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Recent Trends in Transforming Different Waste Materials into Graphene *via* Flash Joule Heating

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# 1 Recent Trends in Transforming Different Waste Materials into Graphene

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- 18 Abstract

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19 The conversion of waste materials into graphene using the Flash Joule Heating (FJH) method

presents an eco-friendly and sustainable approach to addressing the global issue of

21 environmental pollution. FJH efficiently transforms carbon-based materials, such as plastics,

batteries, and food wastes, into high-quality graphene by applying a high-voltage electrical

current that generates temperatures exceeding 3,000°C within milliseconds to seconds. This

technique not only offers a rapid and energy-efficient pathway for graphene synthesis but also

25 tackles the environmental drawbacks of traditional waste disposal methods. Compared to

conventional graphene production techniques, which depend on using harsh chemicals or

27 extreme conditions and tend to be energy-intensive and costly, FJH is scalable, fast, cost-

efficient as it costs 130-135 US dollars per ton using coal as a carbon source, capable of

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handling a wide range of waste materials without requiring solvents or catalysts, and only produces about 10 kg of carbon dioxide (CO<sub>2</sub>) compared to 400-500 kg released from conventional synthesis for each kg of graphene. Flash graphene (FG) shows promise in various applications, including concrete production, water purification, and supercapacitors, making it a crucial contributor to the development of sustainable technologies. This article delves into the basics of FJH, the automation, and the scaling up of the process over the last four years with an aspiration to produce 100 tons of graphene per day. Various characterization techniques of FG, the advantages of FG compared to conventional graphene, and the challenges and limitations of FJH are discussed.

#### Keywords

- 39 Flash graphene; Turbostratic graphene; Flash joule heating automation; Scaling-up; Carbon-
- 40 based waste materials

#### 1. Introduction

Graphene, a 2D single layer of carbon atoms arranged in a hexagonal structure with Sp<sup>2</sup> hybridization, is sought for its exceptional properties, making it one of the most promising materials of the 21<sup>st</sup> century [1, 2]. The outstanding electrical conductivity, mechanical strength, thermal stability, and flexibility of graphene make it a special material compared to other materials [3, 4]. These unique attributes have placed graphene at the forefront of research in nanotechnology, electronics, and materials science [5].

Graphene's range of applications is extensive and continues to grow. In electronics, it results in faster, more efficient transistors and flexible, transparent conductors for touchscreens and displays [6]. In the energy sector, graphene improves the performance of batteries, supercapacitors, and solar cells, offering greater energy storage capacity and efficiency [7]. Additionally, its high surface area and chemical reactivity make it an ideal material for sensors, catalysis, and water purification [8].

Graphene Field Effect Transistors (GFETs) leverage graphene's exceptional electrical, thermal, and mechanical properties to advance nanoelectronics, offering significant improvements over traditional silicon-based transistors [9]. With high electron mobility (up to 30,000 cm²/V·s) and an ambipolar electric field effect, GFETs enable faster charge transport, high-frequency operation, and potential power savings. Their adaptability allows integration on flexible substrates, paving the way for portable and wearable electronics and

furthering the trend of device miniaturization. Additionally, innovations like silicon-on-insulator structures have enhanced on/off ratios and short-channel control, addressing key challenges in digital applications. However, obstacles such as controlling graphene's bandgap and achieving large-scale uniform production remain. Despite these challenges, graphene's unique attributes, including mechanical flexibility, lightweight robustness, and high conductivity, position it as a versatile material for diverse applications. These applications include energy storage as graphene results in efficient, high-capacity batteries and supercapacitors, as well as advanced composites for the aerospace and automotive industries [10]. The versatility of graphene ensures its continued importance in advancing technology across numerous disciplines.

Several techniques are used to produce graphene, each has its strengths and limitations. Mechanical exfoliation, which involves peeling layers from graphite, was the first method to isolate graphene but is not feasible for large-scale production [11]. Chemical vapor deposition (CVD) offers a more scalable approach, allowing graphene growth on metal substrates [12], though it can be costly and complex. FJH is an emerging method that quickly converts carbon-rich materials, including waste materials, into graphene, making it particularly appealing for sustainable recycling [13].

In the context of upcycling waste materials, FJH has evolved as a novel technique that can revolutionize the way we deal with a wide range of different waste materials such as plastics by converting them into high-quality FG [14]. The process involves subjecting the waste material to a rapid, high-temperature electrical discharge, typically ranging from 2,500 to 3,000 degrees Celsius, within a matter of milliseconds. This sudden heating followed by rapid cooling causes the carbon atoms within the material to rearrange into a turbostratic graphene structure, which is more tuneable, manageable, and easily exfoliated compared to the conventional AB stacked graphene layers [15]. Consequently, FJH helps effectively transforming waste into a valuable product with minimal energy consumption in a very short time.

In this regard, the energy required to convert 1.0 g of mixed plastic waste (PW) into 0.18 g of high-quality FG is approximately 23 kJ using the direct current DC-FJH process as reported by Algozeeb et al. [16]. This transformation is highly efficient as it occurs in a remarkably short duration (~8 seconds) at temperatures close to 3100 K. The electricity cost of this process is around \$124 to convert 1 ton of PW into 180 kg of high-quality FG plus

volatiles, making it significantly more cost-effective than conventional physical and chemical recycling methods. Traditional recycling processes are often not economically profitable, as they tend to produce recycled plastics that are more expensive than new plastics [16]. For example, CVD requires energy-intensive conditions, with temperatures of up to 1020°C per gram of PW maintained for an extended period of 90 min, followed by an additional time for system cooling [17], The long processing time at such high temperatures significantly increases energy consumption. Similarly, pyrolysis involves heating large reactors to 500-600°C for over one hour, with the reaction time strongly dependent on maintaining these elevated temperatures to convert PW into liquid fuels. These prolonged durations at high temperatures lead to inefficiencies in energy use and time [16, 18]. Therefore, FJH drastically reduces energy consumption and also minimizes overall operational time. Additionally, FJH helps decrease environmental pollution and reliance on natural resources, offering a sustainable and economical solution for upcycling PW [16], which aligns with the principles of a circular economy by converting wastes into resources.

One of the key advantages of FJH is its versatility in processing a wide range of carbon-containing materials, such as mixed plastics, coke, biomass waste (eg: food waste), electronic wastes (eg: used batteries and electrodes), and even discarded tires, without the need for pre-sorting or purification [19-22]. The resulting FG exhibits excellent electrical conductivity, thermal stability, and mechanical strength, making it suitable for various applications in electronics, energy storage, and composite materials [21]. In this regard, Saadi et al. [23] reported that the electrical resistance of transforming asphaltene to FG dropped from 64 kiloohms (k $\Omega$ ) to just 60 ohms ( $\Omega$ ) using an asphaltene-based FG as a conductive agent by increasing its loading from 20 wt% to 50 wt% in a 500 mg sample of asphaltene. Another study undertaken by Zhu et al. [24] mentioned the decreased resistance of carbon black from 0.62  $\Omega$  to 0.45  $\Omega$  upon the first flashing and to 0.34  $\Omega$  upon the second flashing. Concerning the thermal stability, Advincula et al. [19] showed that a mix of carbon black with shredded rubber tires started the thermal degradation at 300°C, but it started at 550-600°C after adding FG into this composite as shown in Fig. 11.d. Similarly, adding FG to high-density polyethylene-based PW enhanced its thermal stability to start thermally degrading at 625°C as reported by Algozeeb et al. [16]. Also, the compressive and tensile strength of a carbon black-cement composite, which was measured after seven days, increased by almost 35% from 13.5 MPa to 18 MPa and 19% from 9.4 to 11.2 MPa, respectively, upon adding 0.1 wt% of FG into the composite [13]. However, FJH has its

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drawbacks including the high electrical energy requirement that can result in potential safety concerns such as electrocution and the risk of system explosion, which necessitates high safety precautions and trained personnel [13]. A venting system should also be in use to remove the volatiles (gases and vapours) from the heating chamber and to make use of these gases in other applications [25]. Additionally, there could be some difficulty in converting low-carbon source materials such as rice straw to FG, which requires mixing these carbon sources with already-produced FG or other high-carbon materials [26]. Other limitations of FJH are thoroughly discussed in the following sections.

FG has emerged as a hot topic, with a systematic analysis revealing only 46 publications over the past four years (**Fig. 1.a**), according to data available on Scopus [27]. The first article, published by Duy Luong et al. [13] in 2020, was authored by a PhD student from Professor James Tour's research group. Since then, most of these publications, which are currently 24, have been produced by the Tour group at Rice University, as shown in **Fig. 1.b** and **c**. The rest of the publications, which are 22, are published by other research groups. Therefore, this review article focuses on the recent advancements using the FJH technique in upcycling different waste materials and highlights the improvements and scaling-up of this technique, including the automation of the heating system. Additionally, this work covers the characteristics of FG and its advantages compared to the conventional type as well as the applicability of FG in diverse applications.

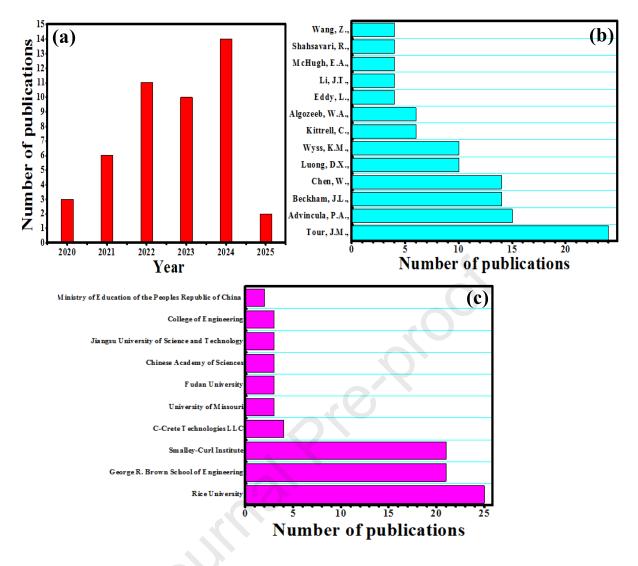


Fig 1. The number of FG -articles published over the last four years (a), and the main contributing researchers (b) and research institutes/universities to these publications (c). The data is obtained from Scopus [27].

#### 2. FJH process and its improvements

FJH is an advanced technique for rapidly converting carbon-rich materials into graphene and other valuable products. This process involves applying a high-voltage electric current to the precursor material, generating an intense, localized temperature surge that reaches several thousand degrees Celsius in milliseconds to seconds, as explained by Professor James Tour [28].

FJH technique was first introduced in 2020 by Professor James Tour's research team at Rice University in producing graphene. The main objective was to develop a more sustainable and cost-efficient approach to graphene production compared to traditional

methods such as CVD or mechanical exfoliation. Although these conventional methods (eg: CVD) produce high-quality graphene, they are not suited for commercial mass production due to their low yield [29, 30]. The need to use specific starting materials such as graphite in the mechanical exfoliation along with the high energy required, which can cause structure defects in the produced graphene and lower its quality, exacerbate the issues of this technique [15]. Additionally, the lack of control over the graphene layers' number and thickness adds to the problems of mechanical exfoliation [31]. Therefore, upon comparing the production cost with the yield of traditional methods, it becomes clear that they are not cost-efficient. On the contrary, FJH can be operated using various carbon waste materials in a very short time and mostly produces high-quality graphene [21]. In their early experiments, the researchers successfully transformed carbon-rich materials, including coal, food waste, and plastic, into FG by applying an electric current to induce rapid heating and graphitization within less than a second.

FJH applies a high electrical current, typically in the range of hundreds of amperes, through carbon-rich materials such as plastics, coal, biomass, or carbon black. Some of these carbon-based materials are electrically resistant, and their resistance can range from 1 to 50  $\Omega$ , as mentioned by Professor James Tour [28], but other materials such as carbon black are conductive. Therefore, the resistant materials are mixed with 2-5 wt% conductive materials such as carbon black or an already-prepared FG sample to improve their conductivity [13]. These precursor materials are positioned in a quartz tube, which could be of different lengths and inner diameters, between conductive electrodes that are made of copper or any other conductive material [13], as shown in Fig 2.a. The temperature during the FJH process can rise above 3,000°C in just milliseconds, as shown in the inset image of Fig 2.a, breaking molecular bonds and every carbon-carbon bond, and leading to the formation of graphene layers. Such a process is like heating toasts in a toaster as described by Prof. James Tour [28]. Other inorganic or non-carbon elements could either remain as impurities or sublime out. Therefore, the system should be properly outgassed either at atmospheric pressure or under vacuum for the sake of avoiding any explosion, which is achieved by loosely fitting the electrodes into the quartz tube [13]. Notably, this method does not rely on external gases, catalysts, or solvents, making it highly efficient and environmentally sustainable.

There is a perfect alignment of the carbon layers in the AB stacked graphene, where the areas of the low electron density, which are in the middle of the benzenoid ring, are over the areas of the high electron density that are at the edges of the benzenoid rings, as shown in Fig 2.b that was depicted by Hao et al. [32] in 2010. Therefore, there is a lot of electrostatic interaction between these stacked layers or sheets, which makes the exfoliation process of these sheets hard. On the contrary, the misalignment of turbostratic graphene layers (Fig 2.c), which were called few-layer graphene [32], makes it much easier for these layers to get separated. This is why FG is more manageable and could be easily applied in numerous applications. In a recent study for the Tour group that was led by Eddy et al. [33] to investigate the role of electric current on the FJH process, it was concluded that the formation of FG from a carbon material is not just a thermal process, but also an electrical process that depends on the electric current and its different parameters. The obtained results indicated that the associated electric field in a carbon source acts as a catalyst by lowering the activation energy required for the reaction of graphene formation [33]. Additionally, by adjusting the current or pulse width, it is possible to control the material's transformation through the two main processes. Firstly, the conversion of amorphous carbon to turbostratic or FG, and secondly, the conversion of turbostratic graphene to ordered graphene structures such as the AB stacking and graphite [33].

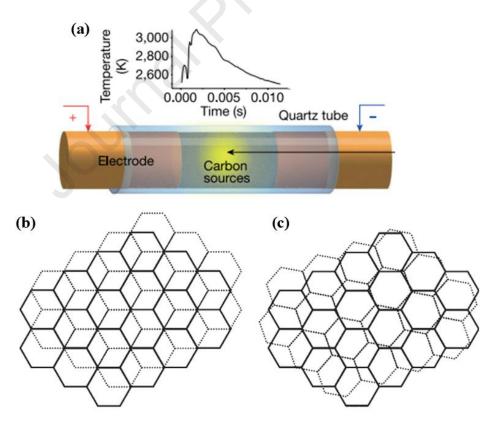


Fig 2. A schematic diagram of the quartz tube in which the FJH process takes place (bottom) and the relation between time in seconds and temperature in K (top) is presented in the insert image to indicate the very rapid increase in temperature in the FJH system (a). a

and the inserted image are reproduced from [13] with permission from Springer Nature,
Copyright [2020]. Top view of AB stacked graphene layers (b) and turbostratic (flash)
graphene layers (c). b and c are reproduced from [32] with permission from Wiley, Copyright
[2010].

The flash annealing process also referred to as the DC-Joule heating method, involves the rapid release of thermal energy into the material by passing an electric current through the sample, and it was used by Murakami and Villas-Boas [34] to produce nanocrystalline magnetic materials in 1999. Within the same context, the FJH process was reported in 2007 by Fujita et al. [35] who used this technique to produce a multi-walled carbon nanotube from an amorphous carbon pillar through flash discharging of gallium droplets, which were incorporated into the carbon pillar to induce a tunnel formation. This tunnel formation is what causes the carbon nanotube to form. Three years later, FJH was used by Cologna et al. [36] to flash sinter nanograin zirconia in a few seconds at 850 °C to produce dense ceramics.

## 2.1. Automation of FJH and scaling up

As the FJH technique advances, efforts are underway to automate the process for industrial-scale applications. Automation involves incorporating programmable systems to regulate voltage, current, and material input, ensuring consistent and efficient production of FG. Researchers are also enhancing electrode designs and employing robotic arms to manage precursor materials, enabling continuous operation and scaling production for commercial use.

Automation is key to transitioning FJH from small-scale lab experiments to industrial applications. It allows for faster production, improved graphene quality control, and more efficient energy usage, making it a practical solution for large-scale graphene production from waste materials. Companies and research institutions are now focusing on integrating FJH into automated systems to support large-scale waste management and material production. In this context, an integrated automatic system was developed by Zhu et al. [26] to replace the manual operational parts for loading and unloading the samples with robot arms as shown in **Fig 3**. To further enhance the mechanical control of this system, an imputation algorithm based on locking the coordinates of the first and last samples onto a sample tray was used. Regarding the electrical design, the cyclic discharge of alternating current (AC) and DC using an AC contactor to cut off the DC discharge circuit during AC-FJH was employed to avoid any breakdown issues of the capacitors responsible for the

discharge of DC. Consequently, this system successfully produced FG with a production rate of 21.6 g per hour upon adjusting the mechanical and electrical components [26]. Through a comparison between sawdust as a low-impurity and rice straw as a high-impurity carbon source material, that study [26] concluded that the purity of the carbon does affect the yield and the quality of produced FG.



**Fig 3**. A photograph of a pilot-scale fabrication device of FG adapted from [26] under the creative commons license CC BY 4.0.

The production scale of FG is constrained by various factors, such as the energy capacity of the FJH system, the rate at which samples are loaded and unloaded, and the decrease in product uniformity as the reaction tube's volume increases [37]. The continuous research and the dedicated workers of Professor James Tour's group, who put a lot of effort into advancing the FJH technique by improving the flashing capacity of the reactor and enhancing the samples loading system, played an essential role in doubling the production rate of FG every two weeks since they first started to use it in 2018. In this regard, Eddy et al. [37] automated the FJH system (**Fig 4**) using an electric system of pulse width modulation (PWM) to produce 1.1 kg of FG from coal-based metallurgical coke in just 90 minutes. PWM was used to cut the continuous discharge of the capacitor banks into sections *via* the rapid on-and-off switching using transistors mounted on each capacitor bank. Using the PWM resulted in a peak current of less than 1000 A. Although this reduced peak current compared to a normal DC flash (> 1000 A) decreased the flashing time, it helped in minimizing the

probability of equipment failure along with increasing the energy efficiency and yield. Upon comparing the characteristics and the quality of FG produced by DC flashing at a higher temperature (3000 °C) and shorter time (half a second) and that of the PWM-modified system produced at a lower temperature (1800 °C) and longer time (around two seconds), it was concluded that both products were similar. Furthermore, the PWM-modified system resulted in a higher consistent quality of the produced FG. Consequently, consecutive flashing cycles consistently producing high-quality FG were allowed using this modified system.



Upon successfully producing 1.1 Kg of FG per day, the Tour group was able to scale the production rate to more than 10 Kg and even up to 1 ton of FG per day [38]. Such an enhancement was achieved by using a variable frequency drive to program the heating and

that holds the sample in a quartz reaction tube before flashing, the second component is

where the flashing chamber stands, the third one is the 3D printer to print the plastic flashing

stand that is responsible for loading, flashing, and unloading of the reaction vessels, the

fourth one is the ramp that is used to collect the reacted quartz tube after flashing and

unloading, the fifth one is the basin where the flashed samples are collected, the sixth one is

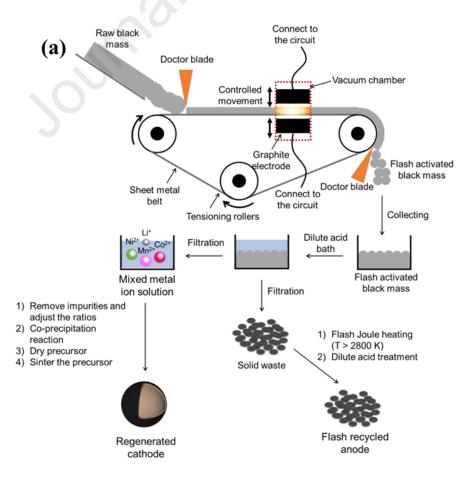
the controlling system for loading and unloading samples, the seventh one is the part

connecting the system to the capacitor banks, and the last one is just an air hose to cool the

3D printer system. This figure is reproduced from [37] with permission from Wiley,

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cooling sequence to have better control over the electrothermal reaction. Subsequently, the team developed a conceptual model to convert cathode and anode waste materials of lithiumion batteries into FG using the design shown in Fig 5.a. Through a few steps, starting with FJH of the battery waste black material, followed by acid leaching and filtration, they managed to get a recycled anode material and an acidic solution containing various metals that could be used to regenerate the cathode material. This design makes it easier to recover most of the metal components of batteries with a recovery percentage of up to 98%, which is better than the direct hydrochloric acid leaching along with reducing energy by 26%, water consumption by 52%, the amount of acid by 59%, and CO<sub>2</sub> emissions by 38% according to their life cycle analysis. The Tour group aspires to produce 100 tons of FG per day by 2024, which would require even higher temperatures than 3000 K and energy densities of about 3600 J/g that are larger than the density used for lithium-ion batteries (about 840 J/g) [38]. However, the group has not achieved this goal yet. Liquid-phase extraction could help achieve this goal of mass production, but the dispersion solvents could be expensive and not environmentally friendly along with cleaning to remove the residuals that can affect the graphene quality [39]. Therefore, further research is required to achieve the goal of 100 tons per day.



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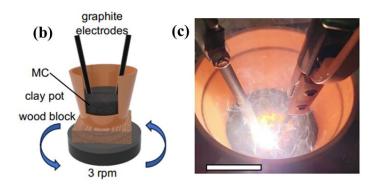
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approximately 750 grams of metallurgical coke in the rotating clay pot with a rotating speed

of 3 rpm (c). b and c are adapted from [40] under the creative commons license CC BY NC

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commercial arc welders starting from only 120 US dollars very recently to substitute

programmable power supplies in conventional FJH systems [40]. These arc welders offer

significant advantages, including low cost, user-friendly operation, compact design, excellent

control over temperature and tunability, and they can be operated swiftly with a minimal

training requirement. The arc welder's power source can be adjusted for mobility. Instead of

relying on a fixed power outlet, a portable system is used to power an arc welder. In this

setup, a car battery is charged using solar panels, creating a mobile FJH system that operates

entirely on solar energy. This solar-powered system was assembled for only 422 US dollars

[40]. A kilogram-scale production of FG from metallurgical coke was carried out using an

Amico Electric ARC-200DC arc welder that costs about 180 US dollars, as illustrated in Fig.

5.b and c. The setup included connecting the arc welder to two graphite rods, which were

placed inside a clay flowerpot with a volume of 13 L. The pot was filled with 500 grams of

MC (material) and left open to the air during the reaction. This pot was placed on a rotating

stand with a speed of 3 rpm to ensure uniform distribution of heat throughout the sample. The

production rate of that system was three kg of FG per hour, accompanied by a very low

production cost compared to the conventional FJH system. The production rate of FG can be

Continuing with the automation and scaling-up of FG production, the Tour group used

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Fig 5. A conceptual design has been proposed for a continuous FJH reactor, along arc welder (b), and a photograph captures the arc welder in operation, processing

with subsequent processing steps to produce regenerated cathodes and flash-recycled anodes (a) adapted from [38] under the creative commons license CC BY 4.0. A schematic illustration shows a cross-section of a clay pot filled with metallurgical coke supported on a rotating stand in a kilogram-scale FJH reaction that was conducted using an Amico Electric

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significantly increased by operating multiple arc welders simultaneously. This can be achieved by connecting them in series to boost the maximum output voltage, or in parallel to raise the maximum output current. This approach is up to 30 times more cost-effective compared to acquiring larger power supplies, making it a highly efficient and economical method for scaling up graphene production.

#### 2.2. FJH Feedstocks

The scalability and energy efficiency of FJH make it a promising solution for sustainable waste management. By reducing environmental harm and enabling the recovery of valuable resources, FJH supports the principles of a circular economy. FJH can process a variety of feedstocks, including inexpensive carbon-based materials such as electronic waste (e-waste), biomass waste, rubber waste, coal, petroleum coke, asphalt, plastics, and even CO<sub>2</sub> (**Fig 6**) [41]. Therefore, FJH offers a sustainable pathway for both of waste valorization and resource recovery.

The proliferation of electronic devices has led to a surge in electronic waste (e-waste), with over 50 million tons generated annually, but less than 20% is properly recycled. E-waste contains valuable metals like gold, silver, and copper, as well as toxic substances such as lead and mercury, posing significant environmental and health risks when improperly disposed of. Traditional e-waste recycling is complex and costly due to the intricate composition of electronics [42]. FJH offers an efficient solution by recovering valuable metals such as gold, silver, and copper from e-waste by vaporizing non-carbon elements and collecting these vapors afterward while converting carbon-based components into graphene [43]. FJH is more energy-efficient than conventional methods and reduces the need for chemical treatments, offering a dual benefit of metal recovery and carbon conversion in an environmentally friendly way.

Biomass waste, including agricultural residues, forestry by-products, and organic municipal waste, represents a vast and underutilized resource. Globally, billions of tons of biomass waste are produced each year, much of which is either burned or left to decompose, releasing significant amounts of greenhouse gases such as CO<sub>2</sub> and methane [44]. Biomass is an abundant carbon source, making it an ideal candidate for graphene synthesis. The FJH process subjects biomass to rapid and intense heating, which allows the carbon atoms in the biomass to reorganize into a turbostratic graphene structure. Biomass offers a renewable source for graphene production, making the process attractive from a sustainability

perspective. Moreover, the advantage of this process lies in its ability to reduce landfill waste while producing high-value carbon products. It also mitigates greenhouse gas emissions from decomposing food waste and supports circular economy goals by transforming organic waste into useful materials [26].

FJH offers an efficient and eco-friendly method to convert rubber waste into high-quality turbostratic FG. Rubber waste, such as tires, presents a significant environmental challenge, with most being burned or sent to landfills. Rubber is highly durable and resistant to microbial degradation, meaning waste tires can persist in the environment for centuries. However, there are some recent promising results about using fungi such as *Botryotinia cinerea* in degrading rubber, and further studies are urgently required [45]. The accumulation of rubber waste not only takes up vast amounts of landfill space but also poses fire hazards and can leach harmful chemicals into the soil and water. Additionally, FJH avoids the use of solvents, making it more sustainable. FJH provides a solution to the environmental challenge of managing non-biodegradable rubber waste, recycling it into valuable materials, and preventing the release of harmful chemicals and gases that usually occurs during conventional disposal methods like incineration [46].

Coal, petroleum coke, and asphalt are major contributors to industrial production and environmental pollution. Waste from these materials often accumulates through industrial processes, road construction, and energy production. Coal and petroleum coke combustion releases high concentrations of CO<sub>2</sub>, SO<sub>2</sub>, and other harmful pollutants, contributing to climate change and worsening air quality. However, these pollutants can be used as feedstocks to produce graphene by FJH. Coal, particularly anthracite, is an ideal feedstock due to its high carbon concentration and global availability. Petroleum coke, a byproduct of oil refining, is another prime candidate due to its high carbon purity and low cost. Asphalt, a byproduct of crude oil distillation used in road construction, can be converted into high-quality graphene through FJH, offering a sustainable route for recycling industrial waste. The versatility of FJH in using coal and these petroleum derivatives as feedstocks makes graphene production cost-effective and environmentally friendly, producing graphene with few defects and high purity, suitable for energy storage, composites, and electronics [13, 47].

CO<sub>2</sub> is a major greenhouse gas responsible for climate change, with over 35 billion tons emitted annually from human activities like fossil fuel combustion. This excess CO<sub>2</sub> leads to global warming, ocean acidification, and environmental degradation. While efforts to

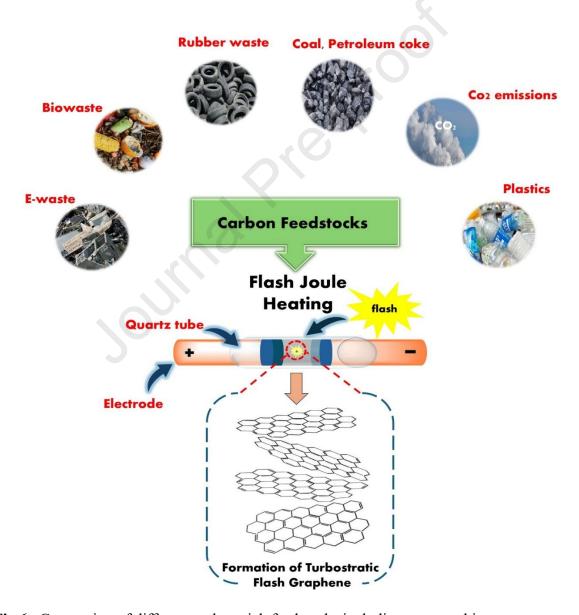
reduce emissions continue, innovative solutions are needed [48]. One such approach is using CO<sub>2</sub> as a feedstock for FG production through FJH. CO<sub>2</sub> can be entrapped and processed into solid carbon through processes like molten carbonate electrolysis, which transforms CO<sub>2</sub> into amorphous carbon. This amorphous carbon can then undergo FJH. Therefore, FJH can indirectly transform CO<sub>2</sub> emissions into graphene and offers a sustainable solution to climate challenges [49].

Plastics are currently a vital part of modern life, with their versatility and low cost driving their widespread use in packaging, consumer products, construction, and beyond. However, the rapid increase in plastic production has led to severe environmental challenges [50]. Globally, PW production surpasses 300 million tons annually, and only a small fraction is effectively recycled, while the rest ends up in landfills, incinerators, or the natural environment, especially oceans. The durability of plastics, which makes them useful, also means they can remain in nature for hundreds of years, contributing to microplastic contamination and environmental destruction. Traditional recycling methods are often inefficient or impractical for many plastic types, and incineration releases harmful greenhouse gases and toxic chemicals, exacerbating environmental issues [51].

FJH presents a promising solution to the growing PW problem. Unlike conventional recycling processes, which are energy-intensive and limited by plastic types, FJH can transform a wide range of plastics, including those that are difficult to recycle, such as polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET). One of the advantages of using plastics as a feedstock for FG is the high carbon content in many common plastics, which makes them ideal candidates for graphene production. Additionally, the process produces useful byproducts such as hydrogen gas, which can be captured and repurposed as a clean energy source. Hydrogen, a zero-emission fuel, represents a valuable resource in the transition to sustainable energy systems, adding a further layer of sustainability to the FJH process. Therefore, FJH offers the dual benefit of producing clean hydrogen energy and generating valuable FG, which has numerous environmental applications [14].

Notably, to initiate the FJH process for converting many of the above-mentioned materials into graphene, a conductive agent like carbon black is usually required. Materials, such as e-waste, biomass waste, rubber waste, asphalt, and plastics have high resistance. The addition of carbon black facilitates the rapid passage of electrical current, enabling the

material to reach the high temperatures necessary for the FJH process [13, 21]. Achieving these high temperatures using the conductive agent helps trigger the breakdown of the waste's molecular structure, allowing the carbon atoms to reconfigure into turbostratic graphene. Without this conductive aid, the high resistance of these waste materials would prevent the effective generation of the required heat for the FJH reaction. This is in line with the observation of Saadi et al. [23] who reduced the resistance from 64 k $\Omega$  to 60  $\Omega$  by increasing the loading of the conductive agent, which was an asphaltene-sourced FG, from 20 wt% to 50 wt% upon converting a 500 mg sample of asphaltene into FG.



**Fig 6**. Conversion of different carbon-rich feedstocks including e-waste, biomass waste, rubber waste, coal, petroleum coke, asphalt, CO<sub>2</sub>, and PW into valuable turbostratic FG.

#### 2.3. Advantages of FG compared to conventional graphene

FG offers several advantages, as it is non-toxic, requires no water or solvents, and can be made of different types of carbon feedstock such as plastics, e-waste, biomass waste, rubber waste, coal, and asphalt, and processed into other products. Additionally, these products can be re-flashed at the end of their life cycle to produce FG that can be used again in other industries. In general, graphene is a very good way to store carbon since it is merely bio-degraded by microorganisms as it is for graphite, so it is very slow to enter the carbon cycle [28]. Therefore, it is considered an excellent alternative to storing carbon in other forms, which would eventually decompose into CO<sub>2</sub>, further exacerbating the global issue of CO<sub>2</sub> emissions. A ton of FG made of coal is estimated to cost about 130-135 US dollars since a ton of coal costs 100 US dollars and the electricity required to convert that coal into FG costs about 30-35 US dollars, according to Prof James Tour [28]. There is a huge difference upon comparing these numbers with the average cost of conventional graphene, that ranges from 60000 to 100000 US dollars, according to Prof James Tour as well [28].

Conventional production methods of graphene, such as CVD, mechanical exfoliation, and chemical oxidation of graphite followed by chemical or thermal reduction of graphene oxide, usually release large amounts of CO<sub>2</sub> emissions, as outlined in **Table 1**. For instance, one gram of graphene produced by chemical reduction of graphene oxide releases about 146 g of CO<sub>2</sub>, and 86 g of CO<sub>2</sub> were released from producing one gram of graphene by the thermal reduction of graphene oxide [52]. CO<sub>2</sub> emissions released from the conventional synthesis of graphene can be up to 407 g per just one gram of graphene [53]. In a recent study that was carried out by Jia et al. [53] to produce FG from four different waste materials, 9.88, 6.56, 8.94, and 6.95 g of greenhouse gas emissions, which are mainly CO<sub>2</sub> and CH<sub>4</sub>, were released from the FJH of sawdust, wheat straw, corn straw, and rice straw, respectively. Within the same context, Zhu et al. [26] managed to reduce CO<sub>2</sub> emissions to just 1.9 g per gram of FG produced from rice straw-based biochar. Moreover, if the electricity grid is powered by renewable energy, FG systems could potentially attain carbon neutral [53] or even carbon-negative outcomes due to the reduction in fossil fuel usage [26].

Graphene is often produced at high temperatures (up to 1000 °C) by conventional methods such as CVD over a long time (more than 10 minutes) [54]. On the other hand, FG only needs milliseconds to get formed. FG can be produced from different waste materials, but conventional graphene is usually sourced from specific carbon sources such as graphite

and gaseous carbon materials like methane and acetylene [55]. Milliseconds are required to produce FG, whereas the synthesis of conventional graphene is done over several minutes and even hours, such as the mechanical exfoliation of graphene by ball milling which can take 24-48 hours [55]. This long synthesis time represents a major hindrance to scaling up conventional graphene synthesis.

**Table 1**. A comparison between FG and conventionally produced graphene.

	FJH-produced Graphene	Conventionally Produced Graphene (e.g., CVD, Mechanical Exfoliation)	References
CO <sub>2</sub> Emissions	Low CO <sub>2</sub> emissions (~10 kg CO <sub>2</sub> /kg graphene) and could be less than that	High CO <sub>2</sub> emissions (~400-500 kg CO <sub>2</sub> /kg graphene)	[53]
Feedstock	Uses any carbon-based waste material such as plastics, batteries, biomass, and food waste	Requires purified carbon sources like methane or graphite	[13, 55]
Energy Consumption	Low energy consumption (due to rapid heating in milliseconds)	High energy consumption (CVD requires continuous heating at ~1000 °C)	[13, 54]
Production Time	Short production time (graphene is produced in the range of milliseconds to a few seconds)	Long production time (CVD can take minutes to hours; exfoliation is slower and less scalable)	[54, 55]
Scalability	Good scalability (can use large quantities of waste material) as it currently produces kilograms of graphene per day	Limited scalability (CVD and exfoliation are not easily scalable for industry)	[38, 54]
Production	Low production cost (estimated cost is significantly	High production cost (CVD systems and purified feedstocks	

	FJH-produced Graphene	Conventionally Produced Graphene (e.g., CVD, Mechanical Exfoliation)	References
Cyanhana	cheaper due to feedstock) as it could be around 130-135 US dollars per ton  Produces turbostratic graphene	are expensive) as it could be 60000 to 100000 US dollars per ton  Produces more crystalline	[28]
Graphene Quality	(less ordered than CVD graphene)	graphene with high quality, especially with CVD	[13]
Sustainability	Sustainable, environmentally friendly (recycles waste into high-value graphene), and can be carbon-neutral or negative if the electricity is sourced from renewable energy	Less sustainable due to high energy use and reliance on non-renewable feedstocks	[26, 53]
Application	Suited for various applications like water treatment, energy storage, concrete and cement production, and applications requiring good purity if the graphene is sourced from a good-quality carbon source	Suited for high-performance electronics and nano-devices requiring high purity and precise control over thickness and composition	[15]

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# 2.4. Characterization of FG

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One of the most important studies that reported the characterization of FG is the one carried out by Luong et al. [13]. In this study, different carbon-based materials including carbon black, anthracite coal, calcined coke, and used coffee were used to produce FG, and the highest yield (90%) was obtained from carbon black. The Raman mapping of carbon black-based FG showed a very high  $I_{2D/G}$  that was 17 (Fig 7.a), where the area of each pixel in the Raman map was 4  $\mu$ m<sup>2</sup> by a 50× magnification. This  $I_{2D/G}$  value is believed to be the

highest reported value so far compared to all other previously produced graphene. The X-ray diffraction (XRD) pattern of FG reveals a prominent (002) peak (**Fig 7.b**), indicating effective graphitization of the amorphous carbon black. This peak corresponds to an interlayer spacing of 3.45 Å, which is slightly larger than the interlayer spacing in traditional AB-stacked graphite (3.37 Å). This increased spacing further verifies the formation of expanded and turbostratic graphene. Folded graphene layers were observed by high-resolution transmission electron microscopy (HRTEM) as shown in **Fig 7.c**.

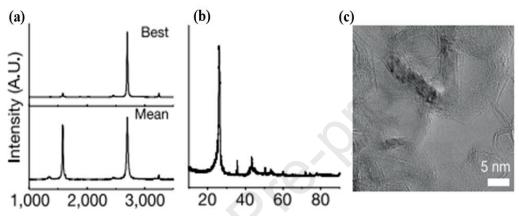
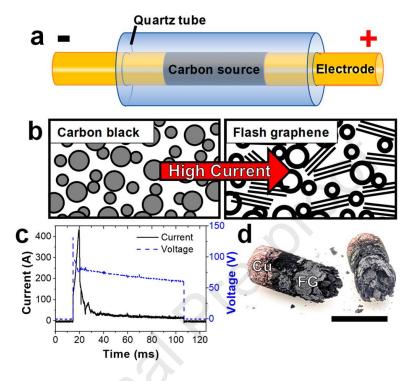


Fig 7. The Raman spectrum (a), XRD pattern (b), and HRTEM image (c) of carbon black-based FG. a-c are reproduced from [13] with permission from the American Chemical

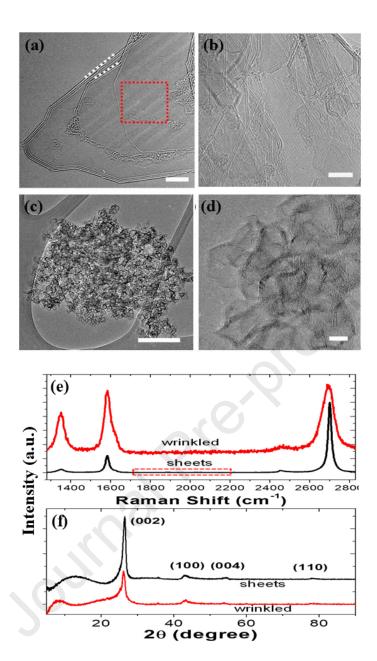
based FG. a-c are reproduced from [13] with permission from the American Chemical Society, Copyright [2020].

Eight months later, the same research group (Tour group) further investigated the characterization of carbon black-FG in a study led by Stanford et al. [30]. In that study, they used the same quartz tube (**Fig 8.a**) of the FJH system in their first paper to produce FG within 100 milliseconds at about 3000 K. As shown in **Fig 8.b**, the carbon black was converted into layers of FG along with small graphitic carbon particles. The main driver of the FJH process is the high and very rapid current that passes through the carbon black, which could be higher than 400 amp in just 20 milliseconds (**Fig 8.c**). **Fig 8.d** further clarifies the difference between the FG layers/sheets which are grey in color and the small graphitic particles which are black colored. To further differentiate between the grey FG layers and the small graphitic dots, TEM analysis was undertaken, and the images showed the sheet-like morphology of grey FG (**Fig 9.a** and **b**), whereas the images of the black graphitic dots showed bent or wrinkled graphene (**Fig 9.c** and **d**). Moreover, the Raman spectrum indicated a higher  $I_{\text{2D/G}}$  ratio for the FG sheets (1.8 up to 2) compared to that of the wrinkled graphene (1.1) as shown in **Fig 9.e**. In consistency with all these results, the intensity of the (002) peak

is higher in the XRD pattern of the FG sheets as shown in **Fig 9.f.** Additionally, the authors found that the  $I_{\text{2D/G}}$  ratio decreases by increasing the flashing time over 200 milliseconds, which results in the transformation of FG sheets into AB stacking and then into the graphite structure.



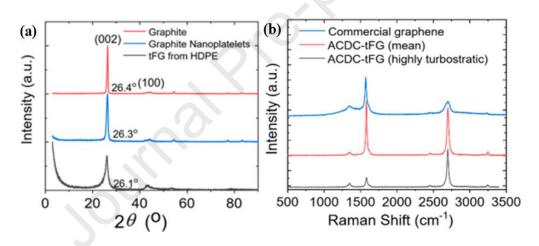
**Fig 8.** The quartz tube used for the FJH reaction (a), a schematic representation of the conversion of carbon black into FG sheets and small graphitic particles (b), the induced current and voltage during the FJH reaction over a very short time in milliseconds (c), and a photograph of the resultant grey and black graphene as well as the copper wool used in the FJH reaction (Scale bar of this photograph is 5 mm) (d). a-d are reproduced from [30] with permission from the American Chemical Society, Copyright [2020].

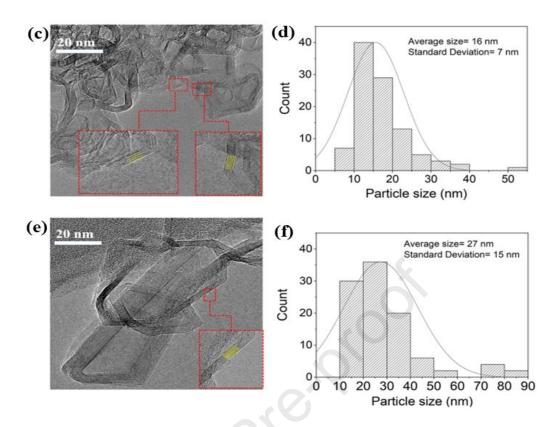


**Fig 9.** TEM images of FG sheets with a scale bar of 5 nm (**a** and **b**), a TEM image with a scale bar of 200 nm (**c**), and an HRTEM image of wrinkled graphene with a scale bar of 10 nm (**d**), the XRD pattern (**e**), and the Raman spectra (**f**) of both types of graphene. a-f are reproduced from [30] with permission from the American Chemical Society, Copyright [2020].

In another study for the same group that was led by Algozeeb et al. [16], high-density polyethylene, which is a very common plastic material, was converted into FG by two types of FJH treatment: the first one using only AC and the other one using AC followed by DC. Comparing the XRD pattern of ACDC-FG with the patterns of graphite and graphite platelets in **Fig 10.a**, it is clear that the (002) peak of ACDC-FG was detected at a lower 2θ value

(26.1) which is in agreement with the value of carbon black-based graphene [13] that is mentioned in the previous paragraph. The Raman spectrum of the highly turbostratic ACDC-FG showed a high  $I_{2D/G}$  ratio compared to that of commercial graphene (**Fig 10.b**) as it ranged from 1 to 6. On the contrary, the  $I_{2D/G}$  ratio of AC-FG (data is not shown) was only in the range of 0.5 to 1.2. Such a result indicated the importance of combining the AC and DC treatment steps as the AC treatment allows the FJH to extend for 8 seconds, which will help subliming the necessary evaporites and produce a low-quality AC-FG. Subsequently, the quality of AC-graphene will be improved by conducting the DC treatment. In consistency with these results, the TEM image of AC-FG shows highly graphitic layers (**Fig 10.c**) with an average particle size of 16 nm (**Fig 10.d**) and the image of ACDC-FG shows turbostratic layers (**Fig 10.e**) with an average particle size of 27 nm (**Fig 10.f**). The increased sheet size of ACDC-FG compared to that of AC-FG agrees with the decrease in the D band of ACDC-FG and its high  $I_{2D/G}$  ratio in the Raman spectra.





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Fig 10. XRD patterns of graphite, graphite platelets, and ACDC-FG (a), Raman spectra of commercial graphene and ACDC-FG (mean and highly turbostratic) (b), a TEM image (c) and particle size analysis of AC-FG (d), and a TEM image (e) and particle size analysis of ACDC-FG (f). a-f are reproduced from [16] with permission from the American Chemical Society, Copyright [2020].

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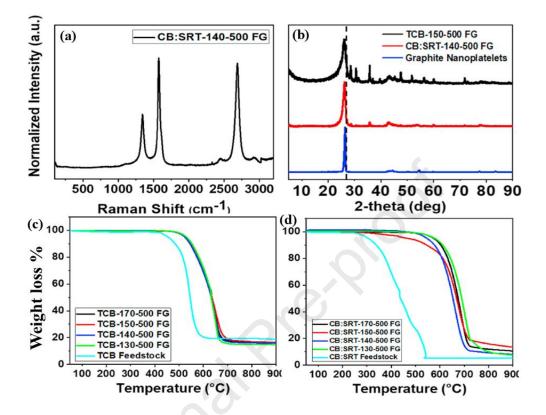
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In another work, rubber waste was used by Advincula et al. [19] to produce FG with I<sub>2D/G</sub> ratios of 0.75 and 0.83 from tire-derived carbon black (TCB) and carbon black with shredded rubber tires (CB:SRT), respectively. Fig 11.a shows the Raman spectrum of TCB in which the three main Raman peaks appear including the D band at about 1350 cm<sup>-1</sup>, the G band at around 1580 cm<sup>-1</sup>, and 2D nearly at 2700 cm<sup>-1</sup>. These values are observed with almost all FG samples such as the sample that was produced along with silicon carbide from fiber-reinforced plastics [56]. The authors investigated different factors such as the pulsing voltage and the pulsing time on the quality of the FG and they found out that the intermediate pulsing voltage (140-150 V) and time (500 milliseconds) are necessary to produce highquality FG. Low values of these parameters resulted in a lower conversion rate of the carbon source to FG and the high values resulted in the AB stacking of the FG layers. The XRD pattern (Fig 11.b) showed that the Full Width Half Maximum of the (002) peak for TCB-FG **CB:SRT-FG** increased, relative indicating and to graphite nanoplatelets,

smaller crystallite size of graphene layers. Thermal gravimetric analysis (TGA) confirmed the higher thermal stability of the two FG products, with degradation beginning at higher temperatures, compared to their carbon source materials as shown in **Fig 11.c** and **d**.



**Fig 11**. Raman spectrum of CB:SRT-FG (a), XRD pattern of graphite nanoplatelets, TCB-FG, and CB:SRT-FG (b), TGA of TCB feedstock and its TCB-FG (c), and CB-SRT feedstock and its CB:SRT-FG at different pulsing voltages (d). a-d are reproduced from [19] with permission from Elsevier, Copyright [2021].

#### 3. Applications of FG

#### 3.1. Water Treatment

Graphene's unique physical and chemical characteristics make it a viable material for various applications including water purification. Its high surface area removes numerous pollutants, such as toxic metals, organic pollutants, and even microorganisms, while its mechanical strength and chemical stability ensure durability in filtration systems [57]. Graphene's selective permeability and hydrophobicity allow for efficient water purification, and its conductivity enhances electrochemical processes like capacitive deionization for salt removal. Additionally, graphene can be functionalized or combined with other nanomaterials, making it versatile in addressing a broad spectrum of water quality challenges [58]. Despite graphene's potential, traditional production methods have hindered its widespread adoption,

particularly in water treatment because of their high cost and energy requirement, and low yield.

FJH is a more recent and innovative approach, providing a cost-effective, scalable, and environmentally friendly way for FG production. FG offers superior dispersion and structural advantages over traditional graphene. Unlike regular graphene, which tends to aggregate and stack due to van der Waals forces, FG's unique production process not only results in a highly porous structure with fewer defects and wrinkles but also promotes a turbostratic arrangement of graphene sheets. This rotational misalignment reduces electrostatic forces and increases interplanar spacing. Furthermore, minimizing the stacking tendencies of graphene sheets by weakening van der Waals attractions and enabling better dispersion in solvents or polymers [59]. Such enhanced dispersion is particularly beneficial in applications requiring high uniformity, such as water treatment. Additionally, the increased interplanar distance and reduced sheet interactions significantly enhance FG's adsorption capabilities. Moreover, the higher specific surface area allows for more active sites, while the reduced interaction energy facilitates easier access for adsorbates or chemical reagents.

Liao et al. [60] explored the ultrafast synthesis of 3D porous FG (3D PFG) using FJH. The process involves using bituminous coal as a carbon source, which is subjected to two stages of FJH, resulting in a 3D porous structure in just 21 seconds. In the first stage, bituminous coal (BC) was subjected to continuous discharge for 20 seconds, increasing the temperature to 1200 K, resulting in porous amorphous carbon (PAC) with a surface area of 439 m<sup>2</sup>/g. In the second stage, the PAC was exposed to a higher voltage (140V), reaching temperatures above 2500 K, which caused rapid thermal expansion. This transformed the material into 3D porous FG (3D PFG) with a 504 m<sup>2</sup>/g surface area. Moreover, this study investigates how varying the carbonate ratios and flash voltages during the FJH process influenced the pore formation in 3D PFG. By adjusting these parameters, they were able to control the hierarchical porous structure, which consists of micropores, mesopores, and macropores. The optimal carbonate ratio of 3:1 (K<sub>2</sub>CO<sub>3</sub> to bituminous coal) resulted in a uniform reaction, producing a highly porous material with a surface area of 504 m<sup>2</sup>/g. Higher carbonate ratios led to pore collapse and reduced porosity, while lower ratios resulted in incomplete activation. Similarly, the flash voltage played a crucial role in the synthesis process. At 140V, the FJH system achieved a high enough temperature (>2500K) to cause rapid thermal expansion and activate the porous structure, optimizing the material's

adsorption properties. Lower voltages did not generate sufficient heat to fully activate the pores, while higher voltages caused structural damage due to excessive thermal expansion.

The dispersion test conducted on BC, PAC, and 3D PFG further highlighted the advantages of 3D PFG. When dispersed in a 1% F-127 solution and sonicated for 20 minutes, 3D PFG remained well-dispersed even after 5 days, demonstrating superior stability. In contrast, PAC showed a lower degree of dispersion, and BC showed the worst performance with significant aggregation over time as shown in **Fig 12.a**. The residual concentration of BC, PAC, and 3D PFG upon centrifugation confirmed the excellent performance of 3D PFG (**Fig 12.b**). The excellent dispersibility of 3D PFG is attributed to its hierarchical porous structure and the presence of oxygen-containing functional groups, which improve its interaction with the surrounding medium. This makes 3D PFG particularly suitable for water treatment applications, where stable dispersions are critical for effective pollutant adsorption.

above-mentioned study also demonstrated the exceptional adsorption The performance of the 3D PFG, particularly for removing organic dyes such as methylene blue (MB) and methyl orange (MO) from water. The initial dye concentration was 200 mg/L for both MB and MO. The 3D PFG achieved an 80% removal efficiency for MB within the first few minutes, showcasing its high adsorption efficiency (Fig 12.c and d). Similarly, the 3D PFG reached a 95% removal rate for MO in a short period, further highlighting the material's effectiveness in water purification. Furthermore, the dyes were completely removed within 128 mins. The maximum adsorption capacity (qm) for MB was 368.10 mg/g, while for MO, the capacity was 150.64 mg/g. These high adsorption capabilities are attributed to the welldeveloped hierarchical pore structure of the 3D PFG, which includes micropores, mesopores, and macropores, providing a large surface area of 504 m<sup>2</sup>/g. This structure provides a large surface area for adsorption and enhances the diffusion of pollutants into the porous material. Additionally, functional groups on the graphene surface promote electrostatic attraction, surface complexation, and hydrogen bonding, and enhance the adsorption process. These findings suggest that 3D PFG, synthesized under optimized conditions, could serve as an efficient and scalable solution for wastewater treatment and environmental remediation.

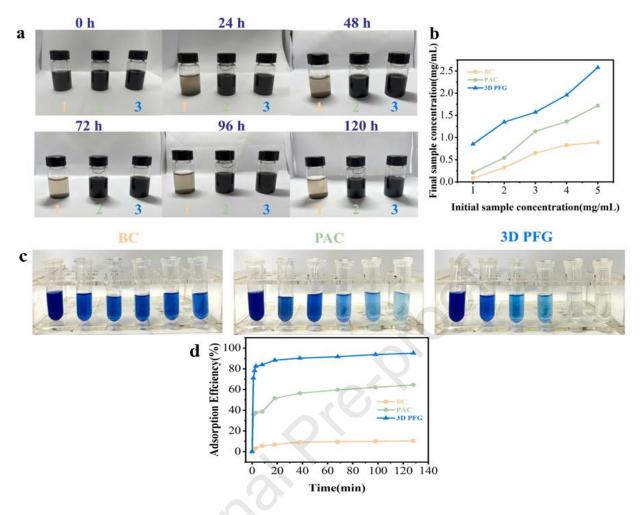


Fig 12. Photos showing a comparison of the dispersion behavior of various samples in solution over 120 hours (a). The samples, labelled 1, 2, and 3, correspond to BC, PAC, and 3D PFG, respectively. The variation in residual concentration of BC, PAC, and 3D PFG following centrifugation at different starting concentrations (b). Photos showing the adsorption of MB by BC, PAC, and 3D PFG samples over 128 minutes (c). The adsorption efficiency of BC, PAC, and 3D PFG over 128 minutes (d). a-d are reproduced from [60] with permission from Elsevier, Copyright [2023].

## 3.2. High-efficiency supercapacitors for energy storage

The increasing global demand for energy storage technologies has driven considerable interest in advanced materials for applications like supercapacitors, which can bridge the gap between traditional batteries and capacitors. Supercapacitors offer rapid charge-discharge cycles, high power density, and long-term stability, but their performance is heavily influenced by the materials used in electrode fabrication. Among various materials, carbon-based nanostructures, particularly graphene, have gained significant attention due to their remarkable electrical, mechanical, and chemical properties [61].

For supercapacitor applications, the graphene produced *via* FJH offers several advantages. The high surface area enables efficient ion adsorption at the electrode-electrolyte interface, enhancing the electric double-layer capacitance mechanism. Additionally, FJH-graphene's few-layered structure and high conductivity ensure fast electron and ion transport, reducing internal resistance and improving overall electrochemical performance. This results in supercapacitors with higher energy and power densities, as well as superior cycling stability, making them ideal for next-generation energy storage devices [62].

Mathan et al. [62] investigated the quick synthesis of graphene-based materials utilizing FJH to create high-performance supercapacitor electrodes. In their study, urea and citric acid are used as precursors for carbon nanodots (CNDs) synthesis. These CNDs were deposited onto flexible carbon substrates and subjected to FJH, where short, high-intensity electrical pulses transformed the CNDs into few-layered graphene within seconds. The study explored how varying flash currents influenced the material's electrochemical properties. Electrodes flashed at higher currents (100 Amperes) showed better electrochemical performance, with specific capacitance values reaching 279 F.g<sup>-1</sup> at 1 A.g<sup>-1</sup>. In contrast, electrodes flashed at lower currents (50 Amperes) and un-flashed electrodes demonstrated drastically lower capacitance by providing 80.9 F.g<sup>-1</sup> and 2.3 F.g<sup>-1</sup>, respectively. The superior performance of the high-current electrodes was attributed to the formation of well-ordered, few-layered graphene with enhanced conductivity and optimized ion transport properties.

Additionally, the researchers assembled a symmetric supercapacitor using the FJH-processed graphene electrodes, which achieved a specific capacitance of 106.5 F.g<sup>-1</sup> and an energy density of 44.4 Wh.kg<sup>-1</sup> at the power of the density of 1000 W.kg<sup>-1</sup>. The supercapacitor also demonstrated impressive cycling stability, retaining 98.5% of its capacitance after 5000 charge-discharge cycles. These results highlight the potential of FJH as a rapid and scalable technique for producing high-performance graphene-based electrodes for energy storage applications, offering FJH-synthesized graphene as a promising material for next-generation supercapacitors.

Zhang et al. [63] proposed a novel method to enhance supercapacitor performance by utilizing FG as a photothermal coating. In their work, FG was synthesized *via* a rapid flash reduction process, transforming graphene oxide into highly conductive, porous graphene films within milliseconds. These films were then applied as photothermal absorbers on supercapacitor devices to elevate operating temperatures through solar irradiation, achieving

a rapid temperature rise above 50°C. This temperature increase significantly improved transport kinetics, resulting in a capacitance enhancement of up to 65%. The FG-based supercapacitor electrodes exhibited exceptional electrochemical performance, with a specific capacitance of 138 F.g<sup>-1</sup> and an energy density of 58.7 Wh.kg<sup>-1</sup> at a power density of 1.2 kW.kg<sup>-1</sup>. Numerical simulations revealed that the elevated temperature improved ion mobility and reduced electrolyte viscosity, enhancing double-layer capacitance and energy storage efficiency. Additionally, the FG-coated supercapacitors demonstrated remarkable cycling stability and effective retention of their enhanced performance even after solar irradiation was ceased, showcasing the thermal retention effects of the photothermal system. The study also emphasized the simplicity and universality of this approach, as the FG coating can be applied to various supercapacitor designs without altering their internal structures. This strategy offers a scalable and sustainable method for boosting supercapacitor performance, with potential applications extending beyond energy storage to areas such as solar steam generation and photothermal catalysis. These findings underscore the potential of FG as a versatile material for next-generation energy storage systems.

## 3.3. Environmentally friendly conductive inks

The progress in the conductive inks industry has evolved significantly due to innovations in materials science and fabrication techniques. Conductive inks are vital in the rapidly growing field of printed electronics, requiring materials that exhibit high electrical conductivity, mechanical flexibility, and stability across various environmental conditions. Traditional conductive inks typically rely on precious metals like silver and gold, which offer excellent conductivity but are expensive, resource-intensive, prone to oxidation, and can compromise the flexibility of devices. This has driven research toward alternative materials that can provide comparable or superior performance at reduced cost and environmental impact [64].

One promising development is the integration of FG into conductive inks, which presents numerous advantages over conventional metal-based options. FG's high electrical conductivity facilitates efficient electron transport, which is critical for applications in printed electronics. The large surface area of graphene, due to its two-dimensional structure, enhances the interaction between the ink and the substrate, leading to a more stable and conductive network. Furthermore, the mechanical flexibility and strength of graphene make it

well-suited for use in flexible and wearable electronics, whereas traditional metal-based inks can fail due to cracking or loss of conductivity under mechanical stress [65].

Damasceno et al. [65] conducted a comprehensive study on the development of a solvent-based conductive ink composed of carbon-black-derived FG (CBFG) and poly(omethoxy aniline) (POMA), to overcome the limitations of traditional metal-based conductive inks, such as high cost, rigidity, and oxidation issues. FJH, which involves rapid electrical discharge, results in highly pure CBFG with a large surface area, offering advantages in flexibility, cost, and environmental sustainability.

POMA, a conductive polymer, was chosen for its enhanced solubility in organic solvents and favorable electronic properties, including  $\pi$ – $\pi$  interactions, which help improve the ink's performance in electronic applications. The combination of CBFG and POMA provided a synergistic effect, improving electrical conductivity (EC) and other functional properties while addressing the drawbacks of metal nanoparticle-based inks. The study employed a factorial design of experiments to evaluate the effect of varying concentrations of CBFG, POMA, and binders on the ink's EC. The optimal ink composition, consisting of 40 mg.L<sup>-1</sup> CBFG, 2 mg.L<sup>-1</sup> POMA, and 4 mg.L<sup>-1</sup> binder, achieved an EC of 0.768 S.m<sup>-1</sup>, demonstrating high conductivity. This makes the ink particularly suitable for printed electronics, wearable sensors, and flexible circuits, where traditional metal-based inks fall short.

Various characterization techniques were employed to investigate the structural and morphological properties of the CBFG and POMA components. These techniques validated the successful production of high-quality CBFG, featuring turbostratic graphene layers that exhibit improved exfoliation, dispersion, and uniformity in inks. The turbostratic arrangement also preserves the material's two-dimensional characteristics, enhancing its conductivity.

Moreover, the study demonstrated that the synthesized ink had excellent adhesion and surface coverage when applied using conventional printing techniques such as doctor blade coating. This ability to form stable, high-performance films on different substrates makes the ink highly versatile.

Overall, this research presents a cost-effective, high-conductivity alternative to metalbased inks. Not only does the combination of CBFG and POMA provide a more sustainable solution, but also it ensures scalability for industrial applications in printed electronics, flexible sensors, and other wearable technologies. The findings underscore the potential of CBFG/POMA-based conductive inks to revolutionize the electronics industry with flexible, affordable, and environmentally friendly materials [65].

### 3.4. Cement and concrete production

Cement is one of the most widely used materials in construction due to its versatility, strength, and durability. It serves as a crucial binder in concrete production. However, the synthesis of cement comes with significant environmental and material challenges. Traditional cement has inherent limitations, such as susceptibility to cracking, high energy consumption during production, and a substantial contribution to global CO<sub>2</sub> emissions. Cement manufacturing accounts for approximately 8% of global CO<sub>2</sub> emissions *via* energy-intensive calcination of limestone, which releases CO<sub>2</sub> as a byproduct [66, 67]. Additionally, the excavation of limestone and clay, which are essential for cement production, results in habitat loss and resource depletion, prompting research into improving cement performance and reducing environmental impact [66].

Cement composites have been enhanced with reinforcement additives like glass fibers [68], carbon nanotubes [69], and graphene oxide [70] to improve resistance to cracking, compressive strength, and thermal stability. However, a more sustainable and efficient solution is needed. FG can be synthesized from waste materials like rubber *via* the FJH process and significantly improves cement's performance. Its high surface area and turbostratic structure enhance bonding with the cement matrix, leading to superior compressive strength and structural integrity. The produced material becomes more resistant to cracking, deformation, and temperature fluctuations, making it ideal for extreme weather infrastructure. Additionally, FG helps to reduce the carbon footprint of traditional cement production.

Advincula et al. [19] investigated the conversion of rubber waste into turbostratic FG using FJH. The research focused on optimizing FJH parameters such as pulse voltage and time to convert pyrolyzed tire carbon black (TCB) and a blend of carbon black with shredded rubber tires (CB: SRT) into high-quality FG. The study achieved a 70% yield using TCB and 47% using the CB blend, with optimal conditions at a voltage of ~140 V-150 V and a pulse time of 500 milliseconds. The dispersion of FG samples in water with 1% Pluronic was investigated. In contrast to commercial graphene and graphite nanoplatelets, which settled out of the solution within 24 hours, FG samples remained stable for over a week (**Fig 13.a**). Moreover, the solubility of FG samples was evaluated by dispersing samples in various solvents, including water with 1% Pluronic F-127, mineral oil, and vegetable oil PAO 6, PAO

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9, and WD-40. After sonication and centrifugation and at a concentration of 5 mg/mL, the FG samples remained stable for over a week as elucidated in **Fig 13.b**. The stability of FG in such a wide range of solvents suggests its potential for easy integration into plastic and cement production processes.

This study also examined the mechanical performance of FG-reinforced cement composites, finding notable improvements in compressive strength. Cement composites incorporating different proportions of TCB FG and CB FG were fabricated to evaluate the effectiveness of FG as a reinforcing agent. After 7 days of curing, the compressive strength of the cement increased by 34% with 0.1 wt% of TCB FG and by 30% with 0.05 wt% of CB FG as elaborated in Fig 13.c. After 28 days, the addition of 0.1 wt% of both TCB FG and CB FG resulted in compressive strength improvements of 31% and 30%, respectively, as shown in Fig 13.d. This suggests that FG can significantly improve the mechanical characteristics of construction materials, potentially reducing the need for large quantities of cement and potentially lowering the carbon footprint of concrete production. From an economic perspective, the energy cost of producing FG via FJH is less than \$100 per ton of rubber waste, making it a promising technology for the large-scale production of graphene and other carbon nanomaterials. In conclusion, the research demonstrated that FJH can efficiently upcycle rubber waste into valuable graphene materials with significant mechanical and environmental benefits, offering a sustainable solution for industries like construction while addressing global waste management challenges.

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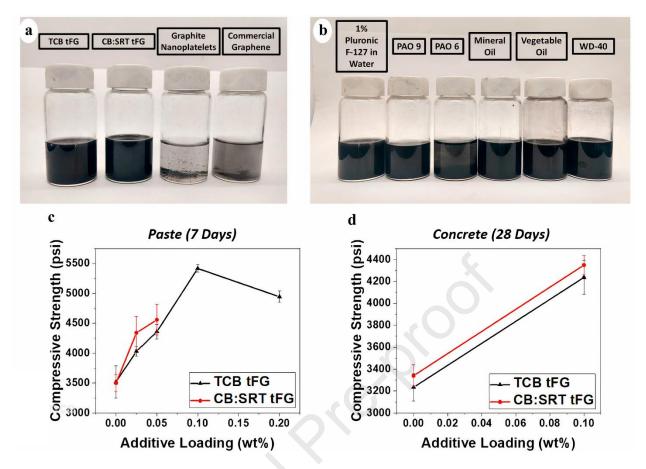


Fig 13. The dispersion of 5 mg/mL of (TCB), (tFG), (CB: SRT), graphite nanoplatelets, and commercial graphene in an aqueous solution of 1% Pluronic F-127 (a). The dispersion of 5 mg/mL of TCB tFG in various solvents and left undisturbed for one week (b). The effect of adding 0.1 wt% of TCB tFG and CB: SRT tFG on the compressive strength of cement in 7 days (c), and concrete in 28 days (d). a-d are reproduced from [19] with

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Algozeeb et al. [71] investigated the conversion of motor oil soot (MOS) and diesel particulates (DP) into high-quality FG using a FJH system. The study focused on the potential of FG as a reinforcement additive in cement mortar, addressing the challenges of cost, scalability, and environmental impact associated with traditional graphene production methods. FJH was optimized with a 500-millisecond discharge time, a voltage of 160 V, and a temperature up to 3000 k, achieving turbostratic graphene with enhanced crystallinity, as confirmed by Raman spectroscopy (I<sub>2D</sub> /I<sub>G</sub> peak ratio of 8–10), XRD, and TEM analyses. FG was incorporated into cement mortar at 0.1 wt%, resulting in significant mechanical improvements after 28 days of curing. The compressive, tensile, and flexural strengths increased by 38%, 27%, and 27%, respectively, while the modulus of elasticity improved by

34%. The enhancement was attributed to FG's nanofiller and bridging effects, which refined pore structures, accelerated hydration, and impeded microcrack formation. Durability tests revealed improved water absorption resistance, though drying shrinkage was slightly higher than control samples, likely due to FG's hydrophobicity and interaction with the cement matrix. Molecular dynamics simulations complemented experimental results, demonstrating that FG reinforced the calcium silicate hydrate structure through stronger Ca–Si and Ca–FG interactions. The simulations also revealed enhanced mechanical properties, with Young's modulus increasing by 49%. This study highlights FG's potential to produce stronger, more durable, and sustainable cement composites using inexpensive waste-derived carbon sources, reducing cement consumption and its associated environmental footprint. Further studies about the application of FG in cement and concrete production are provided in **Text S1** and a comprehensive comparison between these studies is provided in **Table S1**.

### 3.5. Eco-friendly lubricant additives

The growing demand for high-performance lubricants in modern mechanical systems has accelerated interest in innovative materials, that can enhance performance while remaining cost-effective and sustainable. Lubrication technology, essential for reducing friction and wear in mechanical systems, requires materials that enhance performance while maintaining stability under extreme conditions. Traditional lubricant additives often rely on materials like molybdenum disulfide or zinc-based compounds, which can improve wear resistance but may lack the environmental sustainability and cost-effectiveness desired in modern applications [72]. This has led to an increased interest in graphene-based additives due to their superior properties. Graphene-based additives enhance lubricant performance due to their large surface area, mechanical strength, thermal conductivity, ultra-thin structure, chemical stability, and easy dispersion in lubricants. These properties reduce friction, wear, and energy losses, extend lubricant lifespan, and ensure smoother movement, making them highly effective in industrial applications [73].

Advincula et al. [74] explored the applicability of FJH in upcycling waste plastic and metallurgical coke into FG for use as lubricant additives. The study highlighted FG's capability to enhance the tribological properties of poly(alpha olefin) (PAO) lubricants, namely PAO 6 and PAO 9. By incorporating FG derived from waste plastic (WPFG) and metallurgical coke (MCFG), the lubricants exhibited notable decreases in friction, wear scar diameter (WSD), and surface roughness during four-ball tribological evaluations.

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The researchers found that both WPFG and MCFG reduced the coefficient of friction
(CoF) in PAO 9 by 6% and 9%, respectively at $0.1~{\rm mg.mL^{-1}}$ as shown in Fig 14.a. Moreover,
the WSD was significantly reduced, with 14% for WPFG and $8\%$ for MCFG in PAO 9 at $0.1$
$\text{mg.mL}^{-1}$ (Fig 14.b). In PAO 6, a 23% and 6% reduction in CoF % of WPFG and MCFG
were achieved at $0.5~{\rm mg.mL^{-1}}$ , respectively, demonstrating its superior performance (Fig.
14.c). Furthermore, the WSD was significantly reduced, with 12% for WPFG and 14% for
MCFG in PAO 6 at $0.5~\text{mg.mL}^{-1}$ (Fig 14.d). Surface roughness measurements indicated that
reduced roughness was up to 38% and 32% for WPFG and MCFG, respectively, in PAO 6
and by 35% and 29% for WPFG and MCFG, respectively, in PAO 9, further enhancing
lubrication and wear resistance (Fig 14.e and f).

Additionally, life cycle analysis revealed that the production of FG emits up to 99% fewer greenhouse gases and requires 98% less energy and water compared to traditional graphene production methods. Ultimately, the research demonstrated that FG derived from waste materials offers a cost-effective and environmentally friendly alternative to conventional lubricant additives while addressing PW issues. Its ability to enhance lubrication performance while significantly reducing environmental impact makes it a promising solution for industrial applications [74].

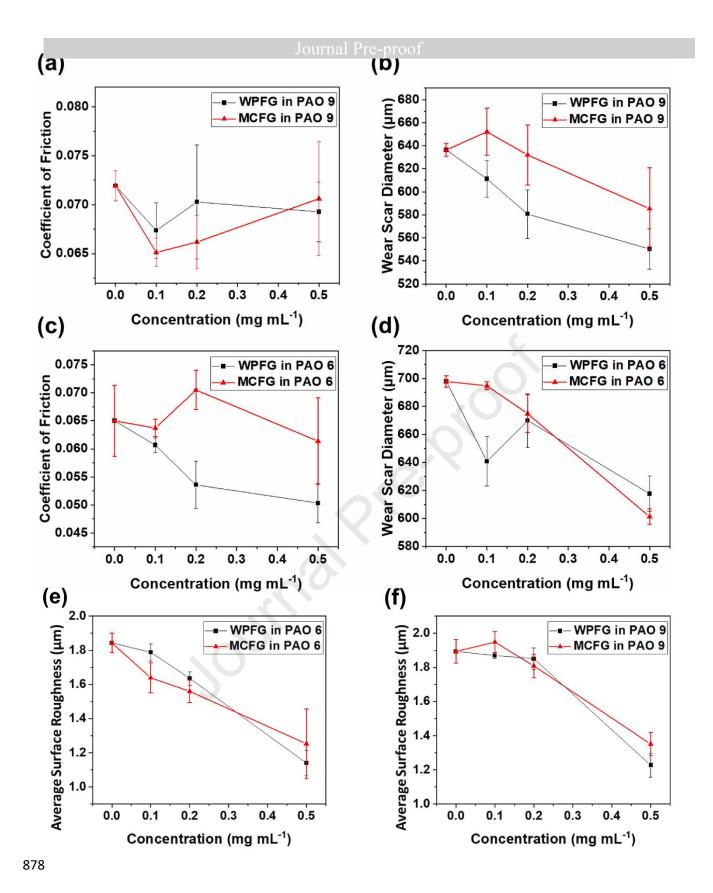


Fig 14. The mean CoF during 1-hour tests for PAO 9 incorporated with WPFG and MCFG additives (a). WSD was recorded for different concentrations of WPFG and MCFG in PAO 9 after 1 hour of testing (b). The mean CoF for PAO 6 mixed with WPFG and MCFG additives in 1-hour tests (c). The mean WSD for different concentrations of WPFG and MCFG in PAO 6 following 1 hour of testing (d). The first point of data only displays the

PAO base oil. The mean surface roughness of wear scars was assessed using white light interferometry (WLI) at 20x magnification after 1-hour tests, followed by 30 minutes of ultrasonic cleaning in an acetone solution. The mean roughness values are presented for PAO 9 mixed with FG (e) and PAO 6 mixed with FG (f). a-f are reproduced from [74] with permission from Elsevier, Copyright [2022].

## 3.6. Functionalization of Polyurethane foam

Polyurethane foam (PUF) is highly valued for its lightweight, flexibility, and durability, making it suitable for insulation, soundproofing, and cushioning applications. PUF is extensively utilized across industries like construction, automotive, and aerospace. However, enhancing its mechanical strength, thermal stability, and acoustic performance is essential to meet modern demands. Incorporating advanced nanomaterials, such as graphene, offers a promising approach to addressing these challenges [75].

Incorporating FG into PUF improves the foam's mechanical resilience and thermal conductivity. Additionally, FG can significantly enhance the foam's acoustic properties by promoting sound absorption and damping, making the material more effective for noise control in applications such as soundproofing panels and automotive interiors. These enhanced foams offer superior noise reduction, making them ideal for environments that require both thermal management and acoustic control, such as buildings, vehicles, and industrial spaces. The synergy between PUF's flexibility and graphene's exceptional physical properties creates a next-generation material that addresses the growing demand for lightweight, durable, and acoustically efficient solutions [76].

Kiddell et al. [77] conducted a study on the use of FG as a reinforcing filler in PUF to enhance its acoustic, thermal, and mechanical properties. They used FJH to produce three different types of FG (FG1, FG2, and FG3) from various feedstock materials. FG was then incorporated into PUF at a concentration of 0.025 wt% to assess its impact on the foam's performance. The study found that FG1 and FG3 significantly enhanced the sound absorption capabilities of PUF, particularly in the 500–2000 Hz frequency range. The presence of FG in the foam matrix improved the dissipation of sound waves by creating frictional forces between the foam and the embedded FG particles.

For thermal properties, FG3 showed a 47% decrease in heat expansion compared to neat PUF, making it more suitable for applications where temperature fluctuations occur, such as insulation materials. Additionally, FG-PUF composites showed improved thermal

decomposition resistance compared to the unmodified foam. In terms of mechanical properties, FG improved both the tensile strength and compressive modulus of the PUF. Tensile strength increased by 16-26% and compressive modulus by 33-37% in all FG-reinforced composites, demonstrating that FG can enhance the foam's ability to resist deformation under stress. However, the addition of FG did not significantly affect the foam's tear strength or compression force deflection, likely due to the relatively small flake diameter and low aspect ratio of the graphene particles. In conclusion, the study demonstrated that the use of FG, even at very low concentrations, can significantly improve the mechanical, acoustic, and thermal properties of PUF. All the above-mentioned results of FG confirm its applicability and high efficacy in a wide range of different applications (**Fig 15**).



**Fig 15**. The applications of FG in water treatment, supercapacitors, conductive inks, lubricant additives, and polyurethane foam.

## 4. Challenges and limitations of FJH process:

• Energy Requirements: Although FJH is efficient in rapidly converting materials, the process requires electrical energy to achieve the extremely high temperatures (up to 3000°C) needed for graphene formation. This can be a limitation in terms of mitigating CO<sub>2</sub> emissions unless the electricity is produced by renewable energy.

- Material Compatibility: Not all waste materials are suitable for FJH. The process is
  most effective with carbon-rich materials, and the presence of non-carbon elements or
  contaminants can affect the quality of the resulting graphene or necessitate additional
  processing steps to remove impurities.
  - Equipment and Infrastructure: FJH requires specialized equipment capable of withstanding the extreme conditions of the process, such as high voltage, temperature, and pressure. The initial investment in this equipment can be high, and maintaining the equipment over time may also pose challenges.
  - **Graphene Quality Control**: While FJH produces FG with desirable properties, there can be variability in the quality and characteristics of the graphene depending on the feedstock and processing conditions. Ensuring consistent quality across large-scale production can be challenging.
  - Unreliability for microfabrication in electronic applications: In microfabrication for electronic applications, CVD is often favored for its ability to produce high-quality thin films of graphene with precise control over thickness and composition, which are the key factors for optimal device performance. In contrast, FJH, while more cost-effective, faces challenges due to the inherent impurities introduced during the fabrication process as the carbon is mostly sourced from waste materials. The preference for CVD in this field is further reinforced by its widespread use in the semiconductor industry, where the consistency of production is essential for ensuring a continuous supply of high-quality graphene. The limitations of FJH in achieving such precision likely contribute to the slower pace of research publications in this area, as its utility is primarily limited to applications where high precision is not a critical requirement.
  - Environmental and Safety Concerns: The high temperatures and rapid heating involved in FJH could potentially lead to safety concerns, particularly regarding the handling of reactive materials and the risk of thermal runaway. Additionally, managing the emissions or byproducts generated during the process may require careful consideration to minimize environmental impact.

#### 5. Conclusions and future recommendations

FJH is a cutting-edge approach to graphene production that effectively overcomes several limitations found in traditional synthesis methods. By applying a high-voltage electric

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current to carbon-rich materials like coal, and plastics, FJH facilitates the rapid transformation of these feedstocks into high-quality graphene within milliseconds to seconds. The highest reported  $I_{2D/G}$  value so far compared to all other previously produced graphene, which is 17, was reported for carbon black-based FG. This technique offers substantial improvements over conventional processes like CVD, which are typically more energyintensive and time-consuming. A significant advantage of FJH lies in its scalability and potential for automation. Researchers are continuously working on improving material handling processes and integrating programmable systems to maintain consistent graphene production on an industrial scale. The scalability of this method holds immense promise for widespread commercial use, especially given its cost-effectiveness and flexibility to process various carbon-based waste materials such as e-waste, plastics, and food waste. However, the goal of producing 100 tons per day has not been yet achieved. FJH enabled researchers to recover 98% of batteries' metal components with a significant reduction in energy, water and acid consumption, and CO<sub>2</sub> emissions. Additionally, if renewable energy powers the electricity grid, FG systems have the potential to achieve carbon neutrality or even become carbon-negative by minimizing reliance on fossil fuels. A ton of FG made of coal is estimated to cost about 130-135 US dollars, whereas conventional graphene would cost 60000-100000 US dollars. FG showed promising results in various applications including concrete, conductive inks, lubricants, and supercapacitors production, as well as water treatment.

According to the conclusions and the major findings of the current review article, there are some recommendations to be made:

- Using renewable energy-based electricity is highly recommended to reduce CO<sub>2</sub> emissions.
- Using different types of bio-waste materials such as plant species should be investigated as plants have different concentrations of cellulose, hemicellulose, lignin, and other organic components that are used in producing different nanomaterials with different properties (eg: different morphologies) as discussed in the literature [78]. Such phytoconstituents could be extracted and used separately or collectively to produce FG, and therefore these variations can influence the quality and properties of the graphene, which is interesting and can lead to promising results. Unlike plastics and coal, plants constitute a natural and renewable source of carbon.
- Although it has been four years since the first publication of FG by Prof. Tour's group, the total number of publications is only 46. These publications covered the

- automation and scaling up of FJH using specific carbon sources (lithium-ion batteries), but the feasibility of doing that by various carbon sources still needs to be investigated.
  - The mechanism of the FJH should be further investigated in future studies using advanced characterization techniques such as operando technique to clarify and improve the understanding of FJH [79, 80].
  - How is the quartz tube cleaned after FJH and reused with different feedstocks to avoid cross-contamination? Or if it is just used once, how much would it cost in the long run? This point should be addressed in the publications to better clarify the costeffectiveness of FJH.
  - Like the emerging nanomaterials, toxicity studies of FG should be undertaken.
  - Collaboration between different research groups around the world and with industrial
    manufacturers especially in developing countries in improving the graphene quality
    and its global production should be pursued. Such collaboration includes the funding
    of workshops and internship programs where school and university students as well as
    young researchers can gain the experience of working in the FG production industry.
  - For industrial applications of FJH, it is essential to incorporate waste heat recovery and off-gas treatment systems into the process [79]. These systems enable the efficient capture and reuse of heat and gases generated during the FJH process, which necessitates the above-mentioned collaboration between industry and academia. All these strategies can increase public and researchers' awareness of FG and result in increasing the number of publications related to this point, which is only 46.

# **CRediT** authorship contribution statement

Mohamed Hosny: Writing — original draft, Visualization, Investigation, Data analysis, Conceptualization, Validation, Review & editing. Ahmed S. Elbay: Writing — original draft, Visualization, Investigation, Data analysis, Formal analysis, Conceptualization.

Ahmed M. Abdelfatah: Writing — original draft, Visualization, Investigation, Data analysis, Conceptualization.

Nourhan El-Maghrabi: Writing — original draft, Visualization, Investigation, Data analysis, Conceptualization, Manal Fawzy: Conceptualization, Project administration, Resources, Supervision, Review & editing.

# **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Data availability

Data will be made available upon request

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$\Box$ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: