Comparison of environmental impacts in the production of graphene from biomass waste and the Hummers' method

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A B S T R A C T

Graphene has found extensive applications in a wide range of critical technologies, and demand is anticipated to grow exponentially in the coming years. This research investigates the environmental impacts of two laboratory-scale methods for graphene production, focusing on their potential applications in electronic devices. The Hummers' method effectively produces high-performance reduced graphene oxide; however, it has faced criticism due to toxic emissions. In contrast, the production of graphene from biomass waste through pyrolysis offers a more environmentally friendly alternative for sustainable manufacturing. A cradle-to-gate life cycle assessment (LCA) is conducted, focusing on three biomass waste materials: banana peel waste, wheat straw waste, and Populus wood waste (PWW), which serve as precursors for graphene production via potassium hydroxide (KOH) activation. The analysis evaluates global warming potential (GWP) and cumulative energy demand using two functional units: 1 kg of produced graphene and a conductivity normalized unit for specific applications. The results indicate that graphene production from PWW is considerably more environmentally sustainable, reducing the GWP from 4841 kg CO_2 equivalent per kg in the Hummers' method to 115.86 kg CO_2 equivalent per kg for PWW-derived graphene. The results obtained from the comparison of hydrazine-reduced graphene oxide (h-rGO) and graphene derived from PWW, based on FU2, illustrate that the total energy consumption for h-rGO per 1 mg. $S^{-1}.m^{-1}$ is 15 times higher than that for graphene derived from PWW. Electricity consumption is identified as the highest contributor to environmental impacts in graphene production, particularly given the current Iranian electricity mix, which heavily relies on fossil fuels. Furthermore, the usage of KOH was a notable contributor to the environmental impact, with 83% of the GWP impact for PWW-derived graphene attributed to electricity consumption and 13% to KOH consumption. A sensitivity analysis based on three scenarios is conducted, which includes a transition to renewable energy, a 20% reduction in electricity consumption, and a 20% reduction in KOH usage. The results showed that transitioning to renewable energy sources, such as wind power, could significantly reduce environmental impacts. This study illustrates the potential environmental advantages of utilizing biomass waste as a precursor for graphene production.

1. Introduction

In recent years, graphene and related materials have attracted considerable attention due to their exceptional electrical properties, thermal conductivity, large surface area, unique mechanical properties, and superior dispersion performance (Somanathan et al., 2015). Graphene has several potential applications, including its use as a semiconductor in electronics, a substitute for indium tin oxide in liquid crystal displays, an additive in composites to enhance strength and conductivity, and in energy storage and conversion applications (Arvidsson et al., 2014).

The utilization of two-dimensional graphene layers in electronic and optoelectronic devices requires only approximately 15 mg per m^2, demonstrating its efficiency at a small scale. However, when used in bulk as conductive filler material, substantial quantities ranging from 2.4 to 48 kg per m^3 are necessary to achieve the desired functionality

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(Serrano-Luján et al., 2019). This variance in material requirements poses challenges for industrial applications, prompting the exploration of more environmentally responsible alternatives (Levchuk et al., 2023).

Graphene can be produced via various methods, which can be broadly categorized into mechanical exfoliation and cleavage (Sinclair et al., 2019), liquid phase exfoliation, molecular intercalation exfoliation, growth on silicon carbide (SiC), chemical vapor deposition (CVD) (Kang et al., 2012; Obraztsov, 2009), precipitation on metals, and chemical synthesis via oxidation and reduction of graphite (Munuera et al., 2022). Notably, bulk graphene production from various biomass waste has emerged as a promising solution to address the demand for environmentally sustainable and cost-effective graphene manufacturing methods (Idris et al., 2023). Treating and recycling biomass waste significantly enhance the utilization and efficiency of natural resources. Properly processed biomass waste can yield valuable chemicals and carbon materials, thereby promoting environmental protection and resource management (Wu et al., 2024).

Numerous studies have synthesized graphene from biomass and examined its potential across a variety of applications. For instance, Chen et al. (2016) synthesized multilayer graphene from wheat straw. Purkait et al. (2017) converted peanut shell biomass into large-area few-layer graphene using KOH activation followed by pyrolysis, achieving in a high specific surface area of 2070 m^2/g, suitable for supercapacitor applications. Baqiya et al. (2020) developed a method for producing reduced graphene oxide from old coconut shells through acid-assisted mechanical exfoliation, yielding nanoscale particle sizes of 1.42–4.99 nm.

These studies highlight the potential of utilizing biomass waste for environmentally friendly production of graphene. Despite these advancements, addressing the environmental implications of various production routes is essential. While biomass waste as a precursor offers significant environmental benefits (Heydarzadeh et al., 2023) the activation materials and processes involved in graphene production can have substantial environmental impacts (Cossutta et al., 2017). Therefore, a systematic approach, such as Life Cycle Assessment (LCA), is necessary to evaluate the overall environmental burdens associated with these processes (Heidari et al., 2019).

LCA plays a crucial role in assessing the potential environmental impacts of emerging technologies by considering all stages, including raw material acquisition, production, use, end-of-life treatment, recycling, and final disposal (i.e., cradle-to-grave) (Cossutta et al., 2020).

Although significant progress has been made in developing various graphene synthesis technologies, research on the LCA of graphene production remains limited (Khanam et al., 2017). On the other hand, previous research has focused on a limited number of graphene production processes and their variations, including chemical oxidation with biological and thermal reduction using various reduction agents to produce graphene oxide (GO) and reduced graphene oxide (rGO), microwave-assisted exfoliation, liquid phase exfoliation (LPE), molecular intercalation exfoliation, ball milling exfoliation, chemical vapor deposition (CVD), and epitaxial growth (Arvidsson et al., 2014, 2016; Beloin-Saint-Pierre & Hischier, 2021; Cossutta et al., 2017; Serrano-Luján et al., 2019). These studies have demonstrated that the environmental impacts of graphene production depend significantly on the production method (Tripathi et al., 2019).

For instance, Arvidsson et al. (2014), conducted a LCA on graphene produced through ultrasonication and chemical reduction, revealing a significant reduction in environmental impacts related to diethyl ether recovery. Similarly, Arvidsson and Molander (2017) found substantial environmental impacts associated with SiC wafer fabrication due to high electricity consumption and suggested mitigating this impact through the adoption of renewable energy source (Arvidsson and Molander, 2017).

Cossuta et al. (Cossutta et al., 2017) evaluated the LCA of three graphene production methods including: electrochemical exfoliation, thermal reduction or chemical oxidation, and chemical vapor deposition (CVD), and found that chemical reduction methods are less suitable for large-scale production. Serrano-Luján et al. (2019) analyzed the environmental and health impacts of producing graphene oxide and reduced graphene oxide using various methods, concluding that Marcano's method is environmentally preferable to Hummers' method, although the use of hydrazine in Marcano's method is more toxic than glucose.

Table S1 in the supplementary information (SI) summarizes studies conducted on the LCA of graphene production. These findings highlight a critical aspect of graphene production: the significant role of electricity consumption in its environmental impacts (Arvidsson et al., 2014; Arvidsson and Molander, 2017; Cossutta et al., 2020). Research by Arvidsson and Molander (2017) emphasizes that electricity consumption contributes to over 99% of the environmental impacts in the worst-case scenario and more than 76% in the best-case scenario (Arvidsson and Molander, 2017).

This finding further highlights the importance of integrating various electricity production systems into the LCA framework, as environmental impacts can vary significantly depending on the electricity mix. Therefore, understanding the electricity supply chain is essential for accurately assessing the environmental impacts of graphene production (Arvidsson and Molander, 2017). Based on these insights, scalability issues often arise in graphene production due to high energy demands and the complexity of the processing (Wu et al., 2024).

For instance, chemical reduction techniques are less viable for large-scale applications due to the involvement of toxic reagents that present substantial environmental and health hazards. Similarly, methods such as epitaxial growth on SiC wafers are constrained by significant electricity consumption and the necessity for specialized equipment, rendering them impractical for widespread production (Arvidsson and Molander, 2017). In light of these challenges and gaps, there is a growing need to compare the environmental impacts of traditional graphene production methods with those derived from biomass materials. However, there is currently a lack of research comparing the environmental impacts of common methods for graphene production and those derived from biomass raw materials. To address this gap, this study compares the LCA of bulk graphene production through the activation process using KOH with three precursors: Banana peel waste (BPW), Wheat straw waste (WSW) and Populus wood waste (PWW) at the laboratory scale with graphene produced by the common Hummers' method.

The experimental data used in this study were collected by the authors through laboratory experiments conducted in Iran, where the electricity supply predominantly relies on fossil fuels. Detailed methodologies and characterization results from these experiments have been previously reported in Bahmei et al. (2020), Ekhlasi et al. (2018), and Kazemi (2018). Given that the electricity supply chain significantly impacts environmental effects, as highlighted in the literature (Arvidsson et al., 2014; Cossutta et al., 2020; Heidari et al., 2019). By analyzing the Iranian electricity mix and exploring the potential benefits of transitioning to renewable energy sources, this study seeks to provide insights into reducing the environmental impacts of graphene production. The goal is to guide the development of more sustainable and environmentally friendly production methods, ensuring that graphene's potential in electronic devices is realized without compromising ecological integrity.

This article is organized as follows: In Section 2, the experimental methods conducted in the laboratory are explained, along with an explanation of the scope and boundaries of the LCA study. Section 3 presents the LCA results based on two functional units, and Section 4 contains the study's conclusion.

2. Material and methods

2.1. Description of the graphene production processes

2.1.1. Materials and equipment

KOH and hydrochloric acid (HCl), which were used for the synthesis of graphene from biomass waste processes, were purchased from Merck. The Nabertherm furnace model RS80/750/11 was used for both carbonization and activation of the materials in the synthesis of graphene from biomass waste. More details are mentioned in our previous studies (Bahmei et al., 2020; Ekhlasi et al., 2018; Kazemi, 2018).

2.1.2. Activation method by KOH

KOH activation is a well-established method for creating porous carbon structures. The chemical reaction between KOH and carbon during the activation process, as represented by Equation (1), forms a network of pores. At temperatures above 973 K, K_2CO_3 decomposes into CO_2 and K_2O, which contributes to high microporosity (Chen et al., 2020). The combined effects of chemical and physical activation, along with metallic K intercalation, result in highly porous carbon structure (Wang et al., 2012; Aluri et al., 2018). Washing removes metallic potassium and other compounds, preventing the carbon sheets from reverting to a nonporous state (Torri et al., 2016).

$$6KOH + 2C \rightarrow 2K + 3H_2 + 2K_2CO_3 \quad (1)$$

The porosity and surface area of graphene are influenced by the choice of biomass waste precursor (Gomez-Martin et al., 2019). In our previous study, BPW underwent carbonization in an argon atmosphere and activation using KOH, resulting in porous graphene with a specific surface area of 2317.5 m^2/g (Bahmei et al., 2020). Briefly, powdered BPW was carbonized under argon at a heating rate of 10 °C.min ^{1} for about 1 h. The carbonized samples were then mixed with a KOH solution at a KOH/C mass ratio of 5:1 (5 g KOH:1 g carbonized BPW). Activation occurred in an argon atmosphere at a heating rate of 10 °C.min ^{1}, an activation temperature of 900 °C, an activation time ranging from 2 h, and a constant argon gas flow rate of 140 mL min ^{1}. The resulting graphene samples were washed with deionized water, dried at 70 °C for 24 h, and subsequently stored in a closed container. Similar methods were applied to PWW and WSW, revealing high surface areas and positive yields for various precursors (Bahmei et al., 2020; Ekhlasi et al., 2018; Kazemi, 2018). This environmentally friendly method exhibits potential for diverse applications, particularly in energy storage systems (Yang, 2021).

2.1.3. Hummers' method

Hummers developed a method for the production of GO which is extensively utilized. This process utilizes a combination of potassium permanganate and sodium nitrate in concentrated sulfuric acid, resulting in expeditious GO synthesis (Hummers Jr and Offeman, 1958). However, GO is initially lacks electrical conductivity and requires chemical and heat treatments to regain its sp^2 character and enhance its electrical conductivity. The production rGO involves various chemical reduction agents, with hydrazine being the predominant reagent (Chen et al., 2013). Serrano-Luján et al. outline the experimental procedure used to produce hydrazine-reduced graphene oxide h-rGO (Serrano-Luján et al., 2019).

Briefly, graphite flakes (5.0 g) and 3.75 g of sodium nitrate ($NaNO_3$) were mixed with 170 mL of concentrated sulfuric acid (H_2SO_4), cooled in an ice bath, and stirred for 30 min 25 g of potassium permanganate ($KMnO_4$) was slowly added and stirred for an additional 30 min. The reaction was then heated to 35 °C and stirred for an additional 2 h. Subsequently, water (250 mL) and 20 mL of 30% hydrogen peroxide (H_2O_2) were gently added. After stirring for an hour and filtering, the powder was periodically washed with 400 mL of HCl: H_2O (1:10) and dried at room temperature for 24 h. Mild bath sonication of an aqueous GO dispersion (1 mg. mL ^{1}) for 2 h, followed by centrifugation at 4500 rpm for 60 min, produced a brown-colored exfoliated GO dispersion with a final concentration of 0.4 mg. mL ^{1}. To produce rGO, excess hydrazine monohydrate was added to the GO dispersion and refluxed for 1 h, with a reducing agent concentration of 6 μL of hydrazine per 1 mL GO dispersion. Filtration, washing with 200 mL of deionized water, and overnight vacuum drying at 80 °C yielded h-rGO powder (Serrano-Luján et al., 2019). To provide a clear visual representation of the graphene production processes employed in this study, a flowchart detailing both the KOH activation method using biomass waste and the Hummers' method is included in Fig. S1 in the SI.

2.2. LCA methodology

2.2.1. Goal and scope, system boundary and functional unit

To evaluate the sustainability of lab-scale graphene synthesis, a cradle-to-gate LCA was conducted. The LCA approach, following the ISO standards 14040/14044, provides a quantitative analysis of the environmental impacts associated with a product, service, or synthetic process route (ISO-14040, 2006). It is important to note that this study only considers the cradle-to-gate framework, and that the impact of the use and end-of-life phases is not considered. Fig. 1 illustrates the overall processes involved, defining the system boundary for graphene production. Three processes based on biomass waste as raw materials were examined in this study. These types of biomass waste contain significant amounts of carbon and volatile matter, which make them suitable for conversion into valuable materials through the pyrolysis process. All types of biomass waste are regarded as real waste, without any associated burdens.

This study focuses on the production of graphene, particularly its application as a semitransparent electronic conductor.

It is essential to tailor the functional unit of graphene to align with the characteristics relevant to its intended applications, such as conductivity and transmittance. Aligning the functional unit of graphene with its electrical conductivity and transmittance is important to for its application as a semitransparent electronic conductor and consider the final uses. Consequently, to thoroughly account for these aspects and consider the diverse final uses of graphene, this study uses two distinct functional unit. This nuanced approach enables a detailed and contextually relevant assessment of graphene's performance and potential in its role as a semitransparent electronic conductor. The first functional unit (FU1) pertains the production of one kg of graphene. Comparing FU1 with most bulk graphene LCA studies that use the same functional unit is straightforward.

A second functional unit (FU2) is designed to link the production of materials with their ultimate applications. Based on the conductivity and surface area of the reported material, FU2 is calculated as the mass of material required per 1 m^2 layer divided by its conductivity (mg. S ^{1}. m ^{1}). It is important to note that for FU2, only the scenario based on waste materials with the lowest environmental impact is compared to the h-rGO production scenario. The surface area and conductivity for h-rGO are assumed to be the highest, equivalent to 745 m^2 g ^{1} and 35.45 S m ^{1} (Aylón et al., 2007; Imam and Capareda, 2012a).

2.2.2. Life cycle inventory (LCI)

The LCI constitutes the second phase of the LCA study, during which all input and output data from the system boundary are collected (ISO-14040, 2006). In this study, data were categorized into two types commonly used in LCA studies: primary and secondary (Michael et al., 2018). Primary data were collected directly from laboratory experiments conducted by the authors, encompassing all inputs and outputs for graphene synthesis from biomass waste, except for air emissions. Details of the synthesis methods for the three precursors are provided in our previous research (Bahmei et al., 2020; Ekhlasi et al., 2018; Kazemi, 2018).

Input and output data are presented in Table 1, while additional

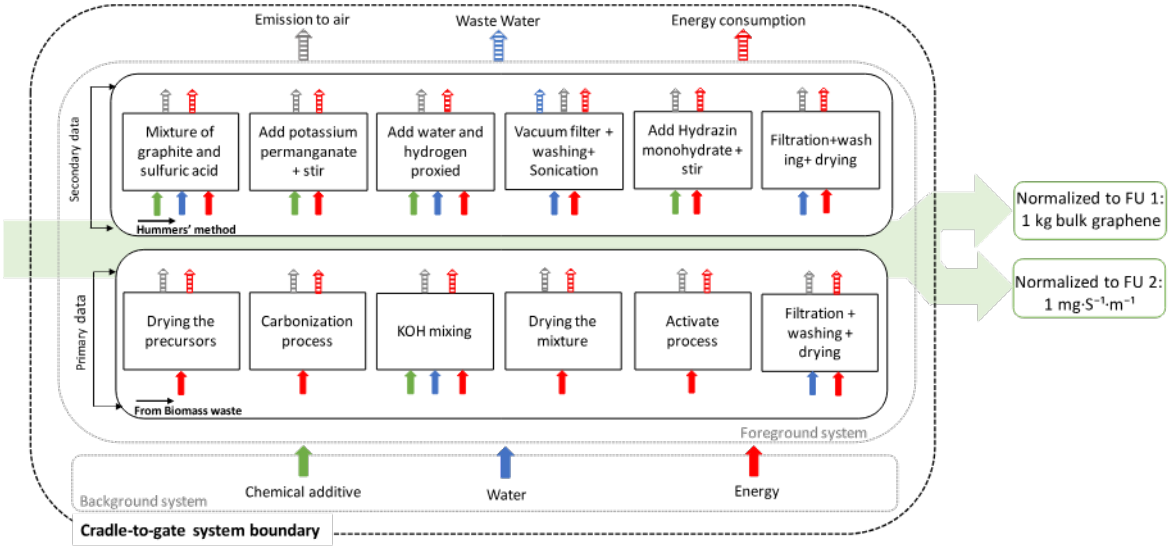

Fig. 1. Cradle-to-gate system boundary applied in the LCA methodology for Graphene Production from Biomass Waste and Hummers' Method (Serrano-Luján et al., 2019).

Table 1
The inventory data for the production of graphene from three biomass wastes.

Populus wood waste (PWW)		Banana peel waste (BPW)		Wheat straw waste (WSW)	
Input					
Substance	Quantity (kg)	Substance	Quantity (kg)	Substance	Quantity (kg)
Populus wood waste	6	Banana peel waste	126	Wheat straw waste	24
Nitrogen, liquid	1.37	Argon, Liquid	3.84	Argon, Liquid	14.98
Potassium hydroxide	5.6	Potassium hydroxide	25	Potassium hydroxide	32.7
Water deionized	8	Water deionized	1000	Water deionized	1200
Electricity, (pyrolysis)	127.2[a]	Electricity, (pyrolysis)	291.36[a]	Electricity, (pyrolysis)	683.52[a]
Electricity, (Treatment process)	10.41[a]	Electricity, (Treatment process)	44.38[a]	Electricity, (Treatment process)	173.68[a]
Hydrochloric acid	4				
Output					
Graphene	1	Graphene	1	Graphene	1
H_2 (emission to air)	0.102	H_2 (emission to air)	0.218	H_2 (emission to air)	0.379
CO (emission to air)	0.292	CO (emission to air)	0.623	CO (emission to air)	1.084
CO_2 (emission to air)	0.349	CO_2 (emission to air)	0.744	CO_2 (emission to air)	1.297
CH_4 (emission to air)	0.185	CH_4 (emission to air)	0.396	CH_4 (emission to air)	0.688
C_2H_4 (emission to air)	0.045	C_2H_4 (emission to air)	0.099	C_2H_4 (emission to air)	0.168
C_2H_6 (emission to air)	0.079	C_2H_6 (emission to air)	0.169	C_2H_6 (emission to air)	0.293
H_2O (evaporation) (emission to air)	0.9	H_2O (evaporation) (emission to air)	98.6	H_2O (evaporation) (emission to air)	3.6
N_2 (emission to air)	1.37	Ar (emission to air)	3.84	Ar (emission to air)	14.98
Ash	0.615	Ash	1.335	Ash	2.345
Waste Water	8	Waste Water	1000	Waste Water	1200

[a] kWh.

details regarding the reference process for each input are provided in Table S2 of the SI. It is essential to highlight that for each type of biomass waste; the synthesis process was optimized to achieve optimal results. Factors such as pyrolysis temperature, KOH consumption, and the use of inert gases (such as argon or nitrogen) were optimized to align with the specific characteristics of each waste type (Bahmei et al., 2020; Ekhlasi et al., 2018; Kazemi, 2018).

Secondary data from the literature were used to estimate air emissions, excluding H_2O, specifically the quantities of gases released (Aylón et al., 2007; Imam and Capareda, 2012b). Evaporation resulting from the moisture content of each precursor is accounted for and categorized as H_2O emissions to the atmosphere, which are also classified as primary data. Secondary data were also utilized for the Hummers' synthesis method. The LCI data for h-rGO production via the Hummers' method were sourced from Serrano-Luján et al. (2019) (See Table S3 of the SI) (Serrano-Luján et al., 2019). Align the system boundaries and functional units of this study, these data were converted to reflect 1 kg and 1 mg $S^{-1}{\cdot}m^{-2}$ of h-rGO production, with adjustments to the temporal boundaries as necessary.

This study aims to compare the environmental impacts of two different graphene synthesis methods. Therefore, transportation is not a necessary consideration, particularly given the high degree of uncertainty and its minimal contribution to the total impact (Liu et al., 2024). Additionally, it should be noted that, as this study was conducted on a laboratory scale, the storage and handling of waste materials were not included within the system boundaries. Background data, including the production of virgin materials and energy sources, were obtained from the Ecoinvent 3.9 database. The analysis was conducted using OpenLCA V2 software.

2.2.3. *Life cycle impact assessment (LCIA)*

A wide range of LCIA methods is available, and their selection depends on various factors, including the latest developments in the field, the type of assessment (e.g., midpoint or endpoint), compatibility with

other studies, and the use of regional normalization factors (Munuera et al., 2022). In this study, an environmental impact assessment was conducted using both the Recipe 2016 midpoint method, a well-established and accepted impact assessment approach within the LCA community (Rybaczewska-Błażejowska and Jezierski, 2024), and the Cumulative Energy Demand (CED) method to calculate and compare potential environmental impacts. A total of 18 impact categories were assessed using the Recipe Midpoint method. Based on the literature, five important categories were selected for discussion: acidification (Cossutta et al., 2017; Dericiler et al., 2024; Jia et al., 2022; Liu et al., 2024; Moon et al., 2022; Vilén et al., 2022), climate change (Cossutta et al., 2017; Dericiler et al., 2024; Jia et al., 2022; Moon et al., 2022; Serrano-Luján et al., 2019; Vilén et al., 2022), human toxicity (carcinogenic) (Arvidsson et al., 2014; Beloin-Saint-Pierre and Hischier, 2021; Moon et al., 2022; Serrano-Luján et al., 2019; Vilén et al., 2022)., particulate matter, and energy resources (Arvidsson et al., 2014; Moon et al., 2022; Serrano-Luján et al., 2019). The Cumulative Energy Demand (CED) method illustrates the energy flow throughout the entire system under consideration (Ampah et al., 2019; Arvidsson et al., 2016; Arvidsson and Molander, 2017; Beloin-Saint-Pierre and Hischier, 2021). This method classifies the overall energy consumption associated with graphene production into distinct categories: non-renewable nuclear energy (NRN), non-renewable fossil fuel (NRF), renewable biomass (RB), renewable wind solar (RWS), and renewable water (RW) (Nowrouzi et al., 2021).

2.2.4. Sensitivity analysis

As previously demonstrated, electricity and KOH consumption are critical parameters affecting environmental impacts in pyrolysis processes (Cossutta et al., 2020; Idris et al., 2023; Nowrouzi et al., 2021). Consequently, a sensitivity analysis was conducted to assess the effects of these variables on the waste-based graphene production process. To accomplish this, three main scenarios were developed.

Reduced Electricity Consumption: Optimizing the pyrolysis process could significantly reduce electricity consumption in waste-based graphene production (Ekhlasi et al., 2018). Therefore, a scenario was examined in which electricity consumption was reduced by 20% while maintaining all other conditions to assess the effects of this reduction.

Recycling of KOH: Nowrouzi et al. (2017) have demonstrated that recycling KOH is a promising strategy for synthesizing high-quality products in the pyrolysis process (Nowrouzi et al., 2017). The properties of products obtained using recycled KOH were found to be comparable to those produced with fresh KOH (Nowrouzi et al., 2017). Given these findings, the present study examines a scenario in which KOH consumption is reduced by 20% through recycling and assesses the potential environmental benefits of this reduction.

Future Electricity Supply Chain: In the final scenario, the implications of modifying the electricity supply chain were analyzed while keeping all other variables constant. In Iran's current electricity mix, natural gas is the predominant source, supplying approximately 81% of the total electricity. Oil contributes 14%, and coal accounts for 4%. Renewable energy sources represent less than 1% of the electricity mix as of 2022 (Zamanipour et al., 2023). Within this 1%, the breakdown includes wind energy (~43.21%), solar energy (~37.39%), small hydro (~15.39%), excess waste heat (~2.27%), and bioenergy (~1.77%) (Noorollahi et al., 2021). To assess the potential environmental benefits of transitioning from a fossil-fuel-dominant electricity mix to a more sustainable alternative, a scenario was modeled in which all equipment involved in the graphene production process, including the pyrolysis furnace, ovens, and other related equipment, operates entirely on wind energy.

3. Results and discussion

3.1. LCIA results per FU 1

Fig. 2 presents the LCIA findings for the five categories examined in this study. The results for all categories are provided in Table S4 of the SI. In all cases, the h-rGO scenario exhibited the highest impact, while the PWW demonstrated the least significant impact (Pizza et al., 2014).

The observed results can be attributed to the production efficiency of graphene derived from each precursor, influenced by both the moisture content and the proportion of lignocellulosic and cellulose materials (Rajendran, 2017). Banana peel contains significant amounts of cellulose, hemicellulose, and lignin, ranging from 23 to 36%, 8–22%, and 8–22%, respectively (Kabenge et al., 2018). However, the humidity content in banana peel can be high, ranging from 75% to 90%, which can negatively impact the quality of the resulting graphene and lead to increased energy demand for active drying (Vu et al., 2017). Wheat straw, on the other hand, is an abundant agricultural waste containing approximately 35–45% cellulose, 20–30% hemicellulose, and 10–20% lignin (Pasangulapati et al., 2012; Ranjan et al., 2022). The humidity content in wheat straw is typically ranges from 10 to 20. Populus wood waste, a type of hardwood, contains approximately 40–50% cellulose, 20–30% hemicellulose, and 20–30% lignin. The humidity content in PWW typically ranges from 10 to 20%, making it a promising feedstock material for graphene production (Lühr and Pecenka, 2020). Among the various components of biomass, hemicellulose exhibits the lowest thermal stability, while lignin demonstrates the highest. The high lignin content, due to its phenolic compounds, significantly enhances the final product (Jin et al., 2012). In this study, the lignin content in biomass samples of BPW, WSW, PWW is quantified at 9.3%, 14.8%, and 35.5%, respectively, on a dry weight basis (Table S5).

According to the experimental results, graphene can be produced with efficiencies of 0.9%, 4.1%, and 17% from BPW, WSW, and PWW, respectively (Bahmei et al., 2020; Ekhlasi et al., 2018; Kazemi, 2018). These efficiencies are closely related to the moisture content of the precursor and the lignocellulosic content (Levchuk et al., 2023). Iran's capacity for cultivating fast-growing trees, particularly Populus species, is noteworthy (Navroodi, 2013). Populus wood, known for its superior fiber characteristics, is extensively used in processing industries and rural applications, making it more marketable than other species (Fallah et al., 2012). Nevertheless, waste from the wood industry in this sector, primarily foliage and sawdust, and often has no specific use and is generally burned improperly, leading to the release of several pollutants, including CO_2 (Emam et al., 2015).The difference in the results for the global warming potential (GWP) category was significant, with h-rGO amounting to 4841 kg CO_2 eq.kg ^{1} graphene, while for PWW amounted to 115.86 kg CO_2 eq.kg ^{1} graphene. For WSW and BPW, the corresponding amounts were 717.24 and 315.52 kg CO_2 eq.kg ^{1} graphene, respectively. As illustrated in Fig. 3, electricity consumption during the pyrolysis steps dominates the overall contributions in all scenarios involving graphene-based waste, accounting for 76%, 66%, and 64% per FU1 for PWW, WSW, and BPW, respectively.

The contributions of each process in producing 1 kg of graphene using Hummers' method are detailed in Table S6 of the SI. Subsequently, for BPW and PWW, KOH consumption follows in significance; whereas for WSW, electricity is directed to the treatment process, including the drying phase. Notably, the electricity required for all unit processes is sourced from the Iranian grid, where approximately 97.5% derives from oil and natural gas (Noorollahi et al., 2021). Additionally, it is important to note that energy consumption of laboratory-scale graphene synthesis is considerably higher than that of industrial-scale production (Heidari et al., 2019). These factors substantially contribute to the elevated role of electricity in GWP. Given the various methods for producing graphene, comparing results with other literature sources is challenging. For instance, findings reported by Arvidsson et al. (2014) also recognize the significant role of electricity in contributing to GWP.

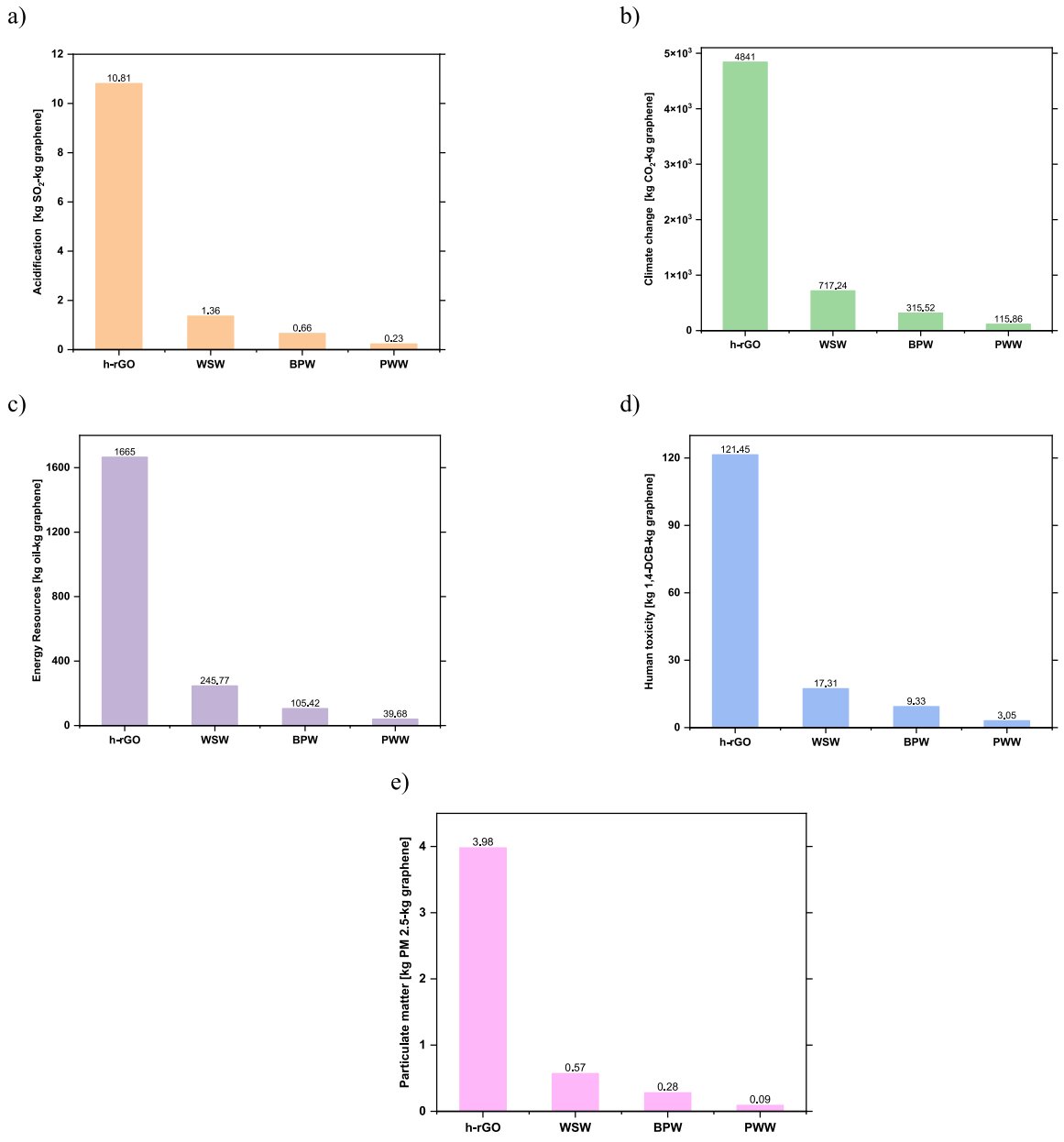

Fig. 2. LCIA results using the ReCiPe method. Values are presented per 1 kg of graphene production as the functional unit. a) acidification, b) climate change, c) energy resources, d) human toxicity, and e) particulate matter potential.

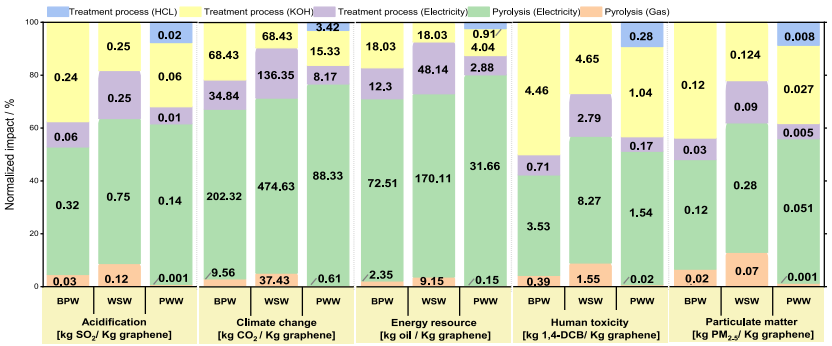

Fig. 3. Synthesis process contribution analysis of environmental impact for three precursors. The absolute values for each synthesis process contribution, expressed in units, are presented in the respective columns.

However, it is noteworthy that in their research, the methods of chemical reduction of graphite oxide and ultrasonication of graphite were employed to produce 1 kg of graphene in solution as the functional unit, resulting in lower CO_2 emissions compared to the current study. This discrepancy can be attributed to the energy supplier; in Arvidsson et al.'s study, Sweden served as the energy supplier, with 50% of its supply sourced from renewable energies (Arvidsson et al., 2014; Navroodi, 2013).

As shown in Fig. 2d, in the human toxicity category (carcinogenic), the h-rGO scenario exhibits percentages exceeding 72%, 78%, and 81% compared to those producing graphene based on WSW, BPW, and PWW, respectively. The impact value is to 121.45 1,4-DCB in the h-rGO scenario, whereas it is only 3.05 1,4-DCB in the PWW scenario, which demonstrates the least impact. The examination of 1 kg of graphene production from biomass precursors in this category revealed that electricity consumption during pyrolysis for PWW and WSW had the most significant impact, accounting for 50% and 48%, respectively, followed by KOH. The analysis of this category (for both carcinogenic and non-carcinogenic substances) showed that metals such as thallium, chromium VI, selenium, nickel, barium, hydrogen fluoride, vanadium, and cadmium are significant contributors to human toxicity. The recovery and reuse of KOH in the synthesis process can significantly mitigate the environmental impacts associated with this category (Nowrouzi et al., 2021).

Electricity pyrolysis contributes between 68% and 80% of the environmental burdens associated with all precursors, exhibiting the highest impact in the energy resource categories (Fig. 2c). In the particulate matter category (Fig. 2e), among the three precursors, the production of 1 kg of graphene-based on WSW had the highest impact, measuring 0.566 kg $PM_{2.5}$.eq. Furthermore, this value was seven times higher, equaling 3.98 kg $PM_{2.5}$.eq for h-rGO. Regarding the acidification category, PWW exhibits the least environmental impact among the three biomass waste. Electricity pyrolysis with 0.139 kg SO_2.eq.kg ^{1} graphene, KOH with 0.055 kg SO_2.eq kg ^{1} graphene, and HCl with 0.017 kg SO_2.eq kg ^{1} graphene had the most significant contribution to environmental burdens. This is primarily due to the release of acid gases, such as SO_2, NO_x, HCl, HF, resulting from the combustion of fossil fuels used in the pyrolysis process (Suganya and Kumar, 2018). During pyrolysis, which is carried out at very high temperatures under inert gas pressure such as N_2 or Ar, the conditions for NOx creation can be optimized. Moreover, the nitrogen and sulfur in the feedstock can react with the remaining oxygen during the early carbonization process, producing SO_2 and NO_2 (Heidari et al., 2019).

The CED values for the cases examined are shown in Fig. 4. The findings indicate that the CED for h-rGO is 80.52 GJ kg ^{1} graphene, which is 6.67, 15.38, and 41.43 times bigger than the CED values for WSW, BPW, and PWW, respectively.

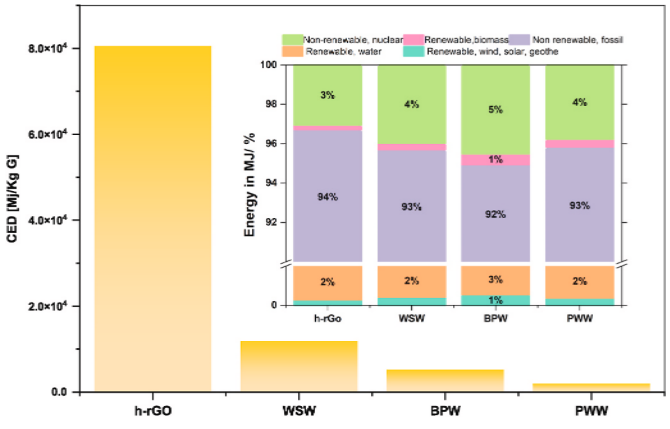

Fig. 4. CED results and parameter contributions attributed to the scenarios per FU1.

According to the CED analysis in this study, NRF was identified as the primary energy source for graphene production from all three precursors, accounting for 93%, 92%, and 93% for WSW, BPW, and PWW, respectively. The assessment also revealed that the pyrolysis process and KOH as an activator were the most energy-intensive NRF procedures. In terms of NRF, natural gas was the primary source of energy in all three scenarios, followed by crude oil. Furthermore, the largest consumption of fossil fuels across all scenarios was associated with electricity supply during the carbonization and activation processes. The results of this study are consistent with prior research of Nowrozi et al. which found that over 56% of energy consumption could be attributed to electricity supply during the activation and carbonization stages of producing activated carbon from biomass waste (Nowrouzi et al., 2021).

3.1.1. *Comparison with existing studies*

LCA results demonstrate substantial variability depending on the data used, methodology, and system boundaries defined. The compared studies encompass diverse impact categories; thus, only the shared category of GWP is considered in this analysis. Notably, LCA investigations of graphene production utilize distinct functional unit. For meaningful comparison, only studies utilizing the same functional unit of 1 kg bulk graphene are included.

For the Hummers' method utilizing hydrazine as the reducing agent, the reported GWP is 586 kg CO_2.eq. kg ^{1} graphene, with the production and utilization of hydrazine and phosphoric acid identified as principal hotspots. Comparative analysis indicates that the production of 1 kg of graphene from PWW is significantly lower than that from the other methods examined, particularly when considering the final application (Serrano-Luján et al., 2019). Serrano-Luján et al. noted a 40% reduction in GWP by using glucose as a reducing agent, which translates to a decrease of 343 kg CO_2 equivalent per kilogram of graphene. However, this value remains 66% higher than the GWP calculated for graphene derived from PWW in this study (Serrano-Luján et al., 2019). Referring to a study by Khanam et al. the GWP for graphene sheets produced via the Hummers' method with hydrazine as the reducing agent is reported at 85 kg CO_2 equivalent. kg ^{1} graphene. However, their study lacks information on the final use of graphene. Although this reported GWP represents the minimum documented value for this production process based on laboratory-scale data, the available information is insufficient to comprehensively explain the observed disparities when compared to findings from other studies (Harding et al., 2007).

3.2. *LCIA results per FU 2*

In this section, as detailed in Section 2.2.1, the FU2 examines the electrical conductivity and surface area of graphene. These variables are crucial when considering the application of carbon materials in energy storage systems (Canal-Rodríguez et al., 2017). Two scenarios were compared as part of this analysis (Fig. 5). The results from section 3.1 demonstrated that the production of 1 kg of graphene using PWW as a precursor has the least environmental impact in comparison to BPW and WSW. To facilitate comparison based on FU2, the inputs and outputs for the PWW scenario and the h-rGO scenario were normalized using a factor of 1 mg S ^{1}·m ^{1}. The results are presented in SI, Table S7 encompassing the 18 categories. Although it is established that h-rGO exhibits greater electrical conductivity than graphene derived from PWW, requiring less material per FU2 (amount of product per m^2 and its conductivity), the environmental impact across all categories remains higher for h-rGO (Kabenge et al., 2018; Pasangulapati et al., 2012; Vu et al., 2017). Despite increased material consumption due to lower electrical conductivity and suboptimal synthesis conditions for graphene derived PWW, this method remains more suitable for producing graphene with the desired functionality in electronic and optoelectronic applications. Additionally, the CED results showed that the total energy consumption for h-rGO per 1 mg S ^{1}·m ^{1} is 15 times higher than that for graphene derived from PWW; further details are presented in SI,

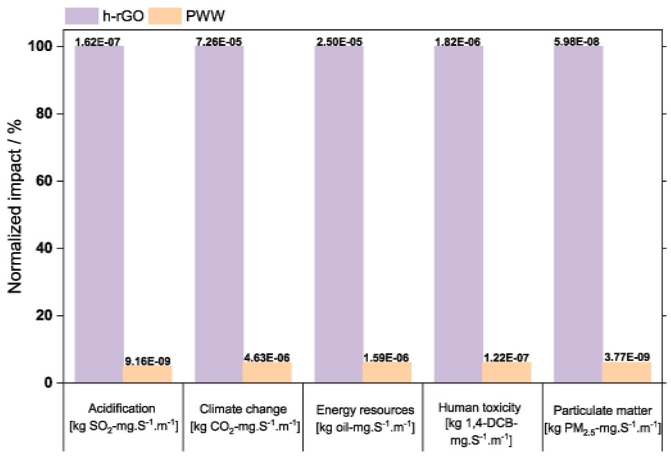

Fig. 5. LCIA results of Recipe method, Values are presented per 1 mg $S^{-1}{\cdot}m^{-1}$ of graphene (FU2). The absolute values, expressed in units, are presented in the respective columns.

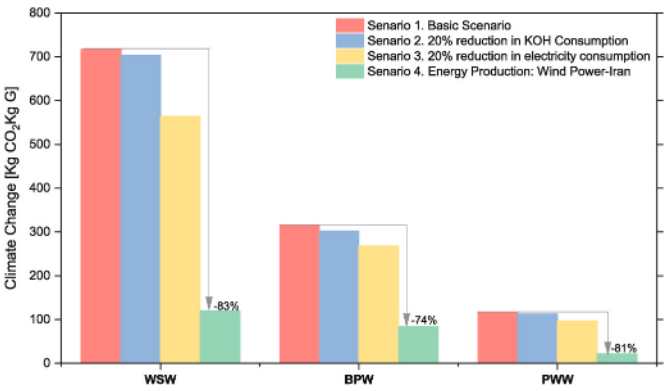

Fig. 6. Sensitivity analysis results for the GWP of biomass waste-derived graphene across all three precursors.

Table S8. Overall, this examination highlights the environmental advantages of graphene derived from PWW as compared to h-rGO, and confirms its viability for application in electronic and optoelectronic systems.

Nevertheless, the lower conductivity of biomass-derived graphene poses a challenge, particularly in applications where high performance and precise quantities are essential. While biomass-derived graphene offers environmental advantages, its conductivity is generally lower than that of h-rGO (Sawant et al., 2022). This lower conductivity can affect its performance in advanced technological applications, such as energy storage and high-performance electronics (Zhou et al., 2022). To address these limitations, ongoing research and technological innovations are imperative. Strategies such as doping, chemical functionalization, and optimized reduction processes show promise for enhancing the electrical properties of biomass-derived graphene (Sawant et al., 2022). Doping introduces various elements into the graphene structure to enhance its conductivity, while chemical functionalization involves attaching specific chemical groups to the graphene surface to improve its performance and interaction with other materials. Moreover, optimizing reduction processes can enhance conductivity by removing oxygen-containing groups that impede electron flow (Kumar et al., 2023). Employing these techniques can significantly enhance the electrical performance of biomass-derived graphene, making it suitable for demanding applications while retaining its environmental benefits. Continued research in these areas is vital for establishing biomass-derived graphene as a viable alternative in high-performance applications, effectively balancing functionality with environmental sustainability.

3.3. Sensitivity analysis results

As detailed in Section 3.1 on graphene production through pyrolysis, the primary contributor to GWP is electricity consumption, followed by KOH consumption. A sensitivity analysis was conducted by reducing both electricity consumption and the amount of the chemical activating agent by 20%. Additionally, a future scenario was introduced to assess the impact of transitioning Iran's current electricity mix to a more sustainable alternative, specifically wind power. Fig. 6 illustrates the GWP results from the sensitivity analysis for biomass waste-derived graphene across all three precursors.

The results indicate that reducing KOH consumption, in comparison to other scenarios, leads to a smaller reduction in GWP for graphene production from all three precursors. Specifically, the GWP decreases by 2.0%, 2.6%, and 4.5% for WSW, PWW, and BPW, respectively. Conversely, the reduction in electricity consumption yields more significant impacts, with decreases of 15% for BPW, 16.5% for PWW, and 21.5% for WSW. The third scenario, involving a transition to wind power, demonstrated the most substantial reduction in GWP. This underscores the vital role of renewable energy sources in mitigating the environmental impact of graphene production.

Notably, the current electricity mix in Iran heavily relies on fossil fuels, comprising over 98%, which significantly contributes to environmental pollution and CO_2 emissions (Noorollahi et al., 2021). Natural gas dominates electricity generation in Iran, comprising approximately 81% of the total; oil contributes 14%, coal accounts for 4%, and renewable sources, including solar and wind, constitute less than 1% as of 2022 (Zamanipour et al., 2023). Although wind energy has substantial potential in Iran, with an estimated recoverable air capacity of around 100 GW, its current utilization remains limited (Fathi et al., 2023). Utilizing wind power as the electricity supply scenario significantly reduces the environmental impact of graphene produced from all three precursors across all impact categories (See Tables S9–S11 in the SI).

Results in the GWP category revealed a substantial decrease from 717, 315, and 115 kg CO_2. kg^{-1} graphene to 119, 83, and 21 kg CO_2. kg^{-1} graphene for WSW, BPW, and PWW, respectively (Fig. 6). The present results are consistent with previous evaluations of graphene, indicating that the use of an electrical supply chain containing a high percentage of renewable energy significantly reduces GWP (Canal-Rodríguez et al., 2017; Cossutta et al., 2020). In conclusion, besides reducing electricity and KOH consumption, transitioning to renewable energy sources, such as wind power, leads to the most significant reductions in GWP and other environmental impacts. This emphasizes the critical importance of adopting renewable energy solutions to minimize the environmental footprint of graphene production.

3.4. Limitations and recommendations

The data used in this study were obtained from laboratory conditions, where the environmental impacts associated with electricity consumption were significantly higher than those observed with industrial equipment, which typically demonstrates greater energy efficiency and reduced energy consumption (Heidari et al., 2019). Consequently, the results of this study may not fully reflect real-world industrial scenarios. Additionally, water consumption and wastewater generation at the laboratory scale can present challenges. Laboratory-scale processes often produce higher volumes of wastewater due to less efficient water management systems compared to industrial-scale operations. Implementing strategies such as counter-current washing systems and wastewater recycling, as demonstrated in the study by Nowrouzi et al. (2017), could significantly reduce both water consumption and wastewater discharge, even at the laboratory scale. This approach would not only improve the sustainability of

the process but also provide a strong foundation for scaling up to more resource-efficient industrial operations (Nowrouzi et al., 2017).

Future research should aim to scale these findings to industrial contexts, where enhanced efficiency can yield a more accurate assessment of environmental impact. Incorporating life cycle costs into these assessments will provide a more comprehensive view of the financial implications associated with the production, use, and disposal of materials such as graphene. Investigating the social impacts of these materials is essential. Comprehensive LCA that encompass all stages of the life cycle are essential for evaluating the advantages of graphene derived from various sources over alternative materials for comparable applications. Additionally, the complexities involved in conducting LCAs for nanomaterials such as graphene highlight the challenges of accurately assessing their environmental impacts (Healy et al., 2008).

Significant gaps remain in understanding the release of nanomaterials throughout their life cycle, including production, use, and disposal (Gilbertson et al., 2015). This uncertainty complicates the evaluation of their environmental and health impacts, as well as their behavior post-release. To improve LCA accuracy, it is proposed to incorporate specific factors related to the environmental toxicity of graphene oxide in freshwater systems (Deng et al., 2017). To enhance LCA accuracy, it is recommended to incorporate specific factors related to the environmental toxicity of graphene oxide in freshwater systems (Deng et al., 2017).

Furthermore, the characteristics of graphene—such as its shape, size, surface functionality, and electrical charge—may influence its interactions with biological tissues, which is vital for predicting potential toxic effects. Therefore, further research is necessary to refine LCA methodologies and ensure the safe use of nanomaterials like graphene. On the other hand, transitioning to renewable energy sources, such as wind and solar power, is essential for mitigating the environmental impacts associated with graphene production, particularly in regions like Iran that heavily rely on fossil fuels.

Given Iran's favorable conditions for solar and wind energy, harnessing these renewable resources could significantly reduce the environmental footprint of industrial activities, including graphene production. Moreover, utilizing biomass waste as a feedstock for graphene synthesis offers considerable environmental and economic advantages. Effective management of biomass waste can transform it into a sustainable and cost-effective resource, thereby reducing reliance on non-renewable materials and addressing waste disposal challenges. Policymakers should encourage investments in technologies that facilitate biomass waste processing to promote more sustainable production practices. Given graphene's potential applications in energy storage and other fields, implementing these strategies could enhance both environmental sustainability and technological innovation.

4. Conclusions

This study aimed to evaluate and compare the environmental impacts of graphene production from biomass waste with those from the Hummers' method, using a cradle-to-gate LCA approach. The biomass waste precursors analyzed included banana peel waste (BPW), wheat straw waste (WWS), and Populus wood waste (PWW). The results demonstrated that the environmental impacts of producing 1 kg of graphene from these biomass precursors were significantly lower than those from the Hummers' method. Among the precursors, PWW exhibited the highest graphene production efficiency during pyrolysis due to its higher lignocellulosic content and lower moisture levels, leading to a greater yield of bulk graphene. Additionally, PWW showed the lowest environmental impacts across all assessed categories compared to graphene derived from BPW and WSW. Specifically, global warming protentional (GWP) results for graphene production from WSW showed 150 kg CO_2 eq per kg of graphene, which was substantially higher than the 110 kg CO_2 eq for BPW and 95 kg CO_2 eq for PWW. Cumulative energy demand analysis further revealed that producing graphene from WSW required 3200 MJ/kg, compared to 2500 MJ/kg for BPW and 2200 MJ/kg for PWW. In addition to the primary functional unit of 1 kg of graphene, a secondary functional unit was developed, focusing on electrical conductivity and surface area. Analysis showed that while PWW-derived graphene exhibited lower electrical conductivity compared to graphene produced using the Hummers' method, the environmental impacts across all categories were still significantly lower for PWW-derived graphene. Specifically, the total energy consumption for hydrazine-reduced graphene oxide per 1 mg $S^{1}\cdot m^{1}$ was 15 times higher than that for graphene produced from PWW. This underscores the environmental benefits of PWW-derived graphene despite its lower conductivity, which is vital for high-performance applications. Therefore, further research and technological innovations are essential to enhance the electrical properties of biomass-derived graphene. A detailed analysis of the LCA results identified electricity consumption and KOH use during the pyrolysis process as the primary contributors to the overall environmental impacts. Scenario analysis showed that substituting grid electricity with wind power in Iran could potentially reduce climate change impacts by up to 80% and human toxicity impacts by 75% across all three biomass precursors. These findings suggest that PWW represents a more sustainable and environmentally friendly option for industrial-scale graphene production compared both to other biomass precursors and to the conventional Hummers' method. Given that PWW is a byproduct of the Iranian wood industry, which is currently underutilized and often improperly disposed of, it presents a valuable and sustainable resource for graphene production. However, this study was conducted under laboratory conditions, which may not fully reflect industrial-scale efficiencies. Future research should scale these findings to industrial contexts, include life cycle cost assessments, and also evaluate social impacts. Transitioning to renewable energy sources, such as wind or solar power, could further reduce environmental impacts, especially in regions like Iran, where fossil fuel reliance is high.

CRediT authorship contribution statement

Fatemeh Bahmei: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Nader Bahramifar:** Writing – review & editing, Supervision, Software, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation. **Shahram Ghasemi:** Writing – review & editing, Visualization, Supervision, Software, Data curation, Conceptualization. **Habibollah Younesi:** Writing – review & editing, Writing – original draft, Supervision, Software, Funding acquisition, Data curation, Conceptualization. **Marcel Weil:** Writing – review & editing, Supervision, Resources, Data curation, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author(s) used ChatGPT in order to improve readability and language. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

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 on request.

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