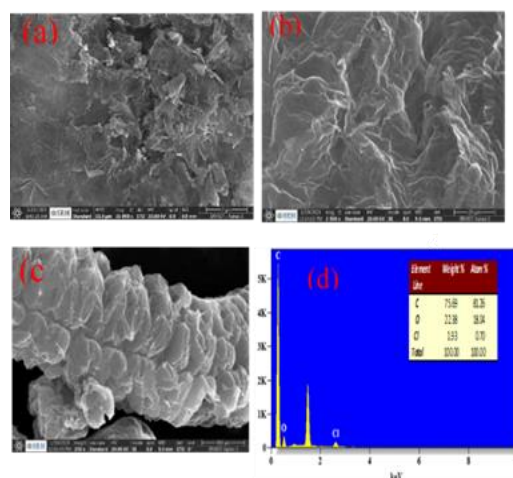


Figure 5. SEM images of (a) Natural flake graphite, (b) graphite intercalation, (c) EG at high magnification, and (d) EDAX spectrum of EG (There is an inset showing the proportions of the elements present) for sample no. 6.

### 3.2. XRD analysis

The X-ray diffraction analysis was crucial in determining the crystal's nature, phase purity, and crystalline structure of the samples prepared. Figure 6. Despite the X-ray diffraction analysis of prepared EG, GIC and NGF. The EG, GIC, and NGF formations were confirmed using JCPDS No 00-012-0212 [38]. In Figure 6, the major XRD pattern of EG, GIC, and NGF is displayed, showing peaks at  $2\theta = 26.4^\circ$  and  $54.5^\circ$ , corresponding to the (002) and (004) crystalline planes [39]. The distinct peaks observed in the analyzed EG, GIC, and NGF samples (refer to Figure 6) can be attributed to the high degree of crystallinity of the

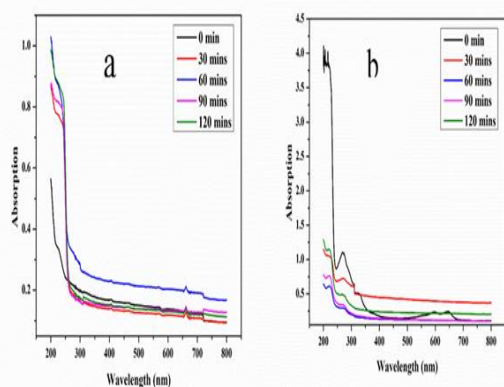


material. From the analysis, it was determined that the EG, GIC, and NGF samples exhibited a hexagonal structure (specifically, space group P63/mmc with a space group number of 194) with lattice parameters of  $a=b= 2.4640 \text{ \AA}$  and  $c=6.7360 \text{ \AA}$ .

The average crystalline grain size was calculated for the prepared material by using the Debye-Scherrer equation (1) [36].

$$D = K\lambda / (\beta \cos \theta) \quad - (1)$$

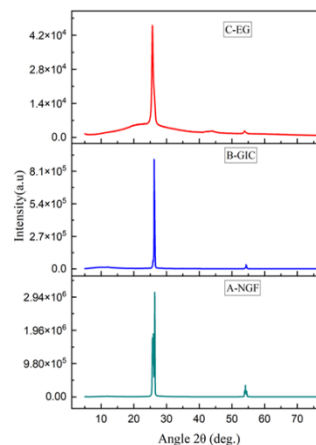
The average crystalline grain sizes for EG, GIC, and NGF were found to be 12.31 nm, 13.60 nm, and 12.52 nm, respectively.



**Figure 6. XRD analysis of (a) NGF (b) GIC (c) EG.**

### 3.3. UV-Vis absorption spectra.

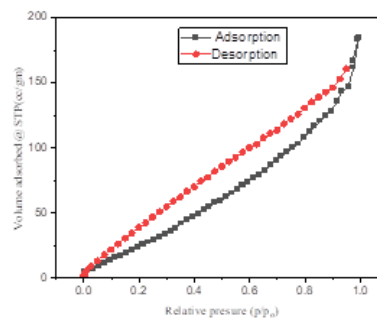
The concentration of kerosene in a sample can also be determined by measuring the absorbance of the sample at 660 nm. The UV absorption spectrum of kerosene can also be used to study its composition of kerosene. For example, the UV absorption spectrum of kerosene can be used to determine the type of crude oil that the kerosene was derived. Overall, the UV absorption spectrum of kerosene is a valuable tool for identifying, quantifying, and studying kerosene Figure 7 (a). and Figure 7(b). The UV absorption spectrum of engine oil can be used to identify engine oil in a sample and to determine its concentration. For example, the UV absorption spectrum of engine oil can be used to identify engine oil in a sample of contaminated water. The concentration of engine oil in a sample can also be determined by measuring the absorbance of the sample at 275 nm.



**Figure 7. UV-Vis absorption spectra analysis of (a) Kerosene oil (b) Engine oil.**

### 3.4. BET analysis.

The Brunauer–Emmett–Teller (BET) gas adsorption method was used to analyze the surface characteristics (surface area, total pore volume, and pore size) of EG. Show the nitrogen The pore volume and the surface area of mesoporous graphite were investigated by nitrogen adsorption/ desorption analysis as shown in Figure 8. the adsorption/ desorption isotherms associated with the type IV indicates the presence of mesopores [36]. The results reveal that prepared mesoporous graphite has many infrequent micropores with a BET-specific surface area of 142.345 m<sup>2</sup> /g. The high volume of small tiny pores is responsible for more surface area and pore volume 0.288 cc/g.



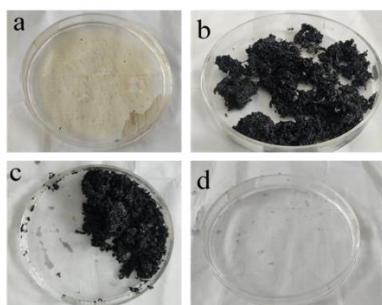
**Figure 8. Nitrogen adsorption/desorption isotherms of as prepared exfoliated graphite.**

### 3.5. Oil sorption

EG can rapidly absorb a large amount of spilled oil. Typical pictures of the appearance of



floating engine oil and kerosene oil before and after adding the exfoliated graphite to the floating oil are shown in Figures 9 (a) and 9 (b) Engine oil and kerosene oil floating on water were completely sorbed into the added EG. Even after the sorption of the spilled oil, the EG remained floating on the water, and the sorbed oil could easily be separated from the water in exfoliated graphite [37]. After removing the EG no contamination remained in the water, into the water, no characteristic color of the oil was observed after transferring the oil-sorbed EG, as shown in Figures 9 (c) and (d). In the case of however, the EG was able to sorb a large amount of oil and was easily separated from the water using an exfoliated graphite. As part of this experiment, 250 ml of water was placed in a 500 ml beaker, and different oils were added individually and gently stirred for 40 seconds. The oil was kerosene oil ( $2.48 \times 10^{-4} \text{ m}^2/\text{s}$ , density  $0.658 \text{ g/cm}^3$ ) and engine oil ( $15 \times 10^{-6} \text{ m}^2/\text{s}$ , density  $0.84 \text{ g/cm}^3$ ) were both added individually and gently stirred for 40 seconds. However, after stirring, the entire amount floated out of the water. After mixing oil with water, EG was added. A large quantity of oil floating on the surface of the water was absorbed by EG. Exfoliated graphite extracted from the beaker did not leave any oil drops on the surface of the water. By changing the weight ratio of heavy oil to EG, the maximum oil absorption capability of EG was determined. In this example, the adsorption capacity was expressed in grams of heavy oil sorbed per gram of EG. A maximum adsorption capacity of 96% g/g was determined for EG concerning kerosene, and 99% g/g for engine oils in the present study. According to our results, the sorption capacity is greater than that reported in previous studies.



**Figure 9.** (a). Shows the floating engine oil on water. (b). after adding exfoliated graphite. (c). Remove exfoliated graphite (d). After removing exfoliated graphite clear water.

#### 4. Conclusion.

This study employed a microwave irradiation technique, which enabled the rapid production of exfoliated graphite (EG) within just one minute. The investigation focused on the influence of varying oxidants and intercalation on the exfoliated volume (EV). Notably, when using a weight ratio of 1:2:1 and a microwave power of 700W, the maximum EV yield of 570 mL/g was achieved for EG. The sorption capacities of EG were evaluated, revealing an impressive oil adsorption rate of 96% for kerosene and 99% for engine oil using only 1 g of EG per oil. With its low-cost and straightforward production method, EG proves to be a promising material for cost-effective oil recovery in spill scenarios. In conclusion, this study demonstrates the potential of EG as an efficient and affordable solution for oil spill remediation.

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#### Competing interests

The Author declares on behalf of all authors that there is no conflict of interest.

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