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Analysis of Energy Consumption in the production of Graphene material and its impact on the Environmental Footprint

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Abstract

This study evaluates the energy consumption of eight different technological pathways for producing graphene and reduce graphene oxide and analyzing their environmental impact. This study is part of the Horizon Europe - GIANCE Project (Graphene Alliance for Sustainable Multifunctional Materials to Tackle Environmental Challenges). Comparing the life cycle assessment (LCA) of the new solutions to the reference solutions is one of Fraunhofer's responsibilities in the project. Therefore, a specific database of Graphene material, i.e., life cycle inventory (LCI), is required. However, there is a lack of LCI data on graphene processes, and this is one of the main challenges, as the database is the core of an LCA. Fraunhofer ICT has developed life cycle inventories to overcome this challenge, considering gate-to-gate system boundaries of graphene-related processes. Eight processes were chosen for graphene production based on the GIANCE project's use cases. They are as follows: (i) Chemical reduction of graphite oxide; (ii) Electrochemical exfoliation of graphite; (iii) Ultrasonic Exfoliation; (iv) Hummers method; (v) Marcano's Method; (vi) Chemical oxidation; (vii) Hydrothermal method; and (viii) Annealing method. This study provides a detailed overview of the current understanding of the environmental impact of graphene production. It seeks to identify potential pathways for development of LCI data while highlighting existing knowledge gaps in this area. The methodological approach uses the Environmental Footprint (EF) 3.1 impact assessment method to analyze the impact of energy consumption of graphene production processes. LCIA results of the different impact categories is transferred in single scores using the normalization and weighting factors of the Product Environmental Footprint from the European Union (EU). This includes conducting a thorough contribution analysis of each process, finding the environmental performance of different production methods, and identifying critical environmental hotspots that require attention to enhance the overall environmental sustainability of graphene production process. The findings underscore the critical need for more precise and comprehensive data on graphene production methods to determine frontrunners in terms of environmental impact accurately. By addressing these elements, the study intends to provide essential insights that will direct towards the LCI database of graphene and its impact on Environmental Footprint.

Kurzfassung

Diese Studie bewertet den Energieverbrauch von acht verschiedenen technologischen Wegen zur Herstellung von Graphen und zur Reduzierung von Graphenoxid und analysiert deren Umweltauswirkungen. Diese Studie ist Teil des Horizon Europe - GIANCE Projekts (Graphene Alliance for Sustainable Multifunctional Materials to Tackle Environmental Challenges). Der Vergleich der Ökobilanz (LCA) der neuen Lösungen mit den Referenzlösungen ist eine der Aufgaben von Fraunhofer in diesem Projekt. Daher wird eine spezifische Datenbasis für Graphen-Materialien, d.h. ein Lebenszyklusinventar (LCI), benötigt. Es gibt jedoch einen Mangel an LCI-Daten zu Graphen-Prozessen, und dies ist eine der größten Herausforderungen, da die Datenbank das Herzstück einer LCA ist. Das Fraunhofer ICT hat Lebenszyklusinventare entwickelt, um diese Herausforderung zu bewältigen, indem es die Systemgrenzen von Graphen-bezogenen Prozessen von Gate zu Gate berücksichtigt. Auf der Grundlage der Anwendungsfälle des GIANCE-Projekts wurden acht Verfahren für die Graphenproduktion ausgewählt. Diese sind wie folgt: (i) chemische Reduktion von Graphitoxid; (ii) elektrochemische Exfoliation von Graphit; (iii) Ultraschall-Exfoliation; (iv) Hummers-Methode; (v) Marcano-Methode; (vi) chemische Oxidation; (vii) hydrothermale Methode; und (viii) Annealing-Methode. Diese Studie gibt einen detaillierten Überblick über das derzeitige Verständnis der ökologischen Nachhaltigkeit der Graphenproduktion. Sie versucht, potenzielle Entwicklungspfade zu identifizieren und gleichzeitig bestehende Wissenslücken in diesem Bereich aufzuzeigen. Der methodische Ansatz verwendet die Methode des Environmental Footprint (EF) 3.1 zur Analyse von Graphen-Produktionsprozessen. Die LCIA-Ergebnisse der verschiedenen Auswirkungskategorien werden unter Verwendung der Normalisierungs- und Gewichtungsfaktoren des Product Environmental Footprint der Europäischen Union (EU) in Einzelscores übertragen. Dies beinhaltet die Durchführung einer gründlichen Beitragsanalyse jedes Prozesses, die Ermittlung der Umweltleistung verschiedener Produktionsmethoden und die Identifizierung kritischer Umwelt-Hotspots, die Aufmerksamkeit erfordern, um die gesamte ökologische Nachhaltigkeit des Graphen-Produktionsprozesses zu verbessern. Die Ergebnisse unterstreichen den dringenden Bedarf an präziseren und umfassenderen Daten über Graphen-Produktionsverfahren, um die Spitzenreiter in Bezug auf die Umweltauswirkungen genau zu bestimmen. Durch die Behandlung dieser Elemente beabsichtigt die Studie, wesentliche Erkenntnisse zu liefern, die zu einer LCI-Datenbank für Graphen und seine Auswirkungen auf den ökologischen Fußabdruck führen werden.

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List of Abbreviations

AM	Annealing Method
CF	Carbon Fiber
CNTs	Carbon Nanotubes
CRR	Chemical Reduction Routes
CVD	Chemical Vapour Deposition
CF	Characterization Factor
EF	Environmental Footprint
FRPCs	Fiber-Reinforced Polymer Composites
GFs	Glass Fiber
GNPs	Graphene Nanoplatelets
GO	Graphene Oxide
HM	Hydrothermal Method
ILCD	International Life Cycle Data System
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
MWCNTs	Multi-Walled Carbon Nanotubes
NF	Normalization Factor
N-rGO	Nitrogen-doped reduced Graphene Oxide
PCs	Polymer Composites
PEF	Product Environmental Footprint
rGO	Reduced Graphene Oxide
SWCNTs	Single-Walled Carbon Nanotubes
TRL	Technology Readiness Level

List of Symbols

$Ca(OH)_2$	Calcium hydroxide
CH_3OH	Methanol
Cp	Specific heat capacity
CTUe	Comparative Toxic Unit for Ecosystems
CTUh	Comparative Toxic Unit for Humans
E	Energy
f	Heat conversion efficiency
h	efficiency
H_2O	Water
H_2O_2	Hydrogen peroxide
H_2SO_4	Sulfuric acid
H_3PO_4	Phosphoric acid
HCL	Hydrochloric acid
I	Inventory
i	Impact category
j	Number of flows
kBq U235 eq	equivalent amount of kilograms of Uranium-235
kg CFC-11 eq.	Kilograms of trichlorofluoromethane equivalent
Kg CO_2 eq.	Kg of CO_2 released into the atmosphere per unit mass of material
Kg N eq.	Kilograms of nitrogen equivalent
kg NMVOC eq.	the equivalent amount of Non-Methane Volatile Organic Compounds
Kg P eq.	Kilograms of phosphorus equivalent
$KMnO_4$	Potassium permanganate
KOH	Potassium hydroxide
kWh	kilowatt-hours
m	Mass

MJ	Megajoules
Mole of H^+ eq.	Moles of hydron concentration
N_2H_4	Hydrazine
$NaNO_3$	Sodium nitrate
NH_4NO_3	Ammonium nitrate
ΔT	Temperature change

1. Introduction

Graphene and similar materials have gained global interest in the last 20 years due to their remarkable properties [2]. Graphene, a single layer derived from graphite, this is a two-dimensional sheet, composed of sp^2 carbon atoms[1] created in related forms such as graphene oxide (GO) and reduced graphene oxide (rGO) [3]. Showcases extraordinary mechanical, electrical, and thermal properties, positioning it as a highly promising material for diverse applications. Why choose graphene instead of other materials for composite production for high-level applications? (e.g. aircraft, vehicles).

Figure 1: Use of Graphene material in high level applications [1]

The figure illustrates GIANCE's strategic approach to addressing environmental challenges through the development of advanced materials based on graphene and related substances (GRM). The platform is designed to foster creativity and innovation in material science, focusing on creating affordable, eco-friendly, lightweight, and recyclable materials known as GRM-bM (Graphene and Related Materials-based Materials) [1].

[1] describes how carbon atoms arrange their electrons, here carbon uses one S orbital and two P orbitals to create a hexagonal structure.

Looking into the automobile industry, where challenges come in the form of decreasing CO_2 emissions, decarbonization targets, safety concerns, and energy consumption issues, graphene emerges as a superior choice. Its exceptional strength, dimensional stability, flame resistance, and heightened durability make it a convincing option [4]. To address the industry's need for lighter vehicles and reduce fuel consumption, as a result, reducing CO_2 emissions. Hence, graphene composites present a potential material to cover these issues [4]. Carbon fiber (CF)-epoxy, and steel are currently used for aerodynamic shields and spare wheel cases in the automobile industry. However, these materials are being replaced by CFs-based graphene nanoplatelets (GNPs) and glass fiber (GFs)-based GNPs to improve impact strength, toughness, and fire resistance. This change also improves recyclability and reduces weight [5].

To ensure a sustainable supply of graphene, it is necessary to use environmentally beneficial production processes. One established method for assessing the environmental impact of products and production processes is life cycle assessment (LCA). Given the anticipated substantial demand for graphene in various applications in the future, it is crucial to examine the environmental consequences of graphene and its manufacturing processes [3]. This work investigates the link between the Environmental Footprint and energy consumption.

LCA is a well-established method for evaluating the environmental impact of products by quantifying both environmental and resource effects. This approach allows for the evaluation of impacts throughout a product's entire life cycle, from raw material extraction to production, use, and eventual disposal [3]. LCAs have mainly focused on automotive, chemical,electronics, and energy industries. The primary challenges for conducting LCAs in graphene production are the same as in other fields. The first obstacle involves raising awareness about incorporating the life cycle concept and preventing unintended shifts in environmental impacts. The second major challenge is insufficient input-output inventory data and impact relationship data. Barriers include confidential details about manufacturing processes, a need for toxicological test results, overall data scarcity, and considerable process variability [6].

The LCA studies mentioned here usually mention specific life cycle stages, although they may only cover some. Nevertheless, qualitative and quantitative findings and thorough explanations for any omissions are included. This approach meets the transparency, acceptability, and credibility criteria for such analyses [7, 8]. While manufacturing and usage stages were commonly considered, impacts related to transportation and end-of-life activities were frequently left out or, at most, minimally discussed [6].

Different methods for producing both monolayer and bulk graphene have been explored in research. Various processes are being developed for graphene production, which can be broadly categorized. Due to the diverse production routes in different stages of development, it's crucial to enhance our understanding of their environmental impacts [8]. Choices in production, such as precursor selection and process temperature, can markedly influence life cycle energy consumption and the associated environmental impact [8]. This research has explored various graphene production methods, incorporating data from patents, research papers, and Ecoinvent databases. In this study, utilizing such data from graphene production would enhance the reliability of the modelling, offering additional insights into how production factors influence the life cycle environmental impacts.

1.1 Research Motivation

Despite the evident need for a new generation of intelligent multifunctional materials, driven by the distinctive characteristics of graphene-related materials (GRM) and other 2D materials (2DM), such as composites, coatings, and foams, there remains a significant gap in conducting a life cycle assessment of graphene-related composite materials due to the lack of available graphene database. An available platform must offer the necessary data to establish such a database. Consequently, this study is compelled to concentrate on existing literature and trends to understand the manufacturing processes of graphene and develop its life cycle inventories and database. This approach aids in assessing the Environmental Footprint of the Energy consumption associated with graphene production.

1.2 Research Objective

To address this gap, the initial step involves the identification of inventories and the electricity requirement for graphene production. This information and a detailed examination of energy usage directly influence the Environmental Footprint. Research into literature and identifying patterns have uncovered methods for creating a graphene database. It also helps better understand the environmental effects caused by using energy in producing graphene; the study has explained various production processes employed in graphene and covered methodologies like the chemical reduction process, Hummers method, hydrothermal method, and others. Each production technique employs distinct functional units that help to meet specific objectives. Using LCA for Expert software, the study constructed models for each production process and calculated the Environmental Footprint associated with the inputs and outputs. By analyzing the impact assessment results, each graphene production method was evaluated, and by doing

that, this study has calculated the Product Environmental Footprint (PEF) single score of each production process. Furthermore, this study has interpreted the impact assessment and energy consumption hotspots inherent in these processes, recognizing them as a pivotal aspect, particularly in environmental Footprint. Finally, the study outlines prospects and discusses limitations encountered, such as the lack of comprehensive data on specific production methods, providing insights for further research in this domain.

1.3 Thesis Framework

Chapter 1 serves as an introduction, setting the stage for the study by introducing the topic, explaining the motivation for the research, stating the objectives, and providing an overview of the framework of the thesis.

Chapter 2 and 3 extensively reviews the historical context of graphene production and its various applications. It also examines the stages involved in LCA in detail and introduces relevant terminology. The chapter 2 concludes with an explanation of previous analyses conducted on graphene production.

Chapter 4 provides a detailed and unique explanation of the Experiment procedure, including a graphical representation of the production process and its methods. It also covers the innovative modelling of the graphene production process.

Chapter 5 is outlining the Product Environmental Footprint and impact assessment results, where each production process is thoroughly discussed and analyzed.

Chapter 6 summarizes the study's essential findings and insights. It also outlines potential areas for future research that could influence the appropriateness of using life cycle assessment for graphene production.

2. State of the Art

Graphene-based materials have gained significant attention in research due to their exceptional physical, chemical, and thermal properties. Graphene has unlocked vast possibilities and emerged as a focal point of exploration across diverse fields, including aerospace, energy, transportation, healthcare, agriculture, and wastewater treatment technology. Although graphene has shown impressive performance in various applications, current research aims to optimize parameters to achieve the best outcomes with graphene-based materials. Despite significant growth, the graphene industry has yet to fully realize its potential, mainly due to the ongoing challenge of scaling up graphene production, which remains a prominent area of research [9].

The mechanical exfoliation method [10], liquid-phase exfoliation [11], oxidation-reduction method [12], and chemical vapor deposition [13] are among the most commonly utilized manufacturing methods for producing graphene. While the quality of graphene produced by these methods is outstanding, the cost associated with preparation remains a significant constraint. Furthermore, achieving monolayer and high-purity graphene presents a challenge, thus limiting the scalability of production and the realization of its full commercial potential. Graphene, the elemental carbon structure, consists of sp^2-carbon atoms arranged in a two-dimensional (2D) honeycomb lattice structure. It can be synthesized through either top-down processes, such as mechanical, electrochemical, or chemical exfoliation of graphite, or bottom-up methods, including chemical vapor deposition and chemical synthesis [9].

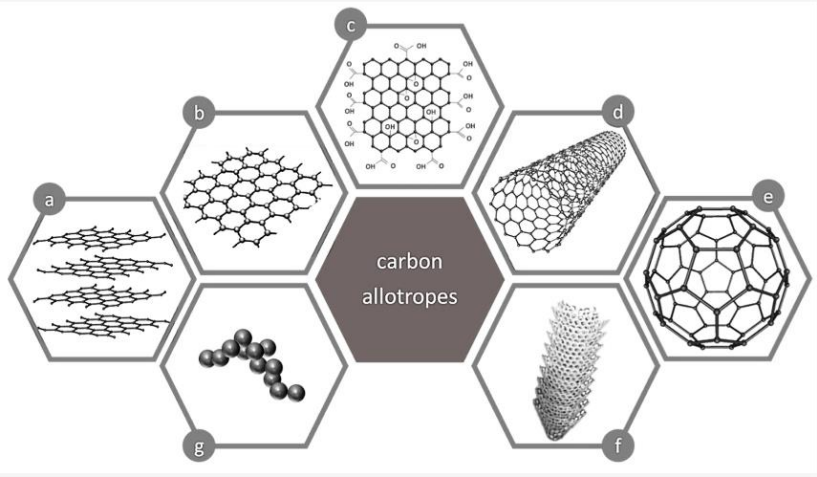

Figure 2: Carbon allotropic forms [14]

As illustrated in Figure 2, 2D graphene serves as the foundational element for all other carbon allotropes, including zero-dimensional (0D) fullerenes, one-dimensional (1D), CNTs, and three-dimensional (3D) graphite. These various allotropes can be derived from graphene through modification processes. Furthermore, graphene possesses remarkable properties: it is lightweight, solid, and rugged, showcasing exceptional electrical and thermal conductivity. It stands out as the thinnest and strongest among nanomaterials. Notably, graphene is not limited to its extreme thermal and electrical conductivity levels and fast electron mobility but also outstanding mechanical strength [9].

- **Graphite:**

  Graphite, a naturally found carbon variant, represents one of carbon's three crystalline allotropes, alongside diamond and fullerenes. Its structure is layered, with two-dimensional hexagonal lattices made up of carbon atoms in each layer. These layers possess weak inter-layer bonds due to van der Waals forces, enabling them to slide easily over one another, thus imparting graphite with its well-known lubricating characteristics [15].

- **Graphene and Graphene Oxide:**

  As explained earlier, Graphene, a two-dimensional material comprising covalently linked sp^2 hybridized carbon atoms, has garnered considerable attention due to its remarkable properties. These include high electronic conductivity, excellent thermal stability, exceptional mechanical strength, and a large surface area, making it particularly suitable for analytical applications. A notable advantage of graphene-based materials over carbon nanotubes (CNTs) is their ability, for specific applications, to be produced from graphite, a readily available and cost-effective material, without the need for metal catalysts. However, alternative preparative methods are required for high-quality applications such as electronics [14].

- **Graphene oxide:**

  Graphene oxide (GO) is a graphene-based material primarily composed of carbon, oxygen, and hydrogen atoms. Its structure differs from that of graphene due to the presence of regions with aliphatic six-membered rings containing hydroxyl, epoxide, carbonyl, and carboxyl groups, in addition to the aromatic regions with unoxidized benzene rings. These oxygen-containing functional groups serve as crucial nucleation sites for subsequent chemical modifications, including decoration and functionalization. These properties make GO an attractive candidate for various applications, including polymer composites, energy-related materials, and sensors [14].

- **Carbon Nanotubes:**
  Carbon nanotubes (CNTs) are unique nanostructures with exceptional electronic and mechanical properties stemming from their close relationship with graphene or one-dimensional appearance. Structurally, Single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) are the two primary categories of carbon nanotubes. SWCNTs are cylindrical tubes made from a single sheet of graphene that has been rolled up, while MWCNTs consist of multiple concentric nanotubes stabilized by van der Waals forces. The presence of concentric graphene sheets in MWCNTs enhances their interaction with analytes, making them valuable in various applications [14].

There is a notable scarcity of comprehensive review articles covering all the production processes of graphene derivatives, including pure and composite graphene fibers, graphene/polymer composites (PCs), and graphene/fiber/polymer composites (gFPCs), as well as their diverse applications across various sectors such as energy, wearable technology, agriculture, wastewater treatment, medical and healthcare, and the automotive industry. Consequently, this review provides an extensive overview of the wide-ranging applications of graphene-based composite materials, accompanied by detailed insights into their fabrication methods. Given graphene's potential to address global concerns related to energy and pollution, understanding the state-of-the-art developments, specific properties, and potential application areas of graphene-based materials becomes imperative [9].

2.1 Introduction to Graphene Production Methods

The production and utilization of graphene-based materials have witnessed a consistent increase over the past two decades. Tables 1 and 2 summarize the number of articles published from 2001 to 2020 concerning alternative methods to produce graphene-based composite materials, including fibers, fabrics, films, polymer composites, and fiber-polymer composites, as well as various production methods employed [9].

Keywords Used for Search	Year Range	No. of Publications
"Graphene based fibres"	2001–2010	57
	2011–2020	3264
"Graphene based fabrics"	2001–2010	0
	2011–2020	410
"Graphene based films"	2001–2010	243
	2011–2020	8658
"Graphene polymer composites"	2001–2010	152
	2011–2020	7655
"Graphene fibre polymer composites"	2001–2010	32
	2011–2020	897

Table 1: Number of articles published between 2001 and 2020 on graphene-based composite materials [9]

Keywords Used for Search	Year Range	No. of Publications
"Wet spinning graphene fibres"	2001–2010	0
	2011–2020	209
"Graphene solution mixing"	2001–2010	13
	2011–2020	719
"Graphene melt blending"	2001–2010	3
	2011–2020	319
"Graphene in situ polymerization"	2001–2010	25
	2011–2020	1817
"Graphene roll to roll milling"	2001–2010	0
	2011–2020	34
"Graphene matrix modification method"	2001–2010	7
	2011–2020	340
"Graphene electrophoretic deposition"	2001–2010	13
	2011–2020	556
"Graphene chemical vapour deposition"	2001–2010	385
	2011–2020	7051
"Graphene chemical grafting"	2001–2010	29
	2011–2020	1341

Table 2: Number of articles published between 2001 and 2020 on different production methods of graphene-based materials [9]

Research interest in graphene-based materials and their manufacturing techniques is steadily growing. According to the author and recent research in the field of composites, the following outline the most utilized approaches for the production of graphene-based materials [9].

2.1.1 Spinning of Graphene Fibres

Graphene fibers are mainly produced using solution spinning techniques, such as wet spinning, dry spinning, and dry jet spinning, due to the impracticality of the melt spinning technique. Wet spinning is widely used for pure graphene and graphene-based composite fibers. To fabricate

pure graphene fibers, graphite must first be converted into GO through chemical or electrochemical exfoliation processes [8]. Chemical exfoliation methods, such as the Hummers and modified Hummers processes, are commonly used [16].

Wet spinning produces GO fibers from a GO solution through a double-diffusion process (Two different substances or components diffuse through a medium or across a boundary simultaneously). Preparing a suitable dope solution is crucial to ensure optimum spinnability, often involving the formation of liquid crystalline graphene oxide (LCGO) dope solution. After spinning, the GO fibers undergo chemical or thermal reduction processes to enhance their electrical conductivity and other properties. Eco-friendly alternatives such as organic acids and plant extracts have replaced hazardous chemicals in chemical reduction. This reduction step is typically more suitable and industrially scalable than thermal reduction [9, 17].

2.1.2 Graphene Polymer Composites (GPCs)

The production of graphene/polymer composites (gPCs) involves various techniques, including solution mixing, melt blending, in situ polymerization, and high shear mixing–calendaring. These techniques affect the dispersion of graphene and its derivatives within the polymer matrices, which affects the performance of the composites [18].

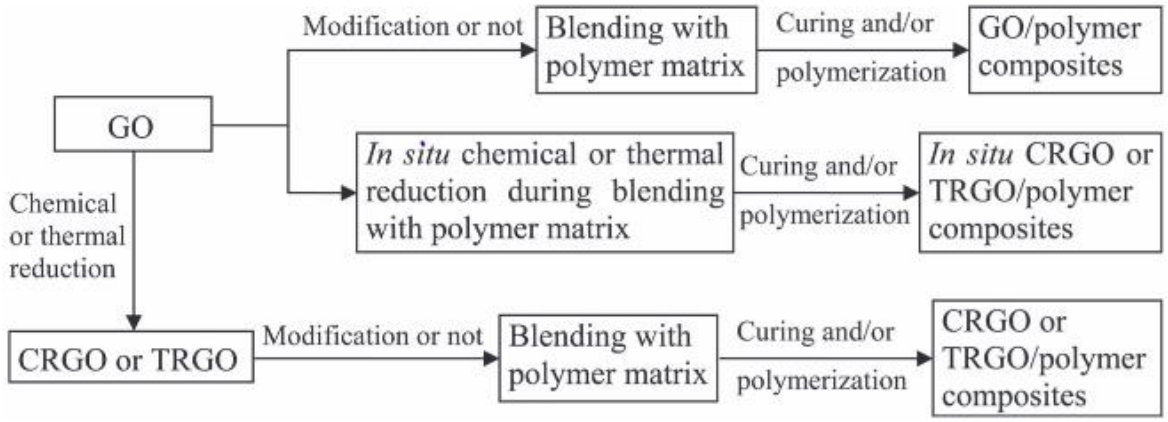

Figure 3: The general fabrication routes for polymer-based composites with GO or RGO as fill [19]

The figure illustrates the production pathways for graphene oxide (GO)/polymer composites and the transformation into in situ chemically reduced graphene oxide (CRGO) or thermally reduced graphene oxide (TRGO) polymer composites. Essential factors in the synthesis of composites include the molecular weight, polarity, hydrophobicity, and reactive groups of the polymer resin, graphene nano-fillers, and solvent. Graphene's hydrophobic nature limits its dispersion in aqueous solvents, so derivatives such as GO, chemically reduced graphene oxide

(CRGO), and thermally reduced graphene oxide (TRGO) are used as fillers in composite fabrication [9].

2.1.3 Graphene/Fibre/Polymer Composites (gFPCs)

Graphene nanomaterials can be added to fiber-reinforced polymer composites (FRPCs) using three main methods. The 'Matrix Modification Method' involves mixing graphene with a suitable polymer and applying it to the reinforcement fibers using techniques such as dip coating, hand lay-up, or spray-up. The 'Fiber Modification Method' involves integrating the graphene nanomaterial directly onto the fiber surface using techniques such as electrophoretic deposition (EPD), chemical vapor deposition, or chemical grafting. This method combines both approaches, enabling graphene incorporation into both the fiber and polymer matrix simultaneously to enhance composite properties for specific applications [9].

2.2 Applications of Graphene-Based Materials

Graphene-based materials have exceptional structural and functional properties, making them widely used in diverse advanced applications. Research on graphene in recent decades has facilitated its use across various fields, from agriculture to aerospace. Notable applications include solar cells, supercapacitors, Li-ion batteries, microbial fuel cells, sensors, and nanomembranes for wastewater treatment. Graphene nanomaterials are used in various medical applications, including drug delivery systems, gene therapy, DNA sequencing, tissue engineering, bio-imaging, and potential cancer therapies. Additionally, graphene-incorporated fiber-reinforced composites are becoming increasingly popular in the automotive, marine, and aerospace industries due to their high strength-to-weight ratio properties [9].

2.2.1 Automobile, Marine and Aerospace Industry

The structural engineering sectors, particularly automotive, aerospace, and marine industries, prioritize lightweight yet durable materials to reduce part weight, fuel consumption, costs, and environmental impact. Fiber-reinforced composites, utilizing high-strength fibers like glass and carbon, are replacing heavy steel components. Thermosetting polymer resins serve as the matrix due to their chemical stability, but they have limitations in thickness properties and susceptibility to crack initiation under cyclic loads. To overcome these challenges, graphene nanomaterials are integrated into fiber/polymer composites, enhancing both in-plane and out-of-plane properties and improving strength-to-weight ratio and mechanical characteristics [3].

Due to their improved interlaminar shear strength and fracture toughness, graphene/carbon fiber/polymer composites are used in various aircraft parts, including ribs, fuselages, and wings. However, cost remains a limiting factor, and further research is needed to reduce costs without compromising quality [3].

2.2.2 Agriculture and Wastewater Management

Even though water is abundant on Earth's surface, water pollution from numerous sources constitutes a severe danger to the sustainability of the ecosystem, resulting in the extinction of aquatic species and a shortage of clean water supplies. Numerous techniques are used to eliminate water pollution and address this problem. Nanoporous membranes based on graphene are an effective method of eliminating many types of contaminants. In wastewater treatment, nanoparticles are efficient barriers to liquid and gaseous materials [9].

3. Methods and Materials

3.1 Life Cycle Assessment

LCA is an approach used to evaluate a product or process's sustainability and environmental impact. For instance, when evaluating a manufactured product, the life cycle includes from cradle to grave, starting from raw material extraction, production, usage, recycling, and disposal. Moreover, its environmental impacts are assessed across various stages, including manufacturing, distribution, usage, recycling, and final disposal. The life cycle can span from cradle to gate to eliminate the grave step, depending on the decision of LCA practitioners. LCA has four interrelated phases by ISO 14040 standards [20].

1. Goal and scope definition

2. Inventory analysis

3. Impact assessment

4. Interpretation

Each stage is not ranked in a hierarchy but is part of a process that involves ongoing revisions. As LCA is repeated with better data and more effort, the overall data quality improves. LCA utilizing databases like eco-invent, Agri-footprint, USLCI, etc. Practitioners also need to choose impact assessment methods, such as Environmental Footprint (EF) 3.1, IPCC GWP 100a, CML-IA, ILCD Midpoint, etc., can be conducted manually or with software such as SimaPro, OpenLCA, LCA for Expert, etc., based on their specific objectives [21].

3.1.1 Goal and Scope Definition

As per the International Life Cycle Data System (ILCD) handbook guidelines, having a clear and well-defined goal at the outset of conducting an LCA is essential [21]. This is the first and most crucial stage of the LCA process, as it lays the foundation for all subsequent steps in the assessment. Establishing a clear goal makes it easier to define the scope of the assessment, the data that needs to be collected, and the methodology used to analyze the data. The first step of any LCA study is defining the goal and scope, where the purpose of the study and the intended audience are stated. These aspects significantly impact methodological decisions throughout the LCA study [20].

Additionally, setting up a system boundary is crucial to clarify which processes are included in the studied product system. This study has three primary goals:

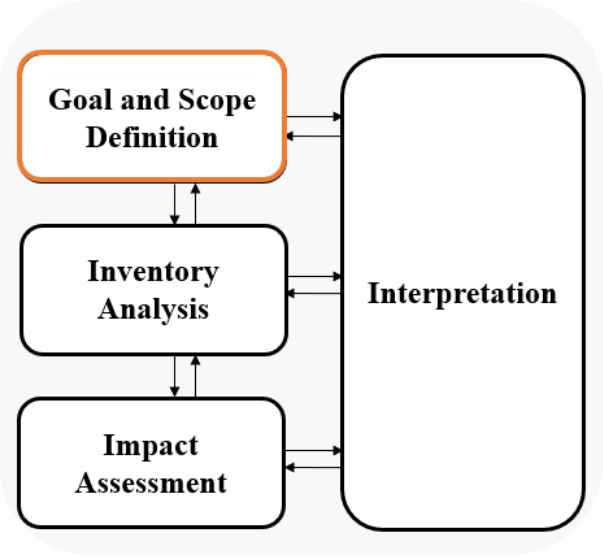

Figure 4: Goal and scope definition [19]

Figure illustrates the Goal and Scope definition of the study. The goal of conducting an LCA defines the specific purpose behind undertaking the assessment. It addresses the question: What are we aiming to accomplish through this analysis? The goal typically entails pinpointing the intended use of the LCA findings and steering the overall focus of the study.

This study has three primary goals:

1. It aims to find inventories for a graphene database, as no such database currently exists in any LCA database platform.
2. It seeks to analyze the energy consumption and evaluate the environmental impacts of various graphene production methods.
3. The study aims to assess how energy consumption influences the product's Environmental Footprint.

The scope of the study is to evaluate the cradle-to-gate life cycle impacts of various graphene production methods. This includes assessing activities from raw material extraction and processing to graphene production. However, the study does not consider the transportation of materials due to the laboratory stage and low technology readiness level (TRL) of the mentioned production processes. Additionally, specific applications were not taken into account, resulting

in the exclusion of the use phase and end-of-life activities from the study. The functional unit is a defined measure of the function or service provided by a product or system. It serves as the reference quantity against which the environmental performance of alternative products or systems can be evaluated. The functional unit for this study was set to 1 kg of Graphene, Nitrogen-doped reduced graphene oxide (N-rGO), and reduced graphene oxide (rGO). The study utilized the EF 3.1 impact assessment method. This method is specifically chosen to determine the Product Environmental Footprint (PEF) single score of the production process [22, 23].

The figure illustrates three distinct system boundaries commonly used in life cycle assessment methodologies: "cradle-to-cradle," "cradle-to-grave," and "gate-to-gate" [20].

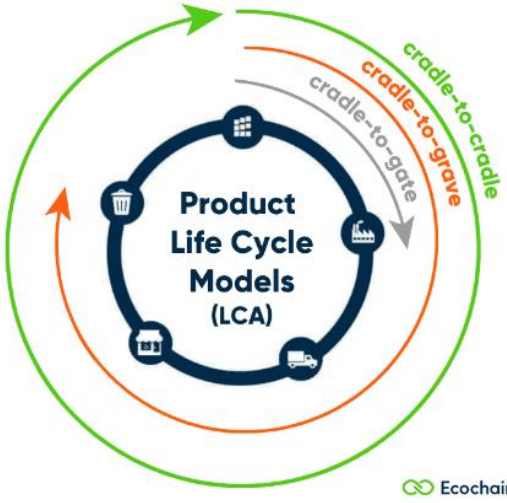

Figure 5: System boundary of life-cycle assessment [20]

System Boundary:

- **Cradle to Cradle:**

  Cradle to Cradle approach in a LCA, the system boundary extends from the extraction of raw materials ("cradle") through the entire life cycle of the product, including production, use, and end-of-life stages, where materials are ideally recovered and reused to create same products ("cradle"). The concept emphasizes continuously cycling materials and resources in closed-loop systems to eliminate waste and pollution [21, 24].

- **Cradle to Grave:**

  The cradle-to-grave approach describes the entire life cycle of a product or material, encompassing all stages from its creation ("cradle") to its disposal or end-of-life management ("grave"). Unlike the cradle-to-cradle approach, which promotes circularity and continuous recycling of materials, cradle-to-grave acknowledges that many products follow a linear trajectory where they are produced, used, and disposed of as waste [21, 24].

- **Cradle to Gate:**

  Cradle to gate refers to a partial life cycle assessment boundary that focuses on the stages of a product's life cycle from its creation ("cradle") to the point where it leaves the manufacturing facility ("gate"). This approach excludes the use phase and end-of-life considerations from the assessment [21, 24].

- **Gate to Gate:**

  Gate-to-gate refers to a life cycle assessment boundary that encompasses only the manufacturing process of a product, starting from when raw materials enter the manufacturing facility (the "gate") to when the finished product exits the facility (another "gate"). This boundary excludes the extraction of raw materials, transportation of materials to the facility, use phase, and end-of-life considerations [21, 24].

3.1.2 Inventory Analysis

Inventory analysis is a critical and often time-consuming step in LCA, following the definition of the goal and scope. The analysis involves gathering and compiling data on elementary flows from all sources. The life cycle assessment's further stages are built upon the collected inventory of elementary flows, which is the outcome [25]. This involves:

1. Identifying processes to be included in the Life Cycle Inventory (LCI) model of the product system.
2. Planning and gathering data required for the inventory analysis.
3. Developing and ensuring the quality of unit processes within the LCI model.
4. Building the LCI model and computing the results based on the compiled data.

During inventory analysis, data is collected on various inputs such as electricity, heat, and raw materials used in production. Similarly, information is gathered on outputs, including emissions

such as carbon dioxide, nitrogen oxides, and hazardous chemicals, as well as by-products and waste generated throughout the life cycle of the product or process [24].

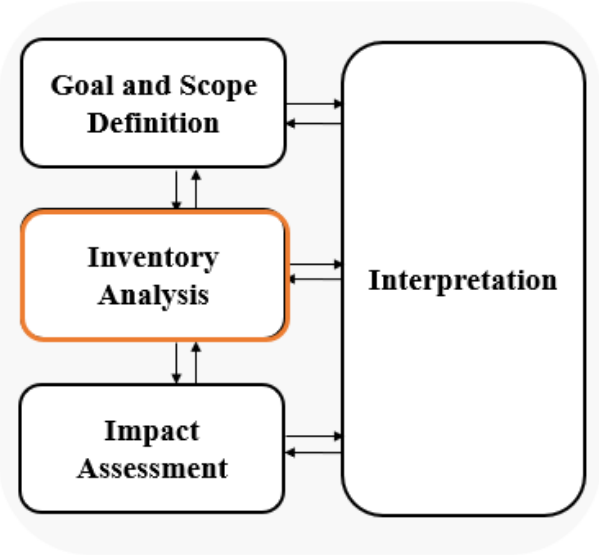

Figure 6: Inventory Analysis [19]

Above figure illustrates Inventory analysis of the life cycle assessment. In Chapter 4, all the inventories of production processes are documented. The collected data is then put into LCA for Expert software to calculate the flows for the product system based on 1 kg of Graphene, N-rGO, and rGO. This assessment is referred to as the Gate to Gate inventories assessment, where the focus is on evaluating the environmental impacts of the production processes from the "gate" of raw material extraction to the "gate" of graphene production [24].

3.1.3 Impact Assessment

This phase represents the third stage of the LCA study, known as Life Cycle Impact Assessment (LCIA), where the data collected in the life cycle inventory phase regarding elementary flows is converted into environmental impact indicators. Unlike the previous phases of LCA, LCIA is predominantly automated through specialized software. However, practitioners must understand the underlying principles, models, and factors to interpret the results accurately and derive meaningful insights from the assessment [25].

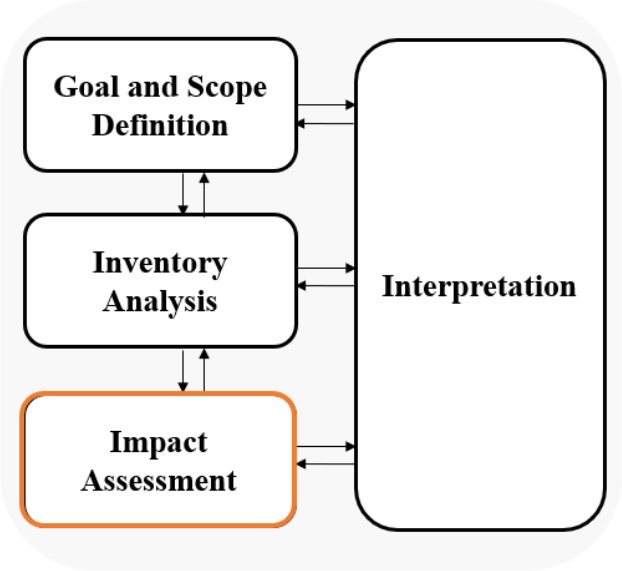

Figure 7: Impact Assessment [19]

This figure illustrates the complete life cycle of the assessed product or process, including all relevant inputs, outputs, and environmental impacts. During the impact assessment phase, each environmental impact associated with the inventories of graphene production is calculated. Classification involves determining which impact indicators have the most significant influence during the production process [24].

Additionally, normalization and weighting factors are applied to the impact assessment results of each production process [22]. This helps to derive a Product PEF single score, which provides a comprehensive understanding of the environmental load associated with each production process through an absolute single score value [23]. The selection of EF 3.1 as the method for measuring the impact of graphene production aligns with the study's focus on the assessment of Environmental Footprint [22].

3.1.4 Interpretation

The interpretation is the final phase of an LCA, when conducting an LCA study to address the goal definition, the interpreting phase takes the iterative steps. During this phase, the results obtained from the LCA model are analyzed in detail to derive robust conclusions and recommendations. This examination involves identifying the significant contributions of the process or product to specific environmental impact categories. By analyzing these contributions, areas for improvement can be identified, aiming to minimize energy consumption and reduce environmental impact categories such as Climate Change – total emissions, Water

use, and Resource use, fossils. The interpretation phase is a critical stage in the LCA process, guiding decision-making and facilitating continuous improvement efforts toward greater sustainability [21, 24].

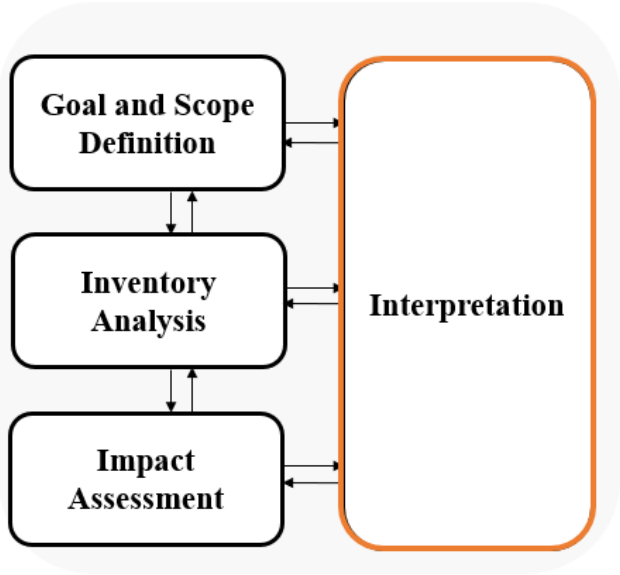

Figure 8: Interpretation [19]

The given figure illustrates the critical process of interpretation within a life cycle assessment, where the comprehensive inventory data and impact assessment results are analyzed and synthesized to derive meaningful conclusions and recommendations regarding the environmental performance of a product or process.

Chapter 5 provides a detailed discussion on identifying the most influential factors affecting the Environmental Footprint and pinpointing where energy is mainly consumed during production. The chapter analyzes various input indicators and highlights vital contributors to environmental impact. It also examines energy usage patterns, revealing critical areas of high energy consumption. This analysis helps to guide efforts to minimize environmental impact and optimize energy efficiency in the production process [21, 24].

3.2 Life cycle assessment studies of graphene production

There are four LCA studies that have been conducted on different graphene production methods. These include chemical reduction of graphite oxide, ultrasonic exfoliation, thermal exfoliation, CVD, and epitaxial growth. These methods represent some of the most commonly

patented production processes for graphene. However, there are also specific variations of liquid exfoliation, CVD, and epitaxial growth. The table presents a comparative analysis of four separate LCA studies conducted on graphene production, each emphasizing distinct impact categories, providing a comprehensive overview of the environmental implications associated with various aspects of graphene manufacturing [3].

Study characteristics	**Chemical reduction of graphite oxide** [26]	**Ultrasonication exfoliation** [26]	**Thermal exfoliation** [27]	**Chemical vapour deposition** [28]	**Epitaxial growth** [29]
Graphene produced	Reduced graphene sheets in water	Graphene sheets in diethyl ether	Pulverized graphite nanoplatelets	Graphene on a quartz substrate	Graphene on a silicon carbide wafer
Carbon feedstock	Graphite	Graphite	Graphite	Methane	Silicon carbide
Functional unit	1 kg	1 kg	1 kg	1 cm^2	1 cm^2
Number of impact categories considered	4: Energy use, water use, human toxicity and ecotoxicity	4: Energy use, water use, human toxicity and ecotoxicity	12, including: Energy use, climate change, acidification, human toxicity and ecotoxicity	2: Energy use and metal use	4: Energy use, climate change, acidification and ecotoxicity
Step with largest contribution to impacts	Reduction step (Hummers' process for water use)	Diethyl ether solvent production	Microwave heating	Methane production	Silicon carbide wafer production
Energy use results	900-1000 MJ/kg	70-500 MJ/kg	2000 MJ/kg	0.007-0.2 MJ/cm2	2-80 MJ/cm2

Table 3: Summary of existing life cycle assessment studies of five graphene production processes

3.2.1 LCA of chemical reduction of graphite oxide and ultrasonication exfoliation

Arvidsson et al. [26] compared the life cycle impacts of two liquid exfoliation processes for graphene production. One method involved the chemical reduction of graphite oxide using hydrazine, as outlined in a patent [27]. The other method involved ultrasonication of graphite, as described in another patent [30]. Different modifications of the Hummers process were assessed to produce graphite oxide from graphite [31], using synthetic and natural graphite as potential raw materials. In the ultrasonication process, diethyl ether was chosen as the solvent due to its widespread industry use and favorable surface properties that aid in exfoliation. Diethyl ether's extensive industrial use and advantageous surface characteristics that facilitate exfoliation led to its selection as the solvent for the ultrasonication procedure [30]. The

functional unit of the study was 1 kilogram of graphene dissolved in a solution. Water was used to reduce the graphite oxide process, and diethyl ether was used for ultrasonication [26].

In this study, four impact categories were evaluated: Energy use, water use, human toxicity, and ecotoxicity. The study found it difficult to differentiate between the environmental performance of the two processes based on the examined baseline cases. Ultrasonication utilized around half the amount of energy and water compared to the chemical reduction of graphite oxide. However, it resulted in approximately twice the level of human toxicity impacts. Ecotoxicity impacts were found to be similar for both processes [3].

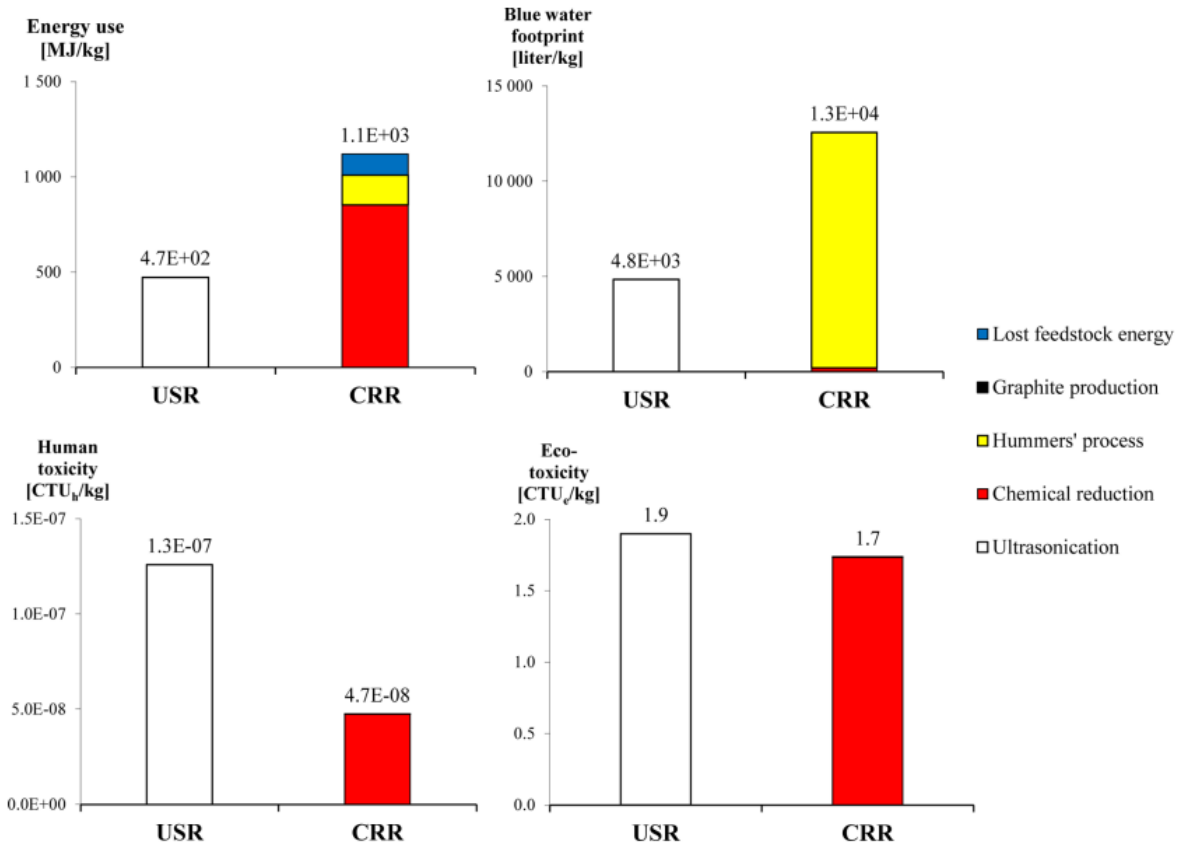

Figure 9: Graphene: USR vs CRR - Energy, water, toxicity, ecotoxicity comparisons succinctly [23]

The figure illustrates distinct impact assessment categories compared to the USR and CRR methods. This comparison highlights the varying environmental impacts assessed by each technique within the analyzed system or process context. During the chemical reduction of graphite oxide, the chemical reduction reaction itself was the main contributor to energy use (approximately 80%), human toxicity (almost 100%), and ecotoxicity (nearly 100%), as shown in figure. Similarly, the Hummers process was responsible for most water use (around 80%). In the case of ultrasonication, the production of diethyl ether solvent accounted for the largest share of all impact categories (almost 100%). The study's primary recommendation was that

industry and research should prioritize the development of ultrasonication techniques with efficient solvent recovery. This approach seeks to reduce the Environmental Footprint associated with liquid graphene exfoliation [3].

3.2.2 LCA of thermal exfoliation

Pizza et al. [27] performed a cradle-to-grave evaluation of an epoxy-based nanocomposite material enhanced with graphene nanoplatelets (GnP) to enhance thermal conductivity. The study's functional unit was defined as 1 kilogram of the nanocomposite. As part of their investigation, Pizza et al. [27] also conducted a cradle-to-gate assessment of GnP production, and the results were presented for 1 kg of GnP. The production of GnP significantly contributes to the overall environmental impacts of the nanocomposite. The impact assessment considered a range of categories, such as metal and fossil depletion, energy consumption, water depletion, Climate change, Ozone depletion, Human toxicity, Ecotoxicity, Photochemical oxidant formation, Acidification, Eutrophication freshwater, marine, and hazardous waste. The results of the thermal exfoliation process show that microwave heating consumes most of the energy in GnP production (>90%). The authors state that this step and its duration are crucial for achieving effective exfoliation. Pulverization accounts for approximately 3% of energy consumption and is the second largest contributor to energy use [3].

3.2.3 LCA of chemical vapor deposition

Arvidsson et al. [26] studied graphene produced via CVD. The study used methane as the carbon source and copper as the substrate metal, supplemented with hydrogen gas to maintain a reducing environment. The investigation was based on experimental research conducted by a University of Texas at Austin team. The functional unit for assessment was defined as 1cm^2 of graphene deposited on a quartz substrate. The study focused on two main impact categories: energy consumption and the use of scarce metals. It compared the resource impacts of graphene produced via CVD to those of Indium Tin Oxide (ITO), a material frequently used in transparent electrodes. Previous research had estimated ITO's life cycle energy consumption to be between 18 and 68 kJ/cm^2 [28]. The CVD-produced graphene has a baseline finding of 22 kJ/cm^2 towards the lower end of the energy consumption spectrum observed for ITO [3].

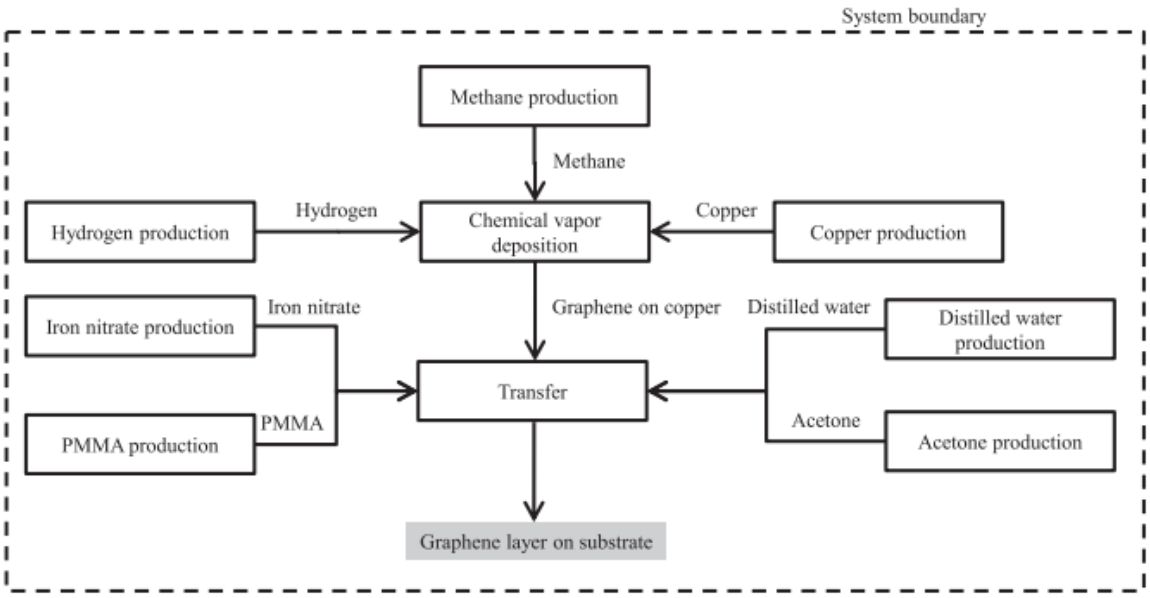

Figure 10: Graphene Layer Production Flowchart [25]

The analysis showed that by recovering excess methane feedstock and minimizing residence time in the reactor, the energy consumption of graphene produced via CVD could decrease to as low as 6.6 kJ/cm². Additionally, CVD-produced graphene was found to be advantageous compared to ITO when considering the use of scarce metals. Although copper is necessary for graphene production, the significantly scarcer material, indium, is required for ITO production. Therefore, CVD-produced graphene seems to be a more favourable alternative to ITO in terms of both energy consumption and the use of scarce metals [3].

3.2.4 LCA of epitaxial growth

Arvidsson and Molander [29] assessed epitaxial growth using a functional unit of 1 cm². The study investigated three production scenarios: lab-scale, pilot-scale, and industrial-scale. The lab-scale scenario drew upon experimental data, whereas the pilot-scale scenario was extrapolated from a modest production facility in Sweden. The industrial-scale scenario was modelled based on assumptions about future advancements. A high-quality silicon carbide wafer with alternating layers of silicon and carbon is required for the process [3].

The study examined the impact of energy use, climate change, acidification, and ecotoxicity. The results showed significant differences between the best and worst cases. The study found that electricity consumption during silicon carbide wafer production was the main contributor

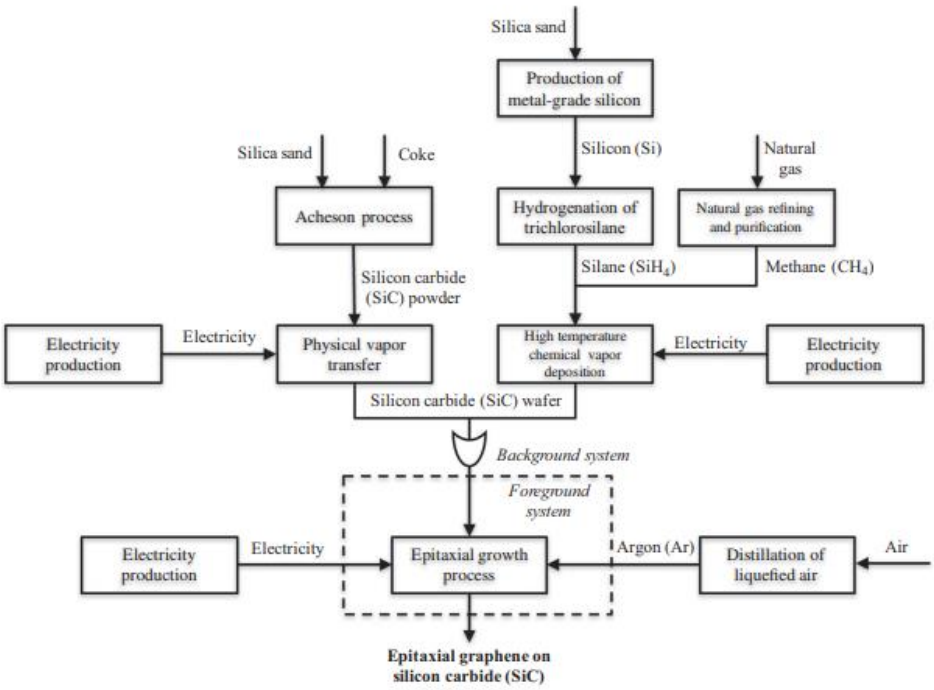

Figure 11: Cradle-to-Gate Life Cycle of Epitaxial Graphene Process Flowchart [26]

to all impacts, accounting for over 70%. Reducing the quantity of silicon carbide wafers could decrease the impact by up to ten times. Additionally, selecting a less harmful or greener energy mix during production might significantly negatively influence the environment, particularly on issues like acidification, climate change, and ecotoxicity, all of which are strongly impacted by the electricity mix selection [3].

3.3 LCA for Expert software

LCA for Expert (GaBi) is a software tool for conducting LCA studies. It offers a thorough framework for assessing how processes, goods, or services affect the environment at every life cycle stage, from extracting raw materials to disposing of them at the end of their useful life [32].

- **Inventory Analysis:** The software ensures accurate data collection and organization of material and energy inputs, emissions, and waste generated throughout a product or system's life cycle [32].
- **Impact assessment:** It allows users to assess how their system or product affects the environment in several impact areas, including resource depletion, ecosystem quality, human health, and climate change [32].
- **Normalisation and weighting:** The software facilitates the normalization and weighting of impact categories, allowing users to compare and aggregate results to

produce a single score or indicator representing overall environmental performance [32].

- **Reporting and communication:** The software provides tools for generating reports and visualizations to effectively communicate LCA results to stakeholders, decision-makers, and other interested parties [32].

3.4 Ecoinvent and Sphera database

Integrating the ecoinvent and Sphera database into LCA studies provides researchers with a robust platform for evaluating the environmental impacts of different products and processes. Both database provides comprehensive LCI data for various industries, covering different geographical regions and allowing detailed analysis of resource use, emissions, and other environmental indicators. Its extensive quality assurance procedures ensure data reliability and consistency, making it a valuable resource for researchers conducting LCA studies [33].

Utilizing the Ecoinvent and Sphera database enables researchers to improve the precision and credibility of their findings, thereby advancing comprehension of human activities' environmental impacts and guiding sustainable decision-making processes. Users can track the effects of their products along the supply chain and utilize the data in various ways tailored to their specific requirements [33].

3.5 Environmental Footprint- Method version 3.1

The European Commission developed the Environmental Footprint (EF) collection of methods to measure and discuss the environmental performance of organizations and products throughout their life cycles. EF 3.1 provides a comprehensive framework for assessing environmental impacts across different impact categories, including both intermediate and final categories. This comprehensive approach ensures that a broad spectrum of environmental impacts are considered, providing a holistic view of the product's environmental performance. EF 3.1 is developed and endorsed by the European Commission, giving it credibility and recognition within the European Union. EF 3.1 represents the latest version of the Environmental Footprint methodology, incorporating improvements and refinements based on scientific advances and stakeholder feedback. Using the latest methodology ensures that the assessment reflects the most up-to-date understanding of environmental impacts and assessment methods. [22].

Apart from EF 3.1, there are many other impact assessment methods, but each serves a different purpose. The Intergovernmental Panel on Climate Change (IPCC), its purpose is same as EF3.1 but the Characterization factors (CFs) are different [22]. Another justification for the GIANCE project's adoption of the EF 3.1 impact assessment method is its compliance with project specifications. This thesis represents an integral aspect of the GIANCE project, which necessitates the use of the EF 3.1 methodology [1].

3.6 Technology Readiness Levels

Technology Readiness Levels (TRLs) are a structured measurement system that facilitates evaluations of a technology's maturity level and enables consistent comparisons across various types of technology. NASA has intermittently utilized this approach in its space technology planning for a considerable period and has recently integrated it into the NASA Management Instruction (NMI 7100), which outlines the comprehensive strategy for integrated technology planning within NASA [34].

- TRL 1 – basic principles observed.
- TRL 2 – Here, the technology concept is formulated, marking a significant step forward in its development.
- TRL 3 – experimental proof of concept.
- TRL 4 – technology validated in the lab.
- TRL 5 – This comprehensive approach ensures that a wide range of technologies are validated in relevant environments, including industrially relevant settings in the case of essential technologies that enable.
- TRL 6 – The technology depicted in an appropriate setting (for example, an industrial setting for critical enabling technologies).
- TRL 7 – Demonstrating system prototypes in an operational environment.
- TRL 8 – System fully operational and certified.
- TRL 9 – The actual system validated in a competitive manufacturing environment is particularly critical for enabling technologies or space applications.

3.7 Product Environmental Footprint

Chapter 3 discussed the Environmental Footprint (EF) 3.1 methodology in detail. This methodology quantifies and assesses the environmental impacts of various processes and activities. Building upon this foundation, EF 3.1 was employed as the analytical framework for evaluating the environmental implications of finding a PEF single score. By utilizing EF 3.1, this study can systematically analyzed key impact categories such as climate change, acidification, ecotoxicity, and resource use, providing a comprehensive understanding of the sustainability performance of Graphene production. Below figure illustrates the Life Cycle Impact Assessment (LCIA) process. First, the inputs and outputs of the life cycle inventory are grouped into 16 impact categories characterized by their mid-points. These categories are normalized, where the results are scaled by the total inventory of a reference unit, to express the impact categories as relative shares of the impacts of the analyzed system. The normalized impact categories are then weighted, i.e., each category is multiplied by a factor to represent its

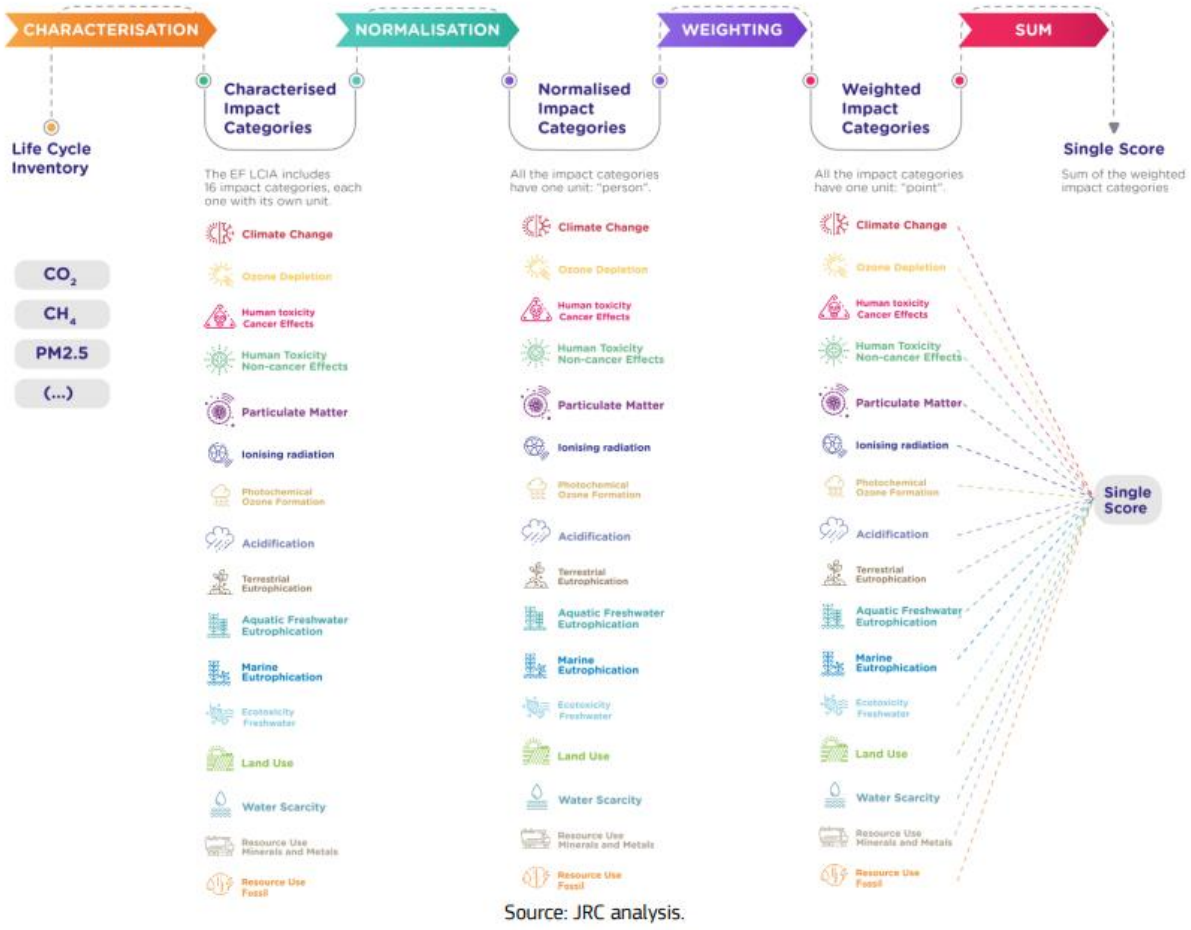

Figure 12: EF3.1 life cycle impact assessment method [1]

perceived relative importance. The weighted impact categories are summed to give a single overall EF score [35].

Impact category	**Indicator**	**Unit**	**Underlying LCIA method**
*Climate change**	Radiative forcing as Global Warming Potential (GWP100)	kg $CO_{2\ eq}$	Bern model - Global warming potential (GWP) over a 100-year time horizon based on IPCC 2021 (Forster et al., 2021).
Ozone depletion	Ozone Depletion Potential (ODP)	kg CFC-11_{eq}	EDIP model based on the ODPs of the World Meteorological Organisation (WMO) over an infinite time horizon (WMO 2014 + integrations)
*Human toxicity, cancer**	Comparative Toxic Unit for humans (CTUh)	CTUh	Based on USEtox2.1 model (Fantke et al. 2017, Rosenbaum et al. 2008), as in Saouter et al. (2018)
*Human toxicity, non-cancer**	Comparative Toxic Unit for humans (CTUh)	CTUh	Based on USEtox2.1 model (Fantke et al. 2017, Rosenbaum et al. 2008), as in Saouter et al. (2018)
Particulate matter	Human health effects associated with exposure to PM2.5.	Disease incidences	PM model (Fantke et al., 2016 in UNEP 2016)
Ionising radiation, human health	Human exposure efficiency relative to U^{235}	kBq U^{235}	Human health effect model as developed by Dreicer et al. (1995) and published in Frischknecht et al. (2000).
Photochemical ozone formation, human health	Tropospheric ozone concentration increase	kg $NMVOC_{eq}$	LOTOS-EUROS model (Van Zelm et al., 2008) as applied in ReCiPe 2008.
*Acidification**	Accumulated Exceedance (AE)	mol H^+_{eq}	Accumulated Exceedance (Seppälä et al. 2006, Posch et al., 2008)
Eutrophication, terrestrial	Accumulated Exceedance (AE)	mol N_{eq}	Accumulated Exceedance (Seppälä et al. 2006, Posch et al., 2008)
Eutrophication, freshwater	Fraction of nutrients reaching freshwater end compartment (P)	kg P_{eq}	EUTREND model (Struijs et al., 2009) as implemented in ReCiPe 2008.
Eutrophication, marine	Fraction of nutrients reaching marine end compartment (N)	kg N_{eq}	EUTREND model (Struijs et al., 2009) as implemented in ReCiPe 2008
*Ecotoxicity, freshwater**	Comparative Toxic Unit for ecosystems (CTUe)	CTUe	Based on USEtox2.1 model (Fantke et al. 2017, Rosenbaum et al. 2008), adapted as in Saouter et al. (2018)
Land use	Soil quality index	Dimensionless (pt)	Soil quality index based on LANCA model (De Laurentiis et al. 2019) and on the LANCA CF version 2.5 (Horn and Maier, 2018)

Water use	User deprivation potential (deprivation-weighted water consumption)	m^3 world eq. deprived water	Available WAter REmaining (AWARE) model (Boulay et al., 2018; UNEP 2016)
Resource use, minerals and metals	Abiotic resource depletion (ADP ultimate reserves)	kg Sb_{eq}	van Oers et al., 2002 as in CML 2002 method, v.4.8
Resource use, fossil	Abiotic resource depletion – fossil fuels (ADP-fossil)	MJ	van Oers et al., 2002 as in CML 2002 method, v.4.8

Table 4: EF3.1 Midpoint Impact Categories and LCIA [44]

3.8 Normalization Factors (NFs)

ISO 14044 provides a structured framework for conducting robust and credible life cycle assessments, including the normalization process. Normalization refers to determining the magnitude of category indicator results by comparing them to reference information. On the other hand, weighting involves converting and potentially combining indicator results across impact categories using numerical factors based on value choices [36]. The normalization factor (NF) for impact category i is determined by summing the products of the inventory (I) of flow j and the characterization factor (CF) of flow j for that specific impact category i [35].

$$NFi = \sum_{j=1}^{x} Ij * CFi,j \text{ [35]}$$

Impact categories	Unit	NF
Acidification	mol H+ eq./person	5.56E+01
Climate change	kg CO_2 eq./person	7.55E+03
Ecotoxicity, freshwater	CTUe/person	5.67E+04
EF-particulate matter	disease incidences/person	5.95E-04
Eutrophication, freshwater	kg P eq./person	1.61E+00
Eutrophication, marine	kg N eq./person	1.95E+01
Eutrophication, terrestrial	mol N eq./person	1.77E+02
Human toxicity, cancer	CTUh/person	1.73E-05
Human toxicity, non-cancer	CTUh/person	1.29E-04
Ionising radiation	kBq U-235 eq./person	4.22E+03
Land use*	pt/person	8.19E+05
Ozone depletion	kg CFC-11 eq./person	5.23E-02
Photochemical ozone formation	kg NMVOC eq./person	4.09E+01
Resource depletion, fossils	MJ/person	6.50E+04
Resource depletion, minerals and metals	kg Sb eq./person	6.36E-02
Water use*	m^3 water eq of deprived water/person	1.15E+04

Table 5: Normalization factor for Environmental Footprint (EF) 3.1 [46, 47]

Normalisation helps provide valuable information in the interpretation phase of LCA. It helps answer the question of whether the results obtained in the assessment are within a credible range in terms of their magnitude [36].

3.9 Weighting

Weighting is allocating relative relevance to various environmental consequences within the context of PEF impact categories. Scientists and researchers have given each category a unique weight, and the European Union is based on factors including overall importance, urgency, effect magnitude, and calculation accuracy (robustness). These weights are referred to as weighting factors [37].

Weighting plays a crucial role in managing different types of environmental impacts. It enables the prioritization and concentration of resources on the most substantial environmental impact categories, aiding decision-makers in focusing on which categories are most important to reduce. In general, climate change in terms of global warming has the highest score in a normalized and weighted set of environmental parameters calculated according to PEF rules. However, other environmental impacts might be more harmful for some materials or production processes [37].

	Aggregated weighting set	**Robustness factors**	**Intermediate Coefficients**	**Final weighting factors (incl. robustness)**
	(A)	(B)	C=A*B	C scaled to 100
Climate change	12.90	0.87	11.18	**21.06**
Ozone depletion	5.58	0.60	3.35	**6.31**
Human toxicity, cancer effects	6.80	0.17	1.13	**2.13**
Human toxicity, non-cancer effects	5.88	0.17	0.98	**1.84**
Particulate matter	5.49	0.87	4.76	**8.96**
Ionizing radiation, human health	5.70	0.47	2.66	**5.01**
Photochemical ozone formation, human health	4.76	0.53	2.54	**4.78**
Acidification	4.94	0.67	3.29	**6.20**
Eutrophication, terrestrial	2.95	0.67	1.97	**3.71**
Eutrophication, freshwater	3.19	0.47	1.49	**2.80**
Eutrophication, marine	2.94	0.53	1.57	**2.96**
Ecotoxicity freshwater	6.12	0.17	1.02	**1.92**
Land use	9.04	0.47	4.22	**7.94**
Water use	9.69	0.47	4.52	**8.51**
Resource use, minerals and metals	6.68	0.60	4.01	**7.55**
Resource use, fossils	7.37	0.60	4.42	**8.32**

Figure 13: Weighting factor for Environmental Footprint (EF) 3.1 [47]

3.10 PEF of Graphene, N-rGO, rGO

A single score in this context refers to a product's overall environmental assessment. The single score is a sum of all the normalized and weighted numbers a product has scored in each impact category. Through the integration of calculations from individual impact categories, a product's overall impact is presented as a single numerical number, which is also known as its Environmental Footprint absolute single score [37]. The process of prioritizing and aggregating the results for the different environmental impact categories assessed in the Environmental Footprint is critical. These impact categories cover many factors, including climate change, acidification, human and eco-toxicity, particulate matter, and impacts related to water, land, and resource use [35].

In the field of Life Cycle Assessment, as described in ISO 14044 (ISO, 2006), normalization and weighting are considered optional steps within the LCIA process. These steps allow practitioners to normalize the results after characterization using a joint reference impact and then combine these results into a single score by assigning different weights to different impacts [35].

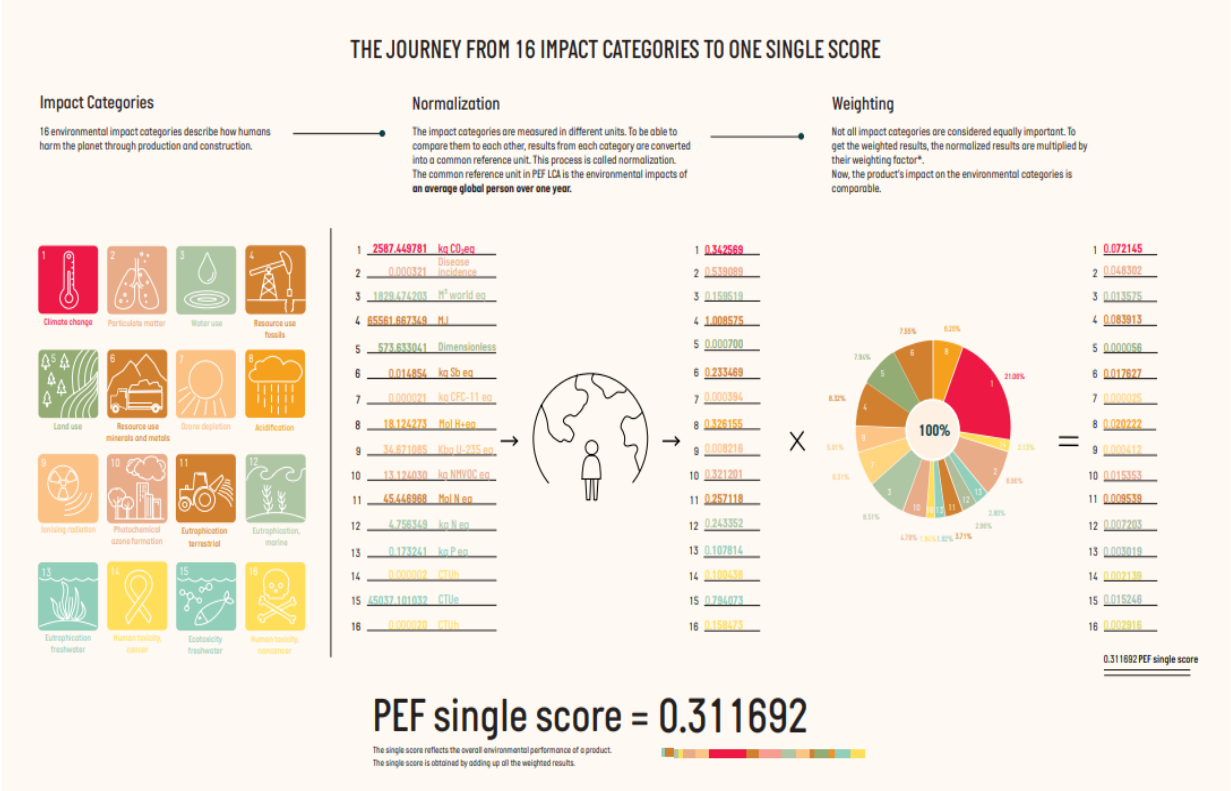

Figure 14: Product Environmental Footprint single score [46]

The figure above demonstrates the methodology of obtaining a product environmental footprint single score through normalization and weighting factors, which involves converting 16 environmental impact categories, measured in different units, into a common reference unit. This process, known as normalization, allows for comparison across categories. Weighting factors are then applied to prioritize certain impacts, resulting in comparable weighted results that reflect the product's overall environmental performance. This single score, derived by summing the weighted results, provides an aggregate assessment of the product's environmental footprint.

3.11 Embodied energy

The embodied energy, is the amount of energy required to produce for example 1 kg of a usable material like steel stock, PET (polyethylene terephthalate) pellets, or cement powder, measured in MJoe/kg (megajoules, oil equivalent per kilogram). Some materials are manufactured using fossil fuels as their primary energy source; for instance, reducing iron ore with coke in a blast furnace. Other materials are produced using electrical energy, a portion of which is typically derived from fossil fuels. Electrical energy needs to be converted into a fossil fuel equivalent, with oil as the standard unit to ensure comparability. In this context, chemical energies (like those from oil) are measured in MJ, while electrical energies are measured in kWh (kilowatt-hours). The conversion rate between these units is 1 kWh = 3.6 MJ (electrical) = 3.6/h MJoe, where h represents the efficiency of converting fossil fuels into electrical energy (usually around 38%) [38].

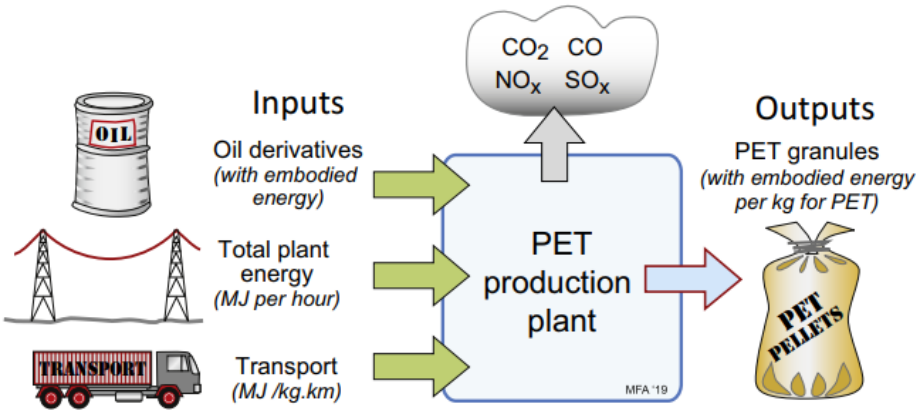

Figure 15: Embodied energy[38]

The material inputs used in any processing operation are commonly referred to as feedstock. These feedstocks can be categorized as either inorganic or organic. Accounting for inorganic feedstock is relatively straightforward because, during processing, it either becomes part of the final product or ends up in waste outputs [38].

However, accounting for hydrocarbons (organic feedstock) is more complex. Hydrocarbons can serve dual roles—as fuel for energy or as a material feedstock. When a hydrocarbon is burned for energy, carbon is immediately released into the atmosphere. On the other hand, if a hydrocarbon is used as a feedstock (e.g., naphtha from oil used in plastics production), the energy content is not lost but instead incorporated into the product. This is what makes it truly "embodied" in the material [38].

Interestingly, the term "embodied energy" does not solely refer to this concept of energy embodied within materials from feedstock use. Instead, it encompasses all energy – both from fuels and feedstocks – associated with material production. This can be a bit confusing because it includes both the energy directly embedded in materials through feedstock use and the energy consumed in the form of fuels during production processes [38].

4. Experiment procedure

In alignment with the LCA principles discussed in the previous chapter, this study delves into the precise definition of functional units and the detailed analysis of production systems for graphene-based materials used as additives in composite materials. In this study, the functional unit is defined as 1 kg of graphene, 1 kg of Nitrogen doped reduced Graphene Oxide(N-rGO), and 1 kg of reduced Graphene Oxide (rGO) in solution, which can be used as an additive in composite materials [26]. This study outlines eight distinct production procedures for producing graphene, each resulting in graphene with varying physiochemical properties. The functional unit of graphene formed by electrochemical exfoliation [8], chemical reduction [26], and ultrasonication [39] produced 1 kg of graphene. The functional unit, 1 kg of N-rGO [2], was produced by the hydrothermal method and annealing processes, and Hummers [16], Marcano's [16], and chemical oxidation processes [8] produced 1kg of rGO. The corresponding production processes and their functional units are listed below.

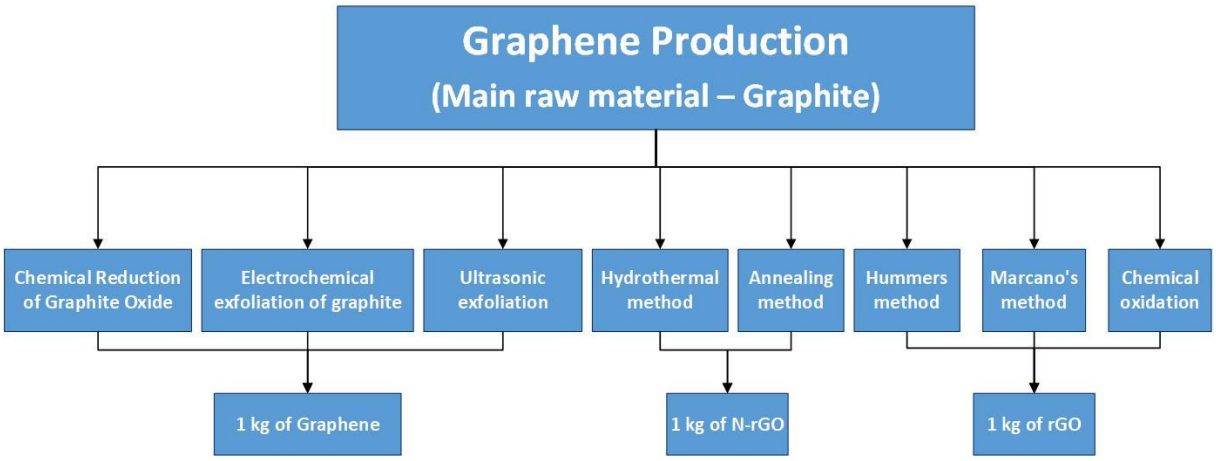

Figure 16: Flowchart of graphene production

Figure 16 shows a flowchart of the eight production routes. The LCA database has economic value for each of the inventories mentioned in the patents and literature obtained from Ecoinvent [40] and Sphera Profesional [41] databases. Patents and literature should disclose production processes that are shown to be technically feasible. These sources provided the majority of the data for graphene production.

The LCA model has been constructed using LCA for Expert software to estimate the environmental impact of graphene manufacturing from raw material extraction to production.

The analysis does not include the usage phase and end-of-life activities, as no specific graphene applications are considered [6]. In addition to that, all production processes mentioned in the study are in the lab scale scenario, which means that the TRL value is set to 3 [42].

4.1 Chemical reduction of graphite oxide

Graphite is the standard raw material for most production routes under study. Synthetic graphite, with a purity level of over 99.9%, or mined graphite, with a purity level of over 93%, can be used [43]. The purity of the graphite does not affect any production route. Because low-quality mined graphite is less expensive, graphite mining provides the foundation for all graphene manufacturing and is thought to represent large-scale production in the future [26]. To convert graphite into graphene using a Chemical Reduction Route (CRR), it must first be oxidized into graphite oxide. The patent proposes the Hummers process, currently the most used method for producing graphite oxide. This is achieved using a combination of sulfuric acid, potassium permanganate, and sodium nitrate. Hydrogen peroxide is also required to reduce excess potassium permanganate during production, while deionized water is required to wash and dilute the graphene oxide [26].

Solvent recovery can be a challenge in the chemical reduction process when using phosphoric and sulfuric acids. Despite this, there is little documentation on the recovery rates in industrial scale, ranging from 0% to 90%. In the baseline case, all acid was neutralized at the end of the procedure due to 0% recovery. It was assumed that the recovery process would involve recycling and reusing the reaction solution, which has minimal environmental impact [26].

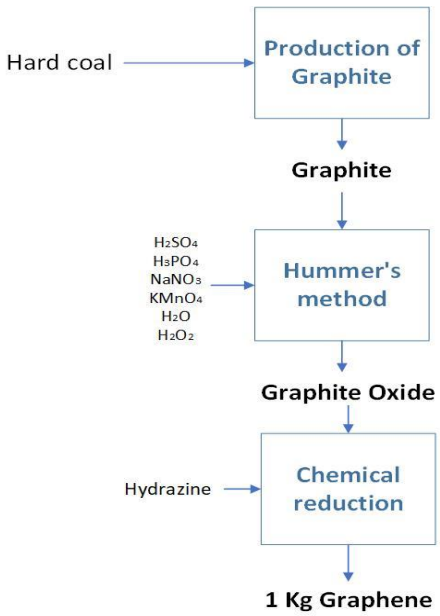

Figure 17: Chemical Reduction Process Life Cycle for Graphene - Cradle to Gate Flowchart [23]

This equation determined the energy used for heating and cooling in this operation based on the input data and tabulated values for heat capacities.

$$E = m \times cp \times \Delta T \quad [26]$$

Where cp is the body's specific heat capacity, m is the body's mass when it is heated or cooled, ΔT is the temperature change that the body undergoes, and E is the energy. The National Institute of Building Services mechanical insulation design guide was used to calculate heat losses [26]. Hydrazine (N_2H_4) is commonly used as a reduction agent to convert graphite oxide to graphene. The specific amount of each input can be shown in Appendix A.1.

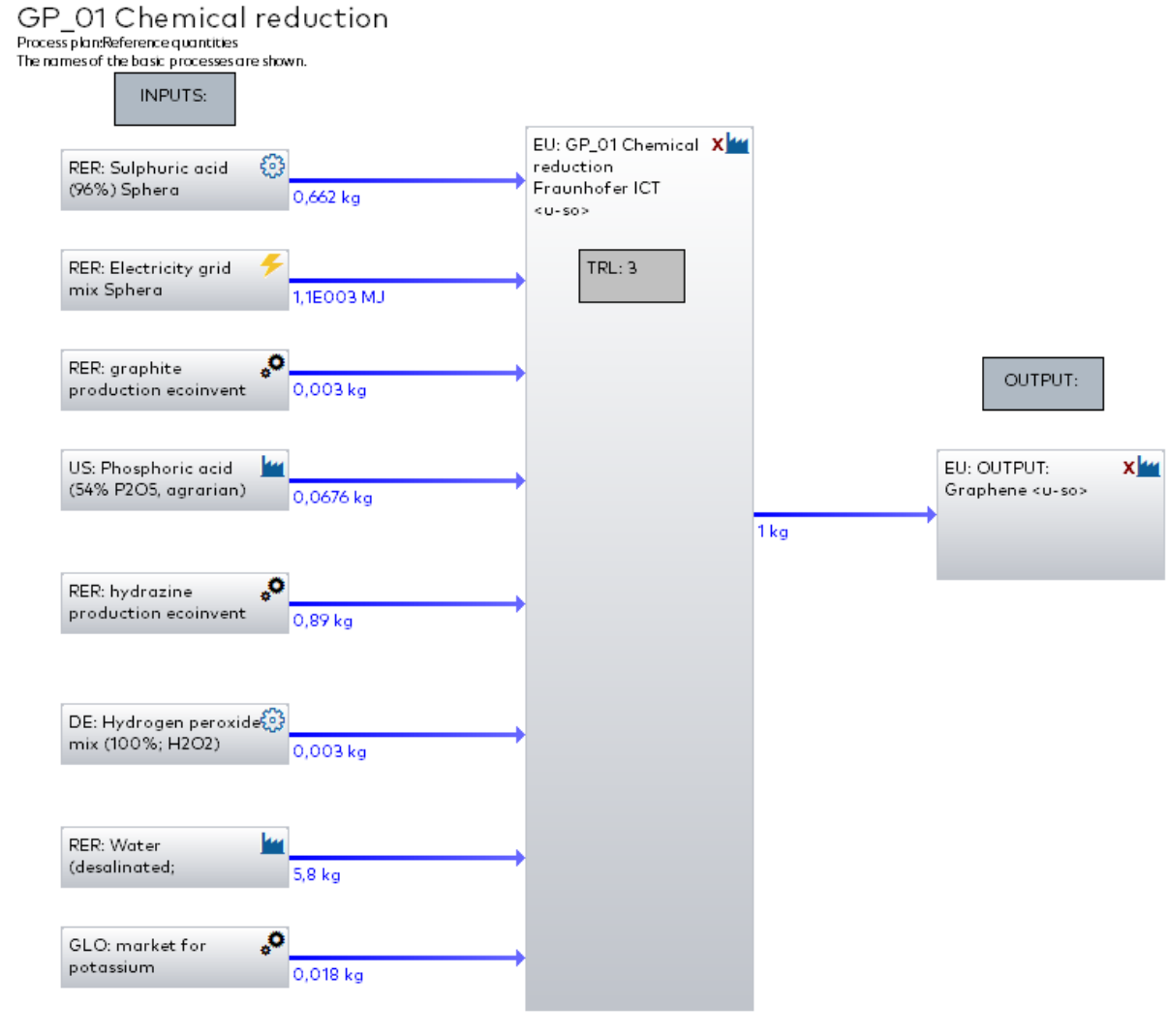

Figure 18: Schematic model of chemical reduction process [44, 45]

4.2 Electrochemical Exfoliation

Graphene is produced through electrochemical exfoliation in an electrolytic cell. The process involves water electrolysis, which produces hydroxyl and oxygen radicals that initiate corrosion at the graphite anode on the edge sides. This reaction opens the graphite edge planes, allowing the electrolyte ions to intercalate and expand the electrode. Consequently, the cohesive Van der Waals forces weaken, causing the electrode material to precipitate from the solution as graphene sheets or nanoribbons, ranging from single to multilayer graphene. To extract additional graphene layers from multilayer graphene, filter the recovered exfoliated material to get a final product. To determine how these factors affect the final production rate, exfoliation is evaluated for a range of electrolytes and operating conditions [8]. During electrochemical exfoliation, the AC-DC power converter is assumed to have 90% efficiency, and the electrode current is monitored. Standard lab equipment, such as scales and beakers, is used to measure electricity [8]. All the information about the inventory used during the process is mentioned in Appendix A.2.

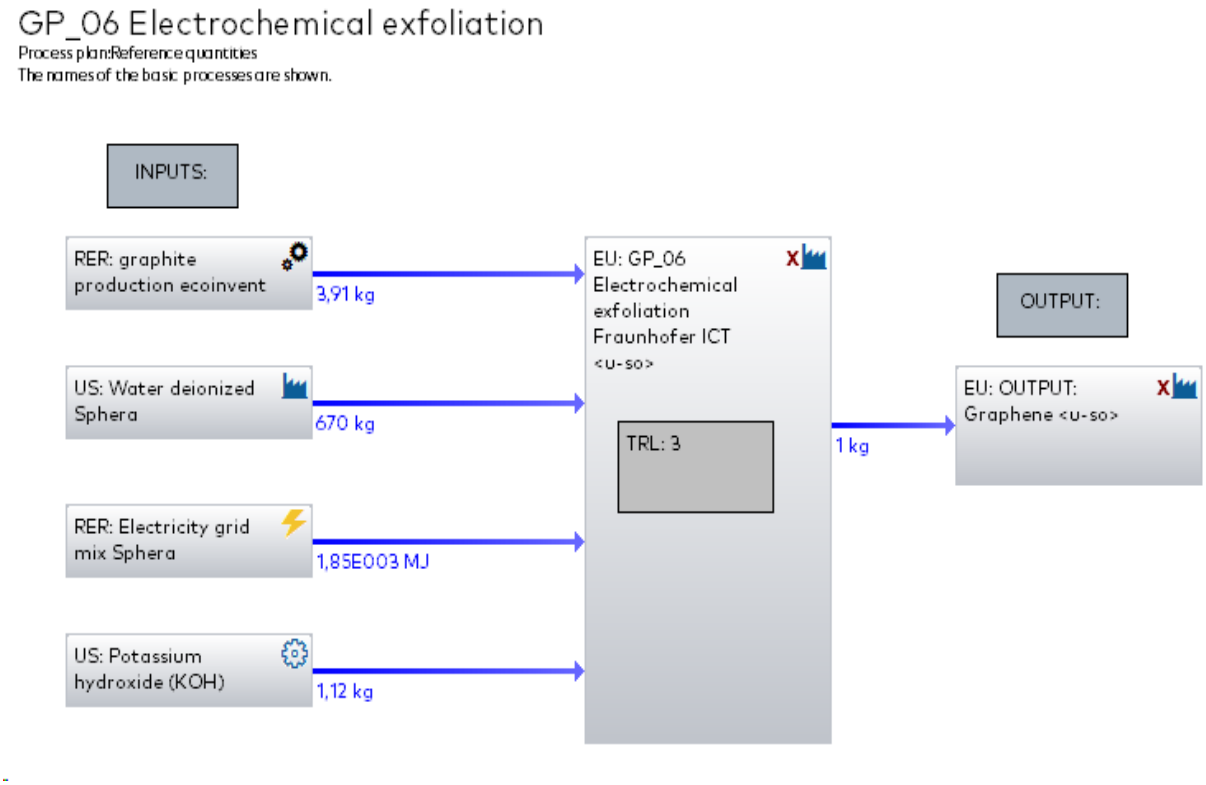

Figure 19: Schematic model of Electrochemical exfoliation [44, 45]

4.3 Ultrasonic Exfoliation

Graphene is produced using graphite, either naturally occurring flakes or Highly Oriented Pyrolytic Graphite (HOPG-artificial), as the primary raw material. To produce graphene, three essential components are required: a binder, a carrier, and a conductive or functional substance. The functional substance is typically carbon in the form of graphite, carbon black, or activated carbon powder, which is electrically conductive. The conductive substance, graphene, is held together by a binder that allows for some flexibility [39].

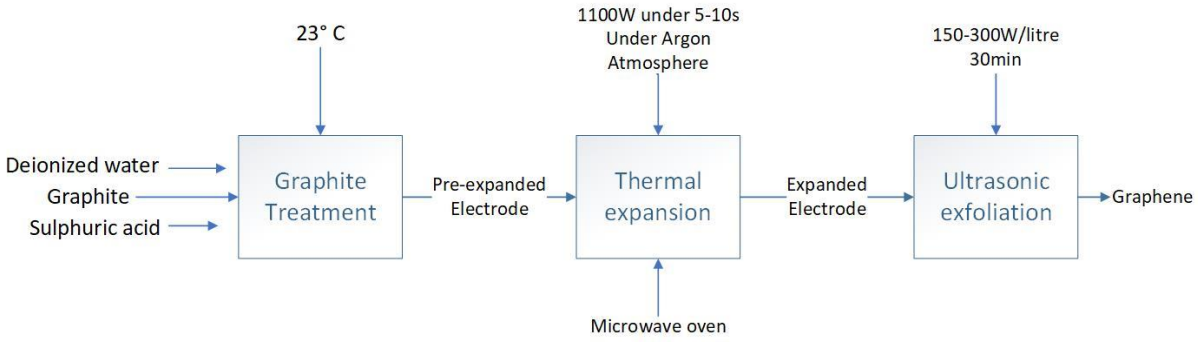

Figure 20: Ultrasonic Exfoliation Process Life Cycle for Graphene [39]

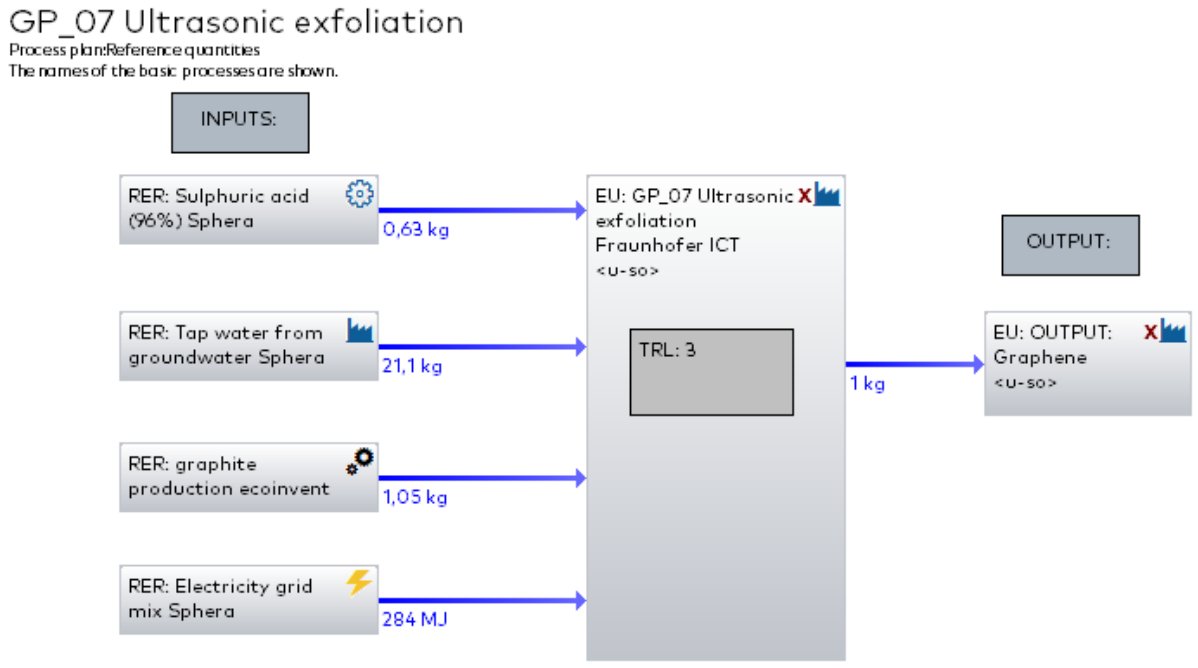

Figure 21: Schematic model of Ultrasonic exfoliation [44, 45]

At 23°C, graphite, deionized water, and sulfuric acid are combined in the precise proportions and amounts listed in Appendix A.3 of the inventory. The resulting mixture is then subjected to a treatment that pre-expands the electrode material. This treatment is carried out using a microwave oven under an argon atmosphere, with the pre-expanded electrode exposed to a power input of 1100W for 5-10 seconds. Once the pre-expanded electrode is obtained, it is further processed in an ultrasonic exfoliation. This exfoliation process uses a power input of 150-300W per liter of solution and lasts 30 minutes. The intermediate product, graphene, is obtained after the ultrasonic exfoliation process. This graphene is then utilized to produce graphene ink, the final product in this literature [39].

4.4 Hydrothermal method

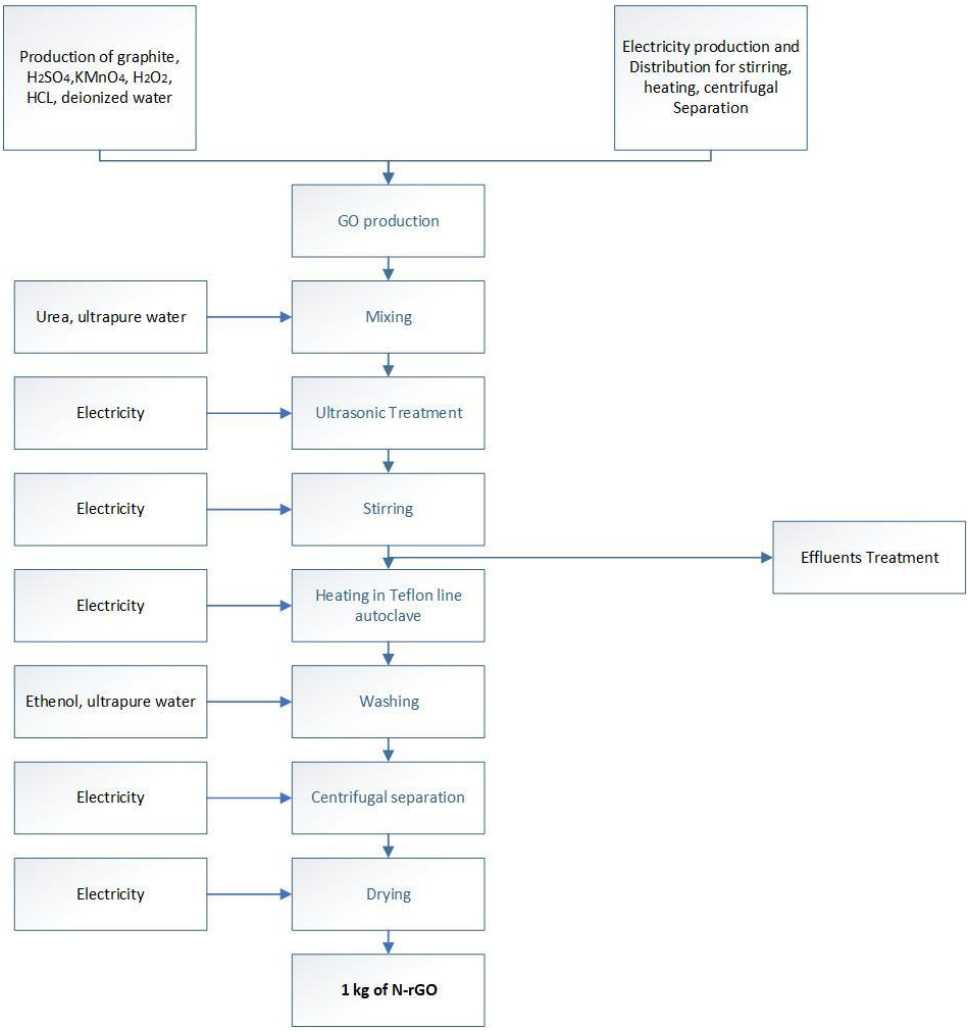

Figure 22: Cradle-to-Gate Life Cycle of N-rGO Produced by Hydrothermal Method (HM) [1]

Modified Hummers' method is used to produce GO, which is used as a basis for synthesizing N-rGO in hydrothermal method (HM) and Annealing method (AM). The Hummers method synthesizes sulfuric acid, permanganate, and graphite. To prevent excessive foaming and to

cleanse GO, deionized water is added. Any residual permanganate is cleaned up with hydrogen peroxide, and then Manganese Dioxide (MnO_2) is added to make soluble Manganese (II)

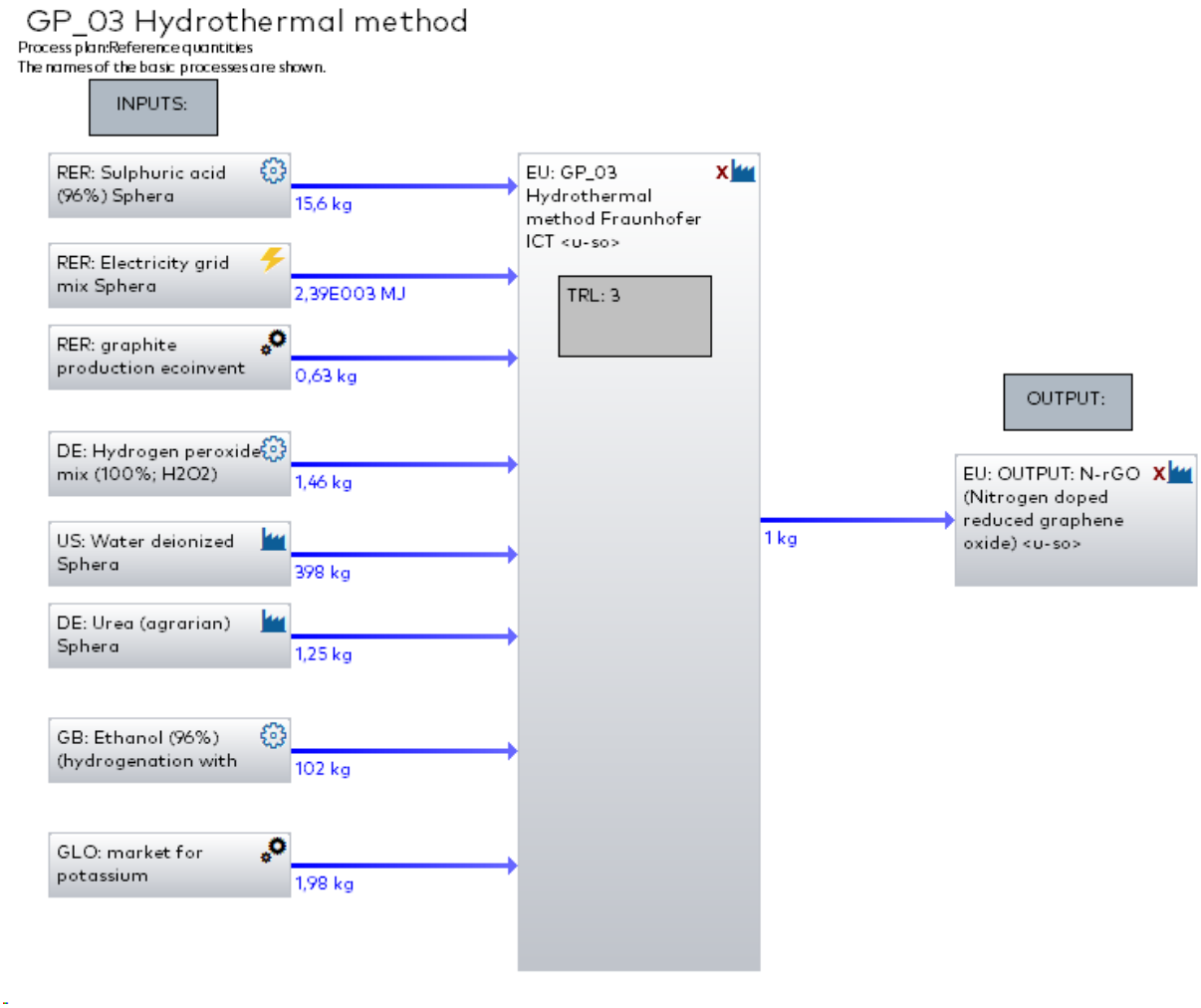

Figure 23: Schematic model of Hydrothermal method [42, 43]

Sulphate ($MnSO_4$). Many operations require electricity, including stirring, heating, centrifugalizing, and drying. Beakers and scales, two standard pieces of laboratory equipment, were used to perform the input inventory to produce GO. Calculations of electricity usage for hot plates and the centrifuge were based on considering both the power factor and the duration of operation. Regarding drying, 100% moisture content was assumed for the graphene oxide. The energy necessary for drying 1 kilogram of water was estimated to be 8 kJ [2].

A mixture of urea and GO is placed inside an autoclave coated with Teflon to create N-rGO. The combination is heated for 18 hours at 180°C. Ultrapure water is employed for the initial mixing of GO and urea and for rinsing the resultant N-rGO. Ethanol is used in the N-rGO washing process subsequently. Electricity is employed in various stages, including stirring, heating, centrifugation, and drying, utilizing equipment such as hot plates, centrifuges, Teflon-

lined autoclaves, ultrasound devices, and ovens. The synthesis inventory data of 1kg N-rGO produced by the Hydrothermal method is described in Appendix A.4 [2].
The electricity consumption for the Teflon-lined autoclave is determined using the following formula:

$$E = \frac{H1+H2}{f} \quad ; \quad H1 = m \times Cp \times \Delta T \quad [2]$$

In this formula, H1 represents the energy needed to heat the materials inside the autoclave, multiplying the mass (m) of the materials being heated by their specific heat capacity (Cp) and the temperature change (ΔT) they undergo. H2 takes into account any heat losses that occur during operation, which are calculated based on the mechanical insulation design guide provided by the National Institute of Building Service [46]. The heat conversion efficiency, denoted as f, is a reliable assumption at 70% [2].

4.5 Annealing method

The same hydrothermal approach is used to synthesize GO, which is the first step in the manufacturing process of graphene. Graphite oxide is transformed into graphene in the next stage. The annealing process involves mixing GO and ammonium nitrate in ethanol, then sonicating and stirring. A hotplate is then used to evaporate the ethanol, and the dry substance that results is pulverized finely. After that, the ground mixture is put into a muffle furnace and heated for an hour at 350ºC. Ethanol and ultrapure water are used to wash the resultant material, referred to as N-rGO [2]. All the input and output data are described in Appendix A.5.

Measuring quantity inputs for producing N-rGO involved utilizing standard laboratory equipment such as beakers and scales. The power factor and usage time were considered when calculating the electricity consumption for appliances such as centrifuges, hot plates, and ultrasound equipment. It was assumed that N-rGO had a 50% moisture content during the drying process, and it consumed 8 kJ of energy to dry 1 kilogram of water. The muffle furnace's electrical usage was determined by a process similar to that used in an autoclave with Teflon lining [2].

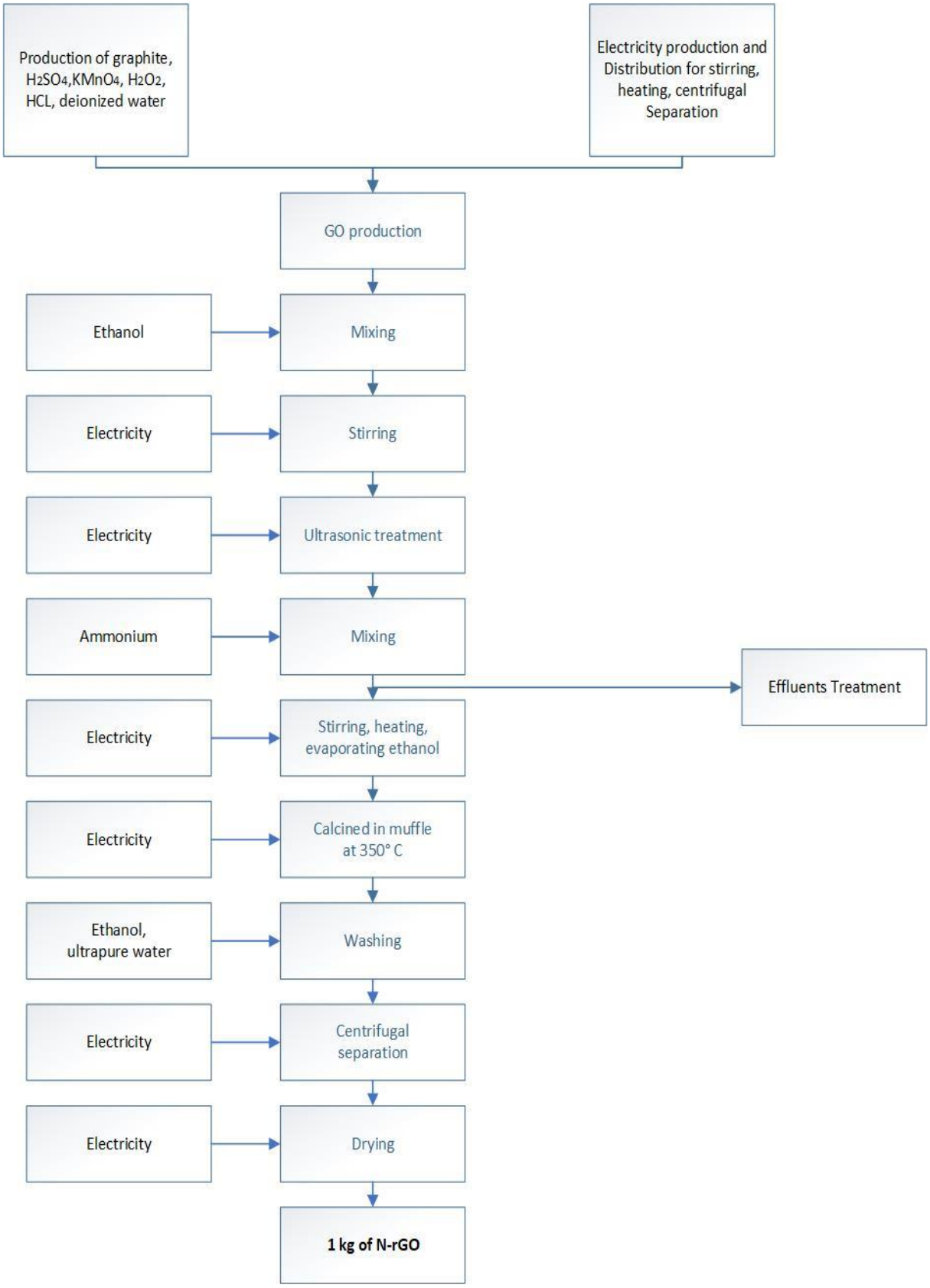

Figure 24: Cradle-to-Gate Life Cycle of N-rGO Produced by Annealing Method (AM) [1]

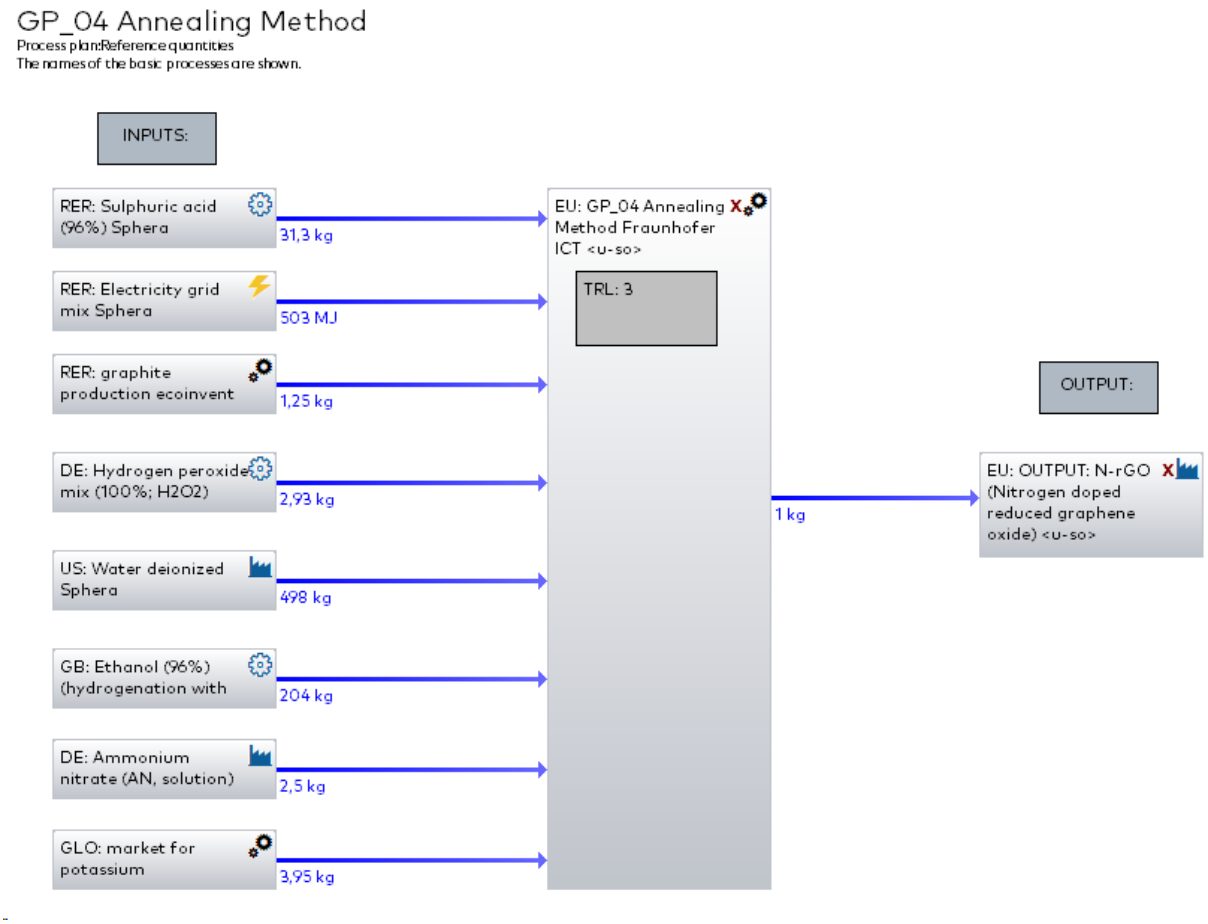

Figure 25: Schematic model of Annealing method [44, 45]

4.6 Hummers method

Graphite oxide was synthesized by stirring a mixture of graphite, sulphuric acid, and sodium nitrate at 5°C for 30 minutes. The resulting mixture was then transferred to a 15 liter container that had been cooled to 0°C using an ice bath for safety. Potassium permanganate was added to the mixture, and the temperature was raised to and maintained at 35 ± 3°C for 30 minutes. Throughout the reaction, the mixture gradually became thicker. After 30 minutes, water was slowly added to the paste with stirring. The suspension was diluted with warm water and treated with hydrogen peroxide to reduce residual permanganate and obtain graphite oxide. Subsequently, the dry graphite oxide was subjected to ultrasonic treatment in water for 2 hours, followed by centrifugation at 4500 rpm for one hour, forming graphene oxide [47].

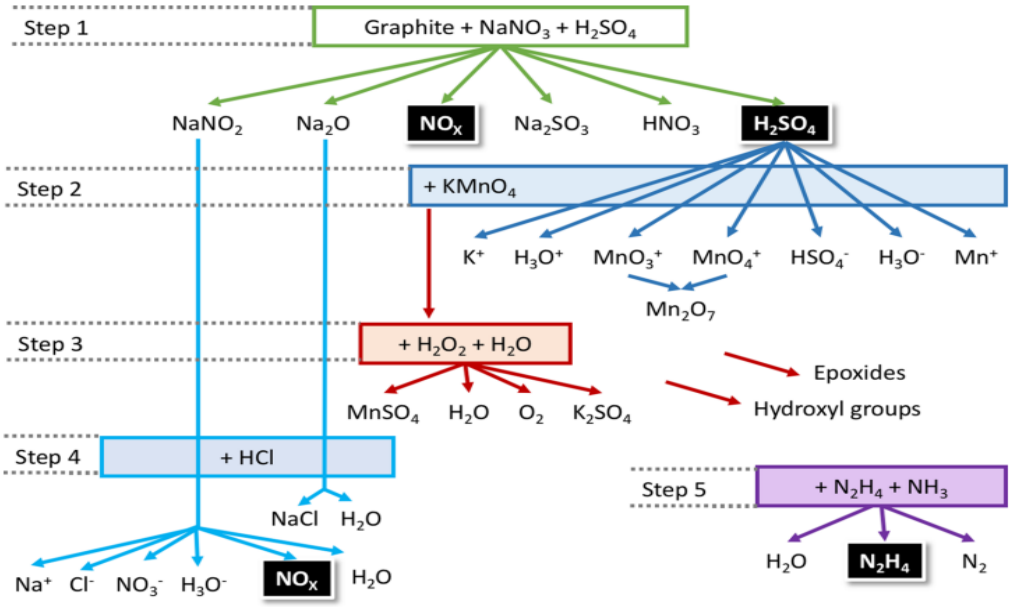

Figure 26: Reactions during Reduced Graphene Oxide Production (Hummers Method) [14]

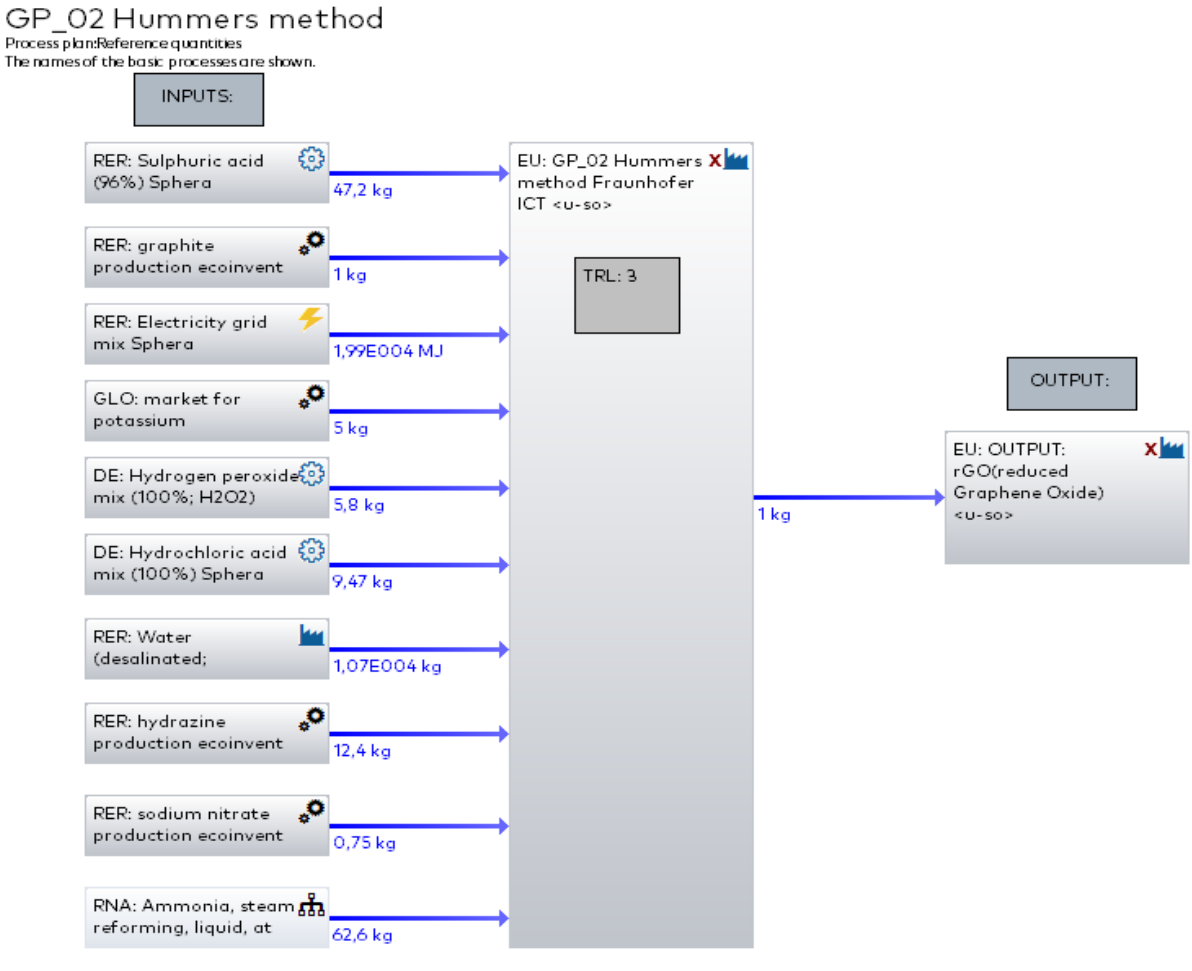

Figure 27: Schematic model of Hummers method [44, 45]

The synthesis of rGO involved adding an excess of hydrazine monohydrate, a reducing agent, to a dispersion of GO and refluxing the mixture for one hour. The concentration of hydrazine used was 6 µL per 1 mL of GO dispersion. The solution was filtered and the material obtained was washed with deionized water. The washed material was then vacuum dried at 80° C overnight, resulting in a powder-like rGO called hGO [16]. Required Input and output for the production of reduced graphene oxide are described in Appendix A.6.

4.7 Marcano's method

The Hummers method, a commonly employed GO synthesis technique, utilizes a concentrated sulfuric acid solution containing potassium permanganate and sodium nitrate. This mixture facilitates the efficient oxidation of graphite, leading to GO production within a relatively short timeframe. Numerous subsequent studies have proposed modifications to the Hummers method, aiming to enhance the efficiency of the oxidation process. The predominant GO synthesis methods often involve increasing potassium permanganate concentrations, enabling the facile production of substantial GO quantities. Notably, a variation eliminates sodium nitrate ($NaNO_3$) by increasing the potassium permanganate quantity and conducting the reaction in a 9:1 ratio of sulfuric acid (H_2SO_4) and phosphoric acid (H_3PO_4). This modification of the Hummers' method, known as Marcano's process, significantly enhances the efficiency of the oxidation process [16].

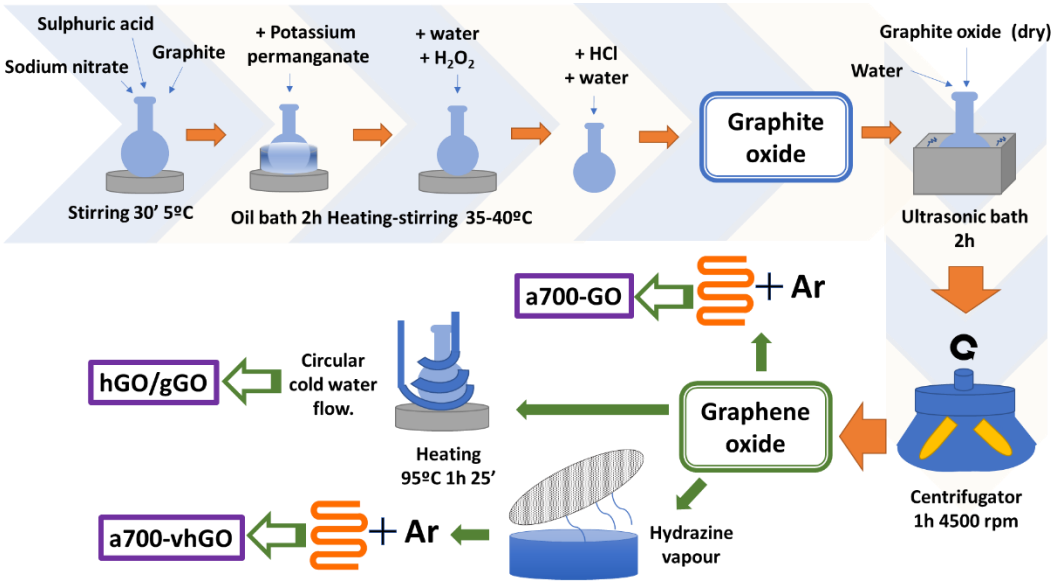

Figure 28: Cradle-to-Gate (LCA) for Modified Marcano Recipe of Reduced Graphene Oxide Production [14]

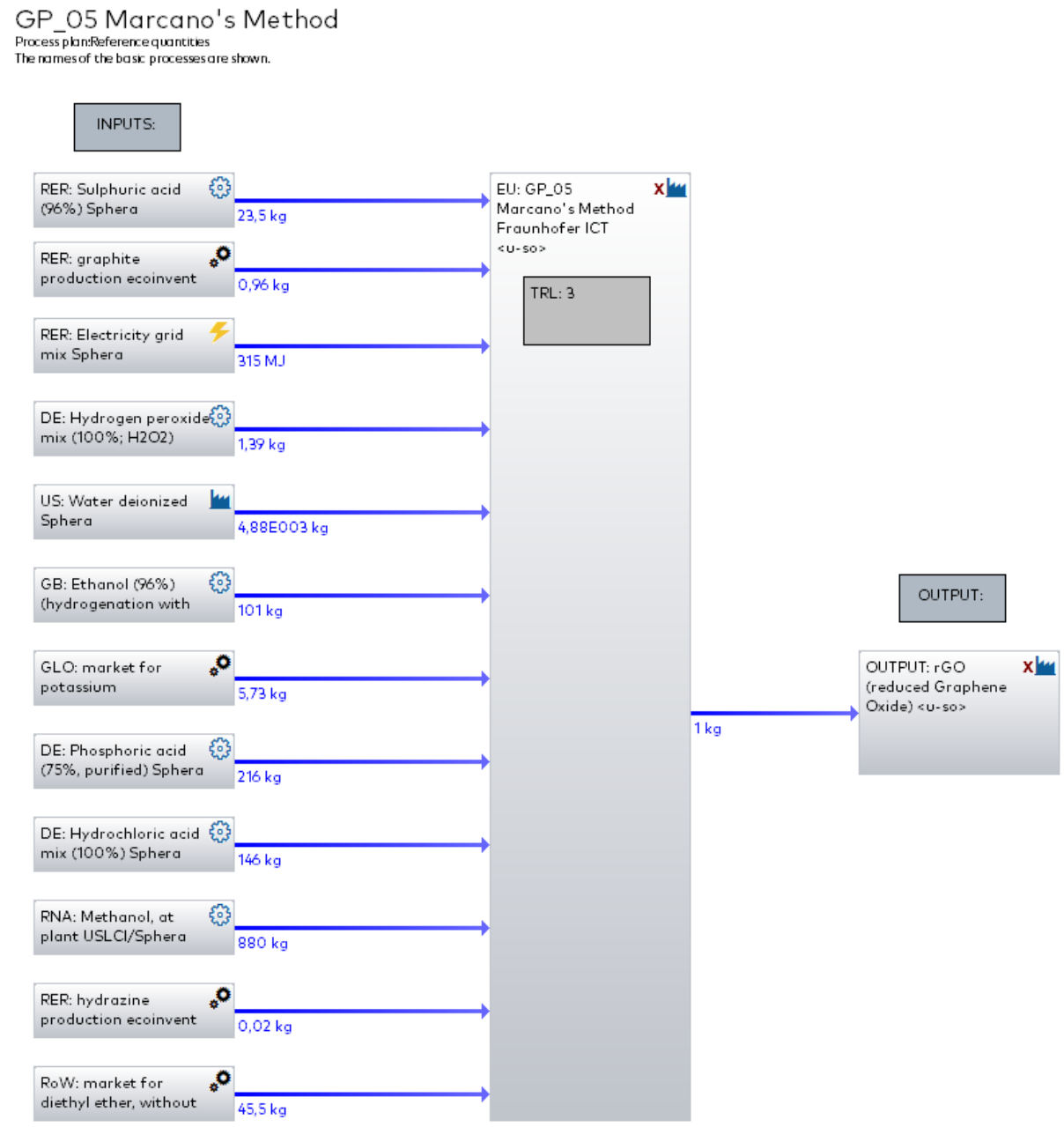

Figure 29: Schematic model of Marcano's method [42, 43]

Graphite oxide was produced through a modified approach based on the Hummers method. In this process, graphite powder was oxidized using a combination of $NaNO_3$, H_2SO_4, and $KMnO_4$ within an ice bath [47]. Initially, concentrated H_2SO_4 was combined with graphite flakes and $NaNO_3$, with the mixture cooled in an ice bath and stirred for 30 minutes. Subsequently, $KMnO_4$ was gradually added and stirred for a further 30 minutes. The reaction temperature was then raised to 35°C and stirred for two additional hours. Water and H_2O_2 were slowly incorporated, stirring for an hour. After filtering the mixture, the resultant powder was repeatedly cleaned with HCL: H_2O (1:10) and dried for 24 hours in the open air [43]. Graphene oxide was produced through mild bath sonication of aqueous graphite oxide dispersion for 2 hours, followed by centrifugation at 4500 rpm for 60 minutes. This process resulted in a brown-colored

dispersion of exfoliated graphene oxide with a specified final concentration and then with the help of methanol, which is required to reduce graphene oxide to get a final product as reduced graphene oxide [16]. All the Input and output inventories are mentioned in Appendix A.7.

4.8 Chemical oxidation of graphite

The chemical processes explored in this work comprise two primary phases. Initially, graphite undergoes oxidation, in which oxygen ions or groups intercalate the graphitic structure, causing its expansion. Subsequently, the oxidized graphite is reduced, wherein the oxygen groups are eliminated, resulting in a porous structure containing randomly arranged graphene nanoplatelets commonly referred to as rGO. Oxidation is accomplished by introducing an oxidative agent into an acidic solution containing graphite powder. The procedure is cooled to regulate the temperature and prevent excessive reactions triggered by the addition of the oxidative agent [8].

The study evaluated a range of graphite oxidation methods, which are denoted as GO1 to GO5.

- GO1 – Modified Hummers method (Fugetsu variant) [48]
- GO2 – Modified Hummers method (Bangal variant) [49]
- GO3 – Modified Hummers method (Jeong variant) [50]
- GO4 – Staundenmaier method [51]
- GO5 – Brodie method [52]

The modified Hummers variations, including Fugetsu, Bangal, and Jeong, involve mixing graphite powder with sodium nitrate ($NaNO_3$) and sulfuric acid (H_2SO_4), employing potassium permanganate ($KMnO_4$) as the oxidative agent. Each variant carefully regulates temperature to meet specific process conditions (temperature, duration). The Bangal variant has the shortest process duration (30 minutes at 35 °C), whereas the Fugetsu and Jeong variants proceed for 3 hours at temperatures slightly below 100 °C. Additionally, the Jeong variant utilizes ten times more sulfuric acid than the others. In contrast, the Staundenmaier process stands out by combining sulfuric acid with fuming nitric acid (HNO_3), employing potassium chlorate ($KClO_3$) as the oxidative agent, with the reaction occurring at ambient temperature for 96 hours.

On the other hand, the Brodie method solely utilizes fuming nitric acid, with sodium chlorate ($NaClO_3$) as the oxidative agent, proceeding at ambient temperature for 24 hours [8].

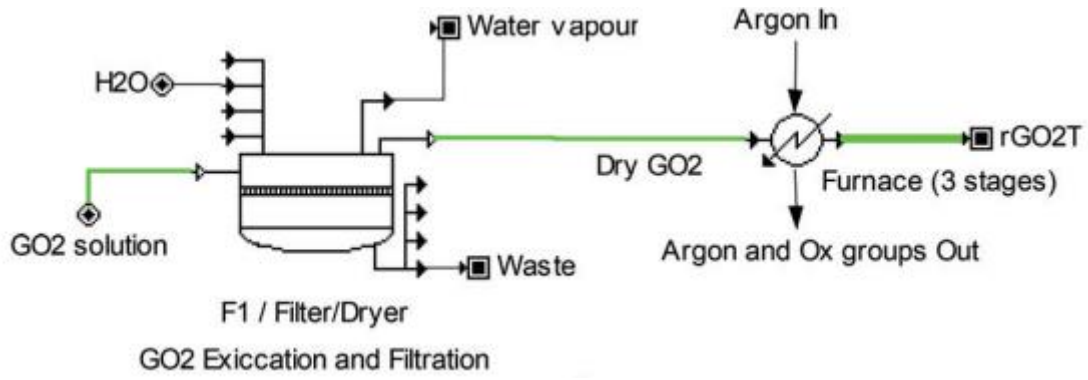

Figure 30: Thermal reduction of graphene oxide [6]

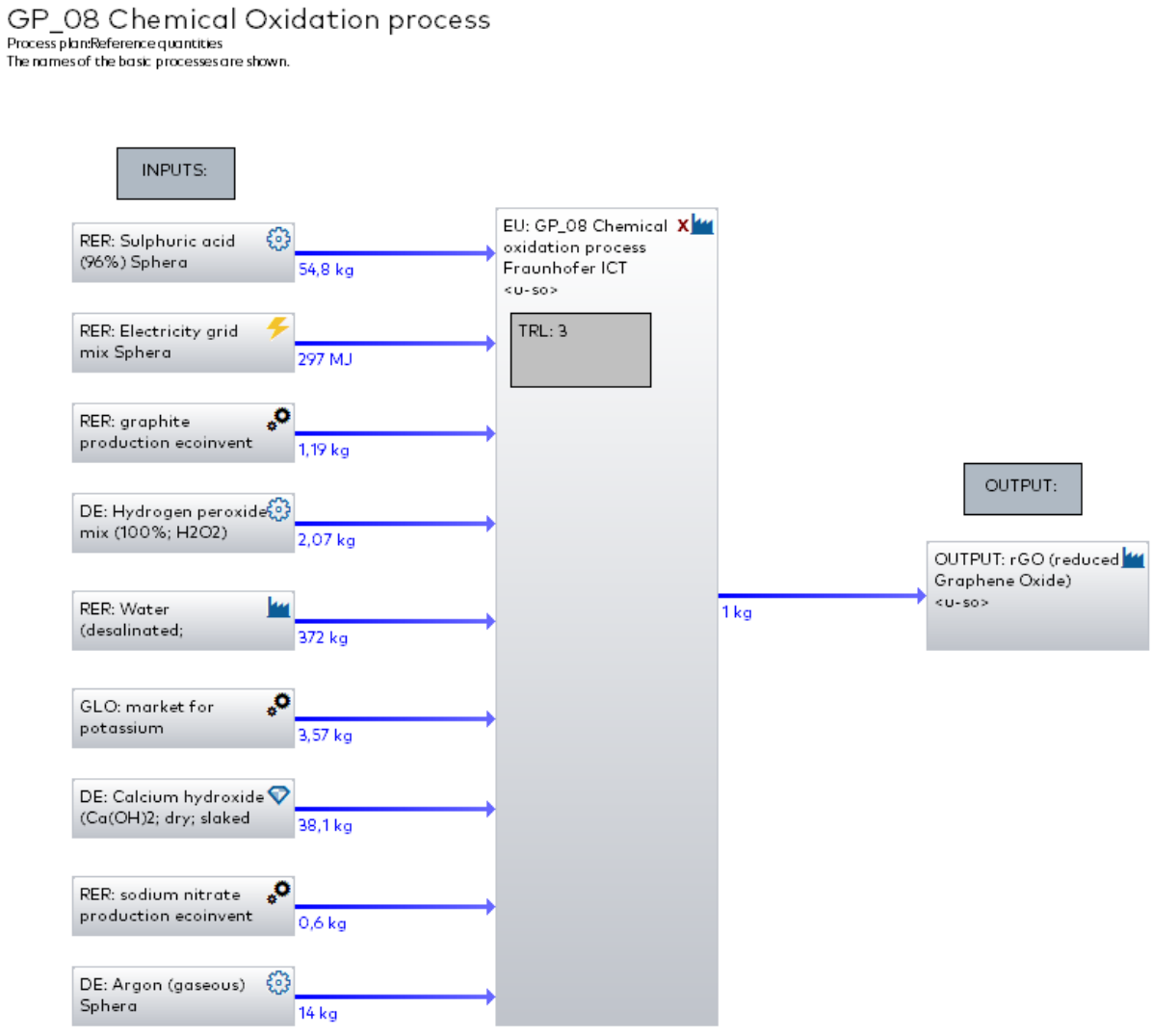

Figure 31: Schematic model of Chemical oxidation process [42, 43]

Graphene oxide reduction can be accomplished through thermal methods. For thermal reduction, a vacuum furnace is employed, in which the graphene oxide undergoes heating to 700°C over approximately 20 hours, with a precisely controlled heating rate. The sample was held at 140°C for 30 minutes by initially keeping the heating rate at 1.5°C per minute. Subsequently, the heating rate decreases to 0.3°C per minute, reaching 350°C, and then the sample is held at this temperature for 30 minutes. In the final stage, the heating rate returns to 1.5°C per minute, elevating the temperature from 350°C to 700°C, with the sample maintained at 700°C for 2 hours before the furnace is turned off, allowing the sample to cool naturally. Before reduction, the graphene oxide must be dried, initiating the thermal annealing process, with water evaporation occurring over 24 hours at 55°C within a furnace [8].

For the analysis, the study included the GO2 method for producing graphite oxide because it uses less energy than other methods. The path after oxidation was thermal reduction, as per the figure, which is the best possible method for having the least impact on the Environmental Footprint [8]. All the input and output data are described in Appendix A.8.

5. Results and Discussion

5.1 Environmental footprint and single score

Table 6 and 7 describes the single Environmental Footprint score for each distinct production process, which is derived through normalization and weighting factors, as explained previously. This is help to understand the PEF single score of different environmental impacts encountered throughout a product's life cycle into a singular numerical metric, facilitating straightforward comparisons among diverse products. The table below presents 25 indicators that have been normalized into 16 impact indicators. Within the Climate Change category, there are three specific indicators: Climate Change, biogenic; Climate Change, fossil; and Climate Change, land use and land use change. Ecotoxicity, freshwater comprises both Ecotoxicity, freshwater inorganics and Ecotoxicity, freshwater organics. Similarly, Human toxicity, cancer combines Human toxicity, cancer inorganics and Human toxicity, cancer organics. The category Human toxicity, non-cancer encompasses the total of Human toxicity, non-cancer inorganics and Human toxicity, non-cancer organics.

Technologies	*Acidification [Mole of H+ eq.]*	*Climate Change [kg CO2 eq.]*	*Ecotoxicity, freshwater [CTUe]*	*Particulate matter [Disease incidences]*	*Eutrophication, freshwater [kg P eq.]*	*Eutrophication, marine [kg N eq.]*
	absolut	*absolut*	*absolut*	*absolut*	*absolut*	*absolut*
Tech01- Chemical reduction	2.11E-01	9.95E+01	5.76E+02	1.77E-06	3.68E-04	5.04E-02
Tech02- Electrochemical exfoliation	3.54E-01	1.67E+02	9.69E+02	2.98E-06	6.18E-04	8.47E-02
Tech03- Ultrasonic Exfoliation	5.44E-02	2.57E+01	1.49E+02	4.58E-07	9.49E-05	1.30E-02
Tech04- Hydrothermal method	4.57E-01	2.16E+02	1.25E+03	3.84E-06	7.98E-04	1.09E-01
Tech05- Annealing method	9.62E-02	4.54E+01	2.63E+02	8.10E-07	1.68E-04	2.30E-02
Tech06- Hummers method	3.81E+00	1.80E+03	1.04E+04	3.20E-05	6.65E-03	9.10E-01
Tech07- Marcano's method	6.03E-02	2.85E+01	1.65E+02	5.08E-07	1.05E-04	1.44E-02
Tech08- Chemical oxidation	5.68E-02	2.68E+01	1.55E+02	4.78E-07	9.92E-05	1.36E-02
Total	***5.10E+00***	***2.41E+03***	***1.39E+04***	***4.28E-05***	***8.90E-03***	***1.22E+00***

Technologies	*Eutrophication, terrestrial [Mole of N eq.]*	*Human toxicity, cancer [CTUh]*	*Human toxicity, non-cancer [CTUh]*	*Ionising radiation [kBq U235 eq.]*	*Land Use [Pt]*	*Ozone depletion [kg CFC-11 eq.]*
	absolut	*absolut*	*absolut*	*absolut*	*absolut*	*absolut*
Tech01- Chemical reduction	5.26E-01	3.05E-08	4.87E-07	5.49E+01	8.14E+02	1.82E-09
Tech02- Electrochemical exfoliation	8.85E-01	5.13E-08	8.18E-07	9.23E+01	1.37E+03	3.06E-09
Tech03- Ultrasonic Exfoliation	1.36E-01	7.88E-09	1.26E-07	1.42E+01	2.10E+02	4.70E-10
Tech04- Hydrothermal method	1.14E+00	6.62E-08	1.06E-06	1.19E+02	1.77E+03	3.95E-09
Tech05- Annealing method	2.40E-01	1.39E-08	2.22E-07	2.51E+01	3.72E+02	8.31E-10
Tech06- Hummers method	9.51E+00	5.52E-07	8.79E-06	9.92E+02	1.47E+04	3.29E-08
Tech07- Marcano's method	1.51E-01	8.74E-09	1.39E-07	1.57E+01	2.33E+02	5.21E-10
Tech08- Chemical oxidation	1.42E-01	8.23E-09	1.31E-07	1.48E+01	2.20E+02	4.91E-10
Total	***1.27E+01***	***7.39E-07***	***1.18E-05***	***1.33E+03***	***1.97E+04***	***4.40E-08***

Technologies	*Photochemical ozone formation [kg NMVOC eq.]*	*Resource use, fossils [MJ]*	*Resource use, mineral and metals [kg Sb eq.]*	*Water use [m³ world equiv.]*	**Environmental Footprint [Single score]**
	absolut	*absolut*	*absolut*	*absolut*	**absolut**
Tech01- Chemical reduction	1.34E-01	2.07E+03	1.52E-05	2.20E+01	7.49E-03
Tech02- Electrochemical exfoliation	2.26E-01	3.49E+03	2.56E-05	3.69E+01	1.26E-02
Tech03- Ultrasonic Exfoliation	3.47E-02	5.36E+02	3.94E-06	5.67E+00	1.94E-03
Tech04- Hydrothermal method	2.91E-01	4.50E+03	3.31E-05	4.76E+01	1.63E-02
Tech05- Annealing method	6.14E-02	9.48E+02	6.97E-06	1.00E+01	3.42E-03
Tech06- Hummers method	2.43E+00	3.75E+04	2.76E-04	3.97E+02	1.36E-01
Tech07- Marcano's method	3.85E-02	5.94E+02	4.37E-06	6.29E+00	2.15E-03
Tech08- Chemical oxidation	3.62E-02	5.60E+02	4.11E-06	5.92E+00	2.02E-03
Total	***3.25E+00***	***5.02E+04***	***3.69E-04***	***5.31E+02***	**1.81E-01**

Table 6: Environmental Footprint [single score] for graphene production

FU (1 kg)	Technologies	Environmental Footprint [Single score]
Graphene	Tech01- Chemical reduction	7.49E-03
Graphene	Tech02- Electrochemical exfoliation	1.26E-02
Graphene	Tech03- Ultrasonic Exfoliation	1.94E-03
N-rGO	Tech04- Hydrothermal method	1.63E-02
N-rGO	Tech05- Annealing method	3.42E-03
rGO	Tech06- Hummers method	1.36E-01
rGO	Tech07- Marcano's method	2.15E-03
rGO	Tech08- Chemical oxidation	2.02E-03

Table 7: Results of the EF single score

From the table above, the Chemical reduction process with the 7.49E-03 EF single score, this method involves using a reducing agent like hydrazine to reduce the graphite oxide to graphene and also consumes a significant amount of energy to produce graphene, often resulting in a considerable amount of environmental impact compared to other methods. Electrochemical exfoliation with a 1.26E-02 absolute single score utilizes electrical energy to exfoliate material and consumes energy higher than Chemical reduction, resulting in a slightly higher environmental impact than chemical reduction. Ultrasonic exfoliation with 1.94E-03 uses thermal expansion to separate electrodes, showing a relatively lower absolute value. The hydrothermal method with a single score value of 1.63E-02 involves using high-pressure and high-temperature water to synthesize N-rGO, resulting in a higher single score value than other production methods except the Hummers method. With an absolute single score of 3.42E-03, the annealing method involves heating a material to a specific temperature and then cooling it slowly, resulting in a moderate environmental impact. Hummers method, with the highest value of EF single score 1.36E-01, states that this method impacts the most on the Environmental Footprint with the highest energy consumption to produce rGO, most of the energy required in sonication and centrifugation stage, and that it directly impacts the single score of PEF. Marcano's and chemical oxidation and thermal reduction methods show similar Environmental Footprint single scores, lower than other production methods discussed earlier. Marcano's method scores 2.15E-03, and chemical oxidation scores 2.02E-03, indicating relatively lower environmental impacts. Marcano's method involves a variation of chemical reduction to produce reduced graphene oxide with minimal environmental impact. The chemical oxidation method utilizes potassium permanganate as an oxidizing agent to oxidize materials, resulting in a relatively low environmental impact.

5.2 Environmental Footprints of 1 kg of Graphene Production Methods:

Table 8 illustrates the environmental impact of producing 1 kilogram of graphene through chemical reduction, electrochemical exfoliation, and ultrasonic exfoliation.

	1 kg of Graphene		
Impact Indicators	**Chemical reduction**	**Electrochemical exfoliation**	**Ultrasonic exfoliation**
EF 3.1 Acidification [Mole of H+ eq.]	2.11E-01	3.54E-01	5.44E-02
EF 3.1 Climate Change - total [kg CO2 eq.]	9.95E+01	1.67E+02	2.57E+01
EF 3.1 Climate Change, biogenic [kg CO2 eq.]	8.71E-01	1.46E+00	2.25E-01
EF 3.1 Climate Change, fossil [kg CO2 eq.]	9.86E+01	1.66E+02	2.54E+01
EF 3.1 Climate Change, land use and land use change [kg CO2 eq.]	1.07E-02	1.80E-02	2.77E-03
EF 3.1 Ecotoxicity, freshwater - total [CTUe]	5.76E+02	9.69E+02	1.49E+02
EF 3.1 Ecotoxicity, freshwater inorganics [CTUe]	5.74E+02	9.66E+02	1.48E+02
EF 3.1 Ecotoxicity, freshwater organics [CTUe]	2.27E+00	3.82E+00	5.87E-01
EF 3.1 Eutrophication, freshwater [kg P eq.]	3.68E-04	6.18E-04	9.49E-05
EF 3.1 Eutrophication, marine [kg N eq.]	5.04E-02	8.47E-02	1.30E-02
EF 3.1 Eutrophication, terrestrial [Mole of N eq.]	5.26E-01	8.85E-01	1.36E-01
EF 3.1 Human toxicity, cancer - total [CTUh]	3.05E-08	5.13E-08	7.88E-09
EF 3.1 Human toxicity, cancer inorganics [CTUh]	4.91E-09	8.27E-09	1.27E-09
EF 3.1 Human toxicity, cancer organics [CTUh]	2.56E-08	4.31E-08	6.61E-09
EF 3.1 Human toxicity, non-cancer - total [CTUh]	4.87E-07	8.18E-07	1.26E-07
EF 3.1 Human toxicity, non-cancer inorganics [CTUh]	4.78E-07	8.03E-07	1.23E-07
EF 3.1 Human toxicity, non-cancer organics [CTUh]	8.86E-09	1.49E-08	2.29E-09
EF 3.1 Ionising radiation, human health [kBq U235 eq.]	5.49E+01	9.23E+01	1.42E+01
EF 3.1 Land Use [Pt]	8.14E+02	1.37E+03	2.10E+02
EF 3.1 Ozone depletion [kg CFC-11 eq.]	1.82E-09	3.06E-09	4.70E-10
EF 3.1 Particulate matter [Disease incidences]	1.77E-06	2.98E-06	4.58E-07
EF 3.1 Photochemical ozone formation, human health [kg NMVOC eq.]	1.34E-01	2.26E-01	3.47E-02
EF 3.1 Resource use, fossils [MJ]	2.07E+03	3.49E+03	5.36E+02
EF 3.1 Resource use, mineral and metals [kg Sb eq.]	1.52E-05	2.56E-05	3.94E-06
EF 3.1 Water use [m³ world equiv.]	2.20E+01	3.69E+01	5.67E+00

Table 8: EF3.1 for Graphene Production Processes for 1kg of graphene [8, 26, 39]

5.2.1 Chemical Reduction of graphite oxide

Graphene production through chemical reduction processes consumes a significant amount of energy, approximately 1100 MJ/kg, as stated in the literature [26]. The chemical reduction process is the primary factor responsible for high energy consumption in CRR, accounting for approximately 75% of the total. This is primarily attributed to the heat required during chemical reduction and the substantial energy demand of hydrazine production [26]. This high energy demand profoundly impacts Environmental Footprint indicators, contributing to elevated environmental burdens associated with graphene production.

According to the table, the chemical reduction of the graphite oxide process has the most significant impact on Resource use, fossils with 2.07E+03 MJ, and Land use with 8.14E+02 Pt, this are two most impact indicators which impact the Environmental Footprint. While, Climate Change – total with 9.95E+01 kg CO_2 eq., in which Climate Change, fossil impact the most

with 9.86E+01 kg CO_2 eq., and Ecotoxicity, freshwater – total with the impact of 5.76E+02 CTUe impact significantly on Environmental Footprint. The observation shows that phosphoric acid, hydrazine, and hydrogen peroxide are primary contributors to Climate Change – total and Land Use impact indicators, highlighting these chemical input's critical role in the chemical reduction process [26]. The main reason for high Climate Change, fossil and Resource use, fossils indicators is higher energy consumption during production [26].

According to the table, Ecotoxicity, freshwater - total, Ecotoxicity, freshwater inorganics, and significantly impact the Environmental Footprint. The chemical reduction process is again the leading cause of the Ecotoxicity, freshwater - total in the case of the CRR. Over 95% of the Ecotoxicity, freshwater inorganics impact indicator is attributed to the emission of vanadium during the hydrazine synthesis process [26].

5.2.2 Electrochemical exfoliation

Graphene production through electrochemical exfoliation consumes 1850 MJ/kg of energy to produce 1 kg of graphene. There are multiple hotspots behind this consumption. First, high voltage and current are required for the electrochemical reactions involved in the process, which inevitably raises the energy requirement. Second, a significant impact on energy usage may come from the kind of electrolyte used in the procedure. In this process KOH used as a electrolyte to produce graphene. Lastly, the properties and composition of the materials used for the electrodes also affect the amount of energy needed. These characteristics combined contribute to the higher energy consumption observed in the electrochemical exfoliation process used to produce graphene [8].

Table above indicates that in Electrochemical exfoliation, Resource use,fossils and Land use, have the most substantial impact on the Environmental Footprint, with an impact of 3.49E+03 MJ and 1.37E+03 Pt, respectively, exceeding that of the Chemical reduction process. This higher impact is primarily attributed to increased energy consumption, surpassing that of the chemical reduction process. The elevated energy consumption is largely derived from fossil fuel sources within the energy grid background data, thus contributing significantly to resource use, fossil impact, and land use within the Environmental Footprint. Ecotoxicity, freshwater - total with the impact magnitude 9.69E+02 CTUe, also significantly affects the Environmental Footprint. Firstly, electrochemical exfoliation involves using chemicals and electrolytes, potentially leading to higher Ecotoxicity due to the release of toxic substances into the water. Disposing and managing these chemicals can significantly impact freshwater resources,

contributing to the observed higher impact on Ecotoxicity, freshwater-total and freshwater inorganics [35]. Using potassium hydroxide as an input material could negatively affect the Ecotoxicity of freshwater and public health because of its acidic nature and ability to contaminate freshwater supplies if improperly handled and disposed of [8]. This data suggests that KOH consumes slightly more water, dissociating into hydrogen and oxygen [8].

5.2.3 Ultrasonic Exfoliation

During graphene production, ultrasonic exfoliation consumed 284 MJ/kg of electricity. According to the literature, the thermal expansion process and graphite extraction from petroleum coke account for a significant portion of the energy used in ultrasonic exfoliation [39].

Process/Material	Quantity (kg)	Embodied Energy	Data origin
Graphite	1	236.52 - 260.41	Literature
Pre-treatment	0.6	1.19	Literature
Microwave expansion	21.6	7.92-15.84	Literature
Ultrasonic exfoliation	21.6	5.83-11.66	Literature

Table 9: Energy requires in each phase to produce Graphene through Ultrasonic Exfoliation [39]

The table 9 illustrates that a substantial quantity of energy is needed for graphite production using the Ultrasonic method. Significant energy is consumed during pre-treatment, Microwave expansion, and Ultrasonic exfoliation processes to manufacture 1 kg of graphene.

The table above shows that Ultrasonic Exfoliation impact the least among other two production process. The major reason behind that is less energy consumption than other two method. For the Ultrasonic exfoliation, Resource use, fossils, and Land use are the primary impact indicators that impact the Environmental Footprint with the impact value of 5.36E+02 MJ and 2.10E+02 Pt. Ultrasonic exfoliation depends on inputs originating from fossil fuels, especially in the mining and processing of graphite, an essential precursor material. A vital energy input and contributor to greenhouse gas emissions is petroleum coke, the primary source of graphite manufacturing. Land is used during production for several reasons, such as waste management, equipment cooling, material preparation, and the extensive use of land, especially in the mining and processing of graphite [39]. Ultrasonic exfoliation depends mostly on mechanical and ultrasonic energy, which offers less danger to freshwater habitats than industrial procedures

using chemical treatments or harmful compounds. The lower impact on Ionizing radiation-human health in ultrasonic exfoliation indicates reduced risks to human health [39].

5.3 Environmental Footprints of 1 kg of N-rGO Production Methods:

Table 10 describes the environmental impact associated with producing 1 kilogram of nitrogen-doped reduced graphene oxide through the hydrothermal method and annealing method [2].

	1 kg of N-rGO	
Impact Indicators	**Hydrothermal method**	**Annealing method**
EF 3.1 Acidification [Mole of H+ eq.]	4.57E-01	9.62E-02
EF 3.1 Climate Change - total [kg CO2 eq.]	2.16E+02	4.54E+01
EF 3.1 Climate Change, biogenic [kg CO2 eq.]	1.89E+00	3.98E-01
EF 3.1 Climate Change, fossil [kg CO2 eq.]	2.14E+02	4.50E+01
EF 3.1 Climate Change, land use and land use change [kg CO2 eq.]	2.32E-02	4.90E-03
EF 3.1 Ecotoxicity, freshwater - total [CTUe]	1.25E+03	2.63E+02
EF 3.1 Ecotoxicity, freshwater inorganics [CTUe]	1.25E+03	2.62E+02
EF 3.1 Ecotoxicity, freshwater organics [CTUe]	4.93E+00	1.04E+00
EF 3.1 Eutrophication, freshwater [kg P eq.]	7.98E-04	1.68E-04
EF 3.1 Eutrophication, marine [kg N eq.]	1.09E-01	2.30E-02
EF 3.1 Eutrophication, terrestrial [Mole of N eq.]	1.14E+00	2.40E-01
EF 3.1 Human toxicity, cancer - total [CTUh]	6.62E-08	1.39E-08
EF 3.1 Human toxicity, cancer inorganics [CTUh]	1.07E-08	2.25E-09
EF 3.1 Human toxicity, cancer organics [CTUh]	5.55E-08	1.17E-08
EF 3.1 Human toxicity, non-cancer - total [CTUh]	1.06E-06	2.22E-07
EF 3.1 Human toxicity, non-cancer inorganics [CTUh]	1.04E-06	2.18E-07
EF 3.1 Human toxicity, non-cancer organics [CTUh]	1.92E-08	4.05E-09
EF 3.1 Ionising radiation, human health [kBq U235 eq.]	1.19E+02	2.51E+01
EF 3.1 Land Use [Pt]	1.77E+03	3.72E+02
EF 3.1 Ozone depletion [kg CFC-11 eq.]	3.95E-09	8.31E-10
EF 3.1 Particulate matter [Disease incidences]	3.84E-06	8.10E-07
EF 3.1 Photochemical ozone formation, human health [kg NMVOC eq.]	2.91E-01	6.14E-02
EF 3.1 Resource use, fossils [MJ]	4.50E+03	9.48E+02
EF 3.1 Resource use, mineral and metals [kg Sb eq.]	3.31E-05	6.97E-06
EF 3.1 Water use [m^3 world equiv.]	4.76E+01	1.00E+01

Table 10: EF3.1 for Graphene Production: Hydrothermal and Annealing Method [2]

5.3.1 Hydrothermal method

In the hydrothermal method, the energy needed to produce 1 kilogram of N-rGO is 2386.80 MJ/kg. This energy is utilized across various equipment such as hot plates, centrifuges, Teflon-lined autoclaves, ultrasound devices, and ovens for stirring, heating, centrifugalizing, and drying purposes. The most significant contributor to energy consumption is the heating process for the Teflon-lined autoclave, constituting approximately 93.7 % of the total energy usage. Additionally, energy consumption from ethanol production and the GO process also plays a role, accounting for approximately 18.3 % and 5.2 %, respectively [2].

The table highlights that in the hydrothermal method, Ecotoxicity, freshwater-total, Resource use, fossils, and Land use contribute significantly to the Environmental Footprint, with impact values of 1.25E+03 CTUe, 4.50E+03 MJ, and 1.77E+03 Pt, respectively. According to data from the ethanol background database, ethanol production is known for its high energy intensity, relying heavily on fossil energy for raw materials and fuels. The higher impact of Land use is attributed to its utilization across different production stages. Notably, electricity production contributes approximately 69.7 % to Ecotoxicity and freshwater-total impact. Additionally, the hydrothermal process, ultrapure water production, and ethanol production contribute 13.5 %, 9.3 %, and 7.3 %, respectively, to overall water usage. Ethanol production (contributing around 17.2%) and electricity (accounting for about 69.7 %) are the primary contributors to increased climate change-total impact, with a magnitude of 2.16E+02 kg CO_2 eq. These elements are critical due to their high energy demands and associated environmental effects. Electricity production, often derived from fossil fuels, emits pollutants that worsen environmental conditions. Similarly, the energy-intensive procedures and reliance on fossil fuels in ethanol production increase Ecotoxicity, freshwater-total impacts by releasing contaminants and chemical residues into water systems [2].

5.3.2 Annealing method

To produce 1kg of N-rGO through the Annealing method, 502.59 MJ/kg of electricity is required. The main contributor to this is ethanol production for stirring, heating, and evaporating ethanol, which contributes (approximately 61.6%); the next is the GO process, ammonium nitrate production, and ultrapure water production, which is in the range of (7.8 %, 4.1 %, and 1.5 %, respectively) [2].

The environmental analysis covers several significant impact indicators such as Ecotoxicity, freshwater – total with impact of 2.63E+02 CTUe, Land use 3.72E+02 Pt, Resource use-fossils 9.48E+02 MJ, which are the three most primary impact indicators affecting the Environmental Footprint for production through the annealing method. Climate change-total with the impact of 4.54E+01 kg CO2 eq. is also a critical concern in Life Cycle Assessment and the production of N-rGO, reflecting its widespread importance. They are considered in this study because N-rGO production uses much electricity and is energy intensive. Water consumption encompasses both foreground and background water usage during production [2].

In the annealing method, ethanol production emerges as the primary driver for elevated Climate change-total and fossil indicators, requirement of ethanol is double than Hydrothermal method.

For higher impact of Ecotoxicity, freshwater – total, Ethanol and graphene oxide (GO) production are the primary contributors to the environmental impact, accounting for 69.2 % and 14.93%, respectively. This is largely attributed to phenol emissions into water, which contribute approximately 52.7 % of the impact. The impacts from the production of other materials account for about 15.9 % collectively. Therefore, reducing ethanol and electricity consumption can effectively decrease the ecotoxicity impact associated with these processes. Consequently, enhancing the efficiency of ethanol recovery emerges as the most effective approach to mitigating the Environmental Footprint associated with N-rGO synthesis via the annealing method. Increasing the efficiency of ethanol recovery could significantly alleviate the environmental impacts across multiple indicators [2].

5.4 Environmental Footprints of 1 kg of rGO Production Methods:

Table 11 describes the environmental impact associated with producing 1 kilogram of reduced graphene oxide through the Hummers method, Marcano's method, and Chemical Oxidation method.

	1 kg of rGO		
Impact Indicators	**Hummers method**	**Marcano's method**	**Chemical Oxidation method**
EF 3.1 Acidification [Mole of H+ eq.]	3.81E+00	6.03E-02	5.68E-02
EF 3.1 Climate Change - total [kg CO2 eq.]	1.80E+03	2.85E+01	2.68E+01
EF 3.1 Climate Change, biogenic [kg CO2 eq.]	1.57E+01	2.49E-01	2.35E-01
EF 3.1 Climate Change, fossil [kg CO2 eq.]	1.78E+03	2.82E+01	2.66E+01
EF 3.1 Climate Change, land use and land use change [kg CO2 eq.]	1.94E-01	3.07E-03	2.89E-03
EF 3.1 Ecotoxicity, freshwater - total [CTUe]	1.04E+04	1.65E+02	1.55E+02
EF 3.1 Ecotoxicity, freshwater inorganics [CTUe]	1.04E+04	1.64E+02	1.55E+02
EF 3.1 Ecotoxicity, freshwater organics [CTUe]	4.11E+01	6.51E-01	6.13E-01
EF 3.1 Eutrophication, freshwater [kg P eq.]	6.65E-03	1.05E-04	9.92E-05
EF 3.1 Eutrophication, marine [kg N eq.]	9.10E-01	1.44E-02	1.36E-02
EF 3.1 Eutrophication, terrestrial [Mole of N eq.]	9.51E+00	1.51E-01	1.42E-01
EF 3.1 Human toxicity, cancer - total [CTUh]	5.52E-07	8.74E-09	8.23E-09
EF 3.1 Human toxicity, cancer inorganics [CTUh]	8.88E-08	1.41E-09	1.33E-09
EF 3.1 Human toxicity, cancer organics [CTUh]	4.63E-07	7.33E-09	6.91E-09
EF 3.1 Human toxicity, non-cancer - total [CTUh]	8.79E-06	1.39E-07	1.31E-07
EF 3.1 Human toxicity, non-cancer inorganics [CTUh]	8.63E-06	1.37E-07	1.29E-07
EF 3.1 Human toxicity, non-cancer organics [CTUh]	1.60E-07	2.54E-09	2.39E-09
EF 3.1 Ionising radiation, human health [kBq U235 eq.]	9.92E+02	1.57E+01	1.48E+01
EF 3.1 Land Use [Pt]	1.47E+04	2.33E+02	2.20E+02
EF 3.1 Ozone depletion [kg CFC-11 eq.]	3.29E-08	5.21E-10	4.91E-10
EF 3.1 Particulate matter [Disease incidences]	3.20E-05	5.08E-07	4.78E-07
EF 3.1 Photochemical ozone formation, human health [kg NMVOC eq.]	2.43E+00	3.85E-02	3.62E-02
EF 3.1 Resource use, fossils [MJ]	3.75E+04	5.94E+02	5.60E+02
EF 3.1 Resource use, mineral and metals [kg Sb eq.]	2.76E-04	4.37E-06	4.11E-06
EF 3.1 Water use [m³ world equiv.]	3.97E+02	6.29E+00	5.92E+00

Table 11: EF3.1 for Graphene Production: Hummers, Marcano's, Chemical Oxidation method [8, 16]

5.4.1 Hummers method

During the production of rGO using the Hummers method, 19881.39 MJ/kg of electricity was consumed, which is the highest amount of energy consumption among all the production

process. This high electricity consumption significantly impacted the Environmental Footprint, as highlighted by the impact assessment results. The primary hotspot where electricity consumption was exceptionally high was during the conversion of graphitic oxide to graphene oxide through sonication and centrifugation processes, followed by the energy-intensive heating process required to produce reduced graphene oxide from graphene oxide [16].

Table above shows that that Resource use, fossils, Ecotoxicity, freshwater - total, Climate change – total, and Land use with impact value of 3.75E+04 MJ, 1.04E+04 CTUe, 1.80E+03 kg CO_2 eq., and 1.47E+04 Pt were the highest among all indicators for the Hummers method. During the oxidation process of graphite following the Hummers method, various chemicals could be produced, including $NaNO_2$, Na_2O, NO_x, $NaSO_3$, HNO_3, H_2SO_4, K^+, $H3O^+$, MnO_3^+, HSO_4^-, Mn_2O_7, H_2O, Mn^{+2}. When adding the H_2O_2 and H_2O, the list grows with potential $MnSO_4$, H_2O, and K_2SO_4 compounds. In the worst-case scenario, H_2SO_4 will react with all $NaNO_3$ to produce NO_x emissions, which is the major contributor for Ecotoxicity, freshwater – total impact indicator. while the remaining H_2SO_4 will react with $KMnO_4$. Any remaining H_2SO_4 is considered potential emissions. During the oxidation of graphite oxide, the reaction between $NaNO_3$ and H_2SO_4 could produce also HNO_3. Furthermore, when hydrochloric acid is poured, it could lead to the formation of NaCl and Cl_2 or Cl^-. From all these substances, the main concern during the production of reduced graphene oxide is the emission of hydrazine and NO_x [16].

Hydrazine poses the most significant concern among the chemicals considered, significantly impacting most Climate Change - total. Following closely are potential NO_x emissions, which contribute to photochemical ozone formation, acidification, terrestrial eutrophication, and marine eutrophication. Hummers production involved land required for the graphene oxide and energy production. Hydrazine and sulfuric acid are the most significant contributors to freshwater ecotoxicity. The NO_x emissions impact the remaining categories [16].

5.4.2 Marcano’s method

In producing reduced graphene oxide via Marcano's method, approximately 315.03 MJ/kg of energy is utilized. The majority of this energy is consumed during the reduction of methanol. Additionally, significant amounts of electricity are also used for heating, stirring, centrifugation, and vacuum drying throughout the production process [16].

Resource use, fossils and Ecotoxicity, freshwater - total, are the most impact indicators that affect the Environmental Footprint of the production process with the impact amount of

5.60E+02 MJ and 1.55E+02 CTUe. Most of the water used during the production process is for the reduction of methanol for reduced graphene oxide in stage 3 of the production process, that impact directly on freshwater ecosystem which is releasing as a emission to water and also puts more impact on the Environmental Footprint. Moreover, the high impact of Resource use-fossils can be attributed to the background data of the electricity grid. An essential factor in the total environmental effect is the reliance on fossil fuels to produce electricity. Besides this, phosphoric acid is the major contributor, affecting all the impact indicator categories except for freshwater ecotoxicity. The modified recipe based on the Marcano's method is demonstrated to be the most efficient option from an environmental perspective, thereby justifying the adoption of an alternative to Hummers recipe, such as the Marcano's method [16].

5.4.3 Chemical Oxidation Method

During the production of reduced graphene oxide using the chemical oxidation method, the energy consumption amounted to 296.70 MJ/kg. Electricity was primarily dependent on the temperature and time required for the oxidation process; a secondary area of high energy usage was in the thermal reduction process, which involved heating the graphene oxide in a vacuum furnace to 700°C for around 20 hours with a specific heating rate and after that energy was required for mixing, filtering, and drying activities were also contributed a significant amount of contribution for energy consumption [8].

The table data highlights that Ecotoxicity, freshwater-total, and Resource use-fossils are notable impact indicators for the chemical oxidation process, with respective impact values of 5.60E+02 MJ and 1.55E+02 CTUe. Sulfuric acid primarily drives the higher Climate change-total impact with the 2.68E+01 kg CO_2 eq. impact of magnitude. The laboratory-scale findings depict a worst-case scenario, where 100% of the acid is discharged as waste in freshwater ecosystem and necessitates neutralization. This elevated result underscores the significance of recovering and reusing acid whenever feasible to minimize the environmental impact. In addition to sulfuric acid, hydrogen peroxide, and potassium permanganate also play significant roles in contributing to Climate change-total impact indicators. The production and utilization of these chemicals entail energy-intensive processes, releasing greenhouse gases into the atmosphere. Moreover, their chemical reactions during the graphene oxide production process may generate by-products that contribute to global warming [8].

Apart from electricity, which uses fossils as a primary input for generating power, two more input materials contribute equally to higher resource use: $Ca(OH)_2$ (Calcium hydroxide) and

$NaNO_3$ (sodium nitrate). These materials are extensively used in various stages of the production process, requiring significant energy inputs for their extraction and processing. Calcium hydroxide, commonly known as lime, is produced through the calcination of limestone, a process that typically involves the combustion of fossil fuels. Similarly, sodium nitrate is derived from natural deposits or manufactured through energy-intensive processes, often involving fossil fuels [8].

5.5 Discussion

The Product Environmental Footprint results from the table provide valuable insights into the environmental impacts of various graphene production methods. Among the methods analyzed, the Hummers method stands out with the highest EF single score of 1.36E-01, indicating a significant environmental impact due to its high energy consumption during stages like sonication and centrifugation. In contrast, methods such as Marcano's method with the value of 2.15E-03 and chemical oxidation with 2.02E-03 exhibit relatively lower environmental impacts, attributed to their less energy-intensive processes and lower emissions. The Annealing method, with a single score of 3.42E-03, falls in between, reflecting a moderate environmental impact driven by energy consumption during the heating and cooling phases. Notably, the Hydrothermal method with a 1.63E-02 single score shows a higher environmental impact, likely due to the intense energy and resource requirements of using high-pressure, high-temperature water for synthesis.

Additionally, Chemical Reduction had an absolute score of 7.49E-03, Electrochemical Exfoliation had 1.26E-02, and Ultrasonic Exfoliation had 1.94E-03. Chemical Reduction involves using hydrazine as a reducing agent to convert graphite oxide to graphene, resulting in a moderate environmental impact driven by significant energy consumption. Electrochemical exfoliation, utilizing electrical energy for material exfoliation, shows a slightly higher environmental impact than chemical reduction and is second most contributing after the Hummers method, primarily due to increased energy requirements. In contrast, Ultrasonic Exfoliation, which relies on thermal expansion for electrode separation, demonstrates a relatively lower environmental impact among the methods analyzed. The PEF single score clearly indicates that the Hummers method has the most adverse environmental impact compared to other graphene production methods, primarily due to its significantly higher energy consumption.

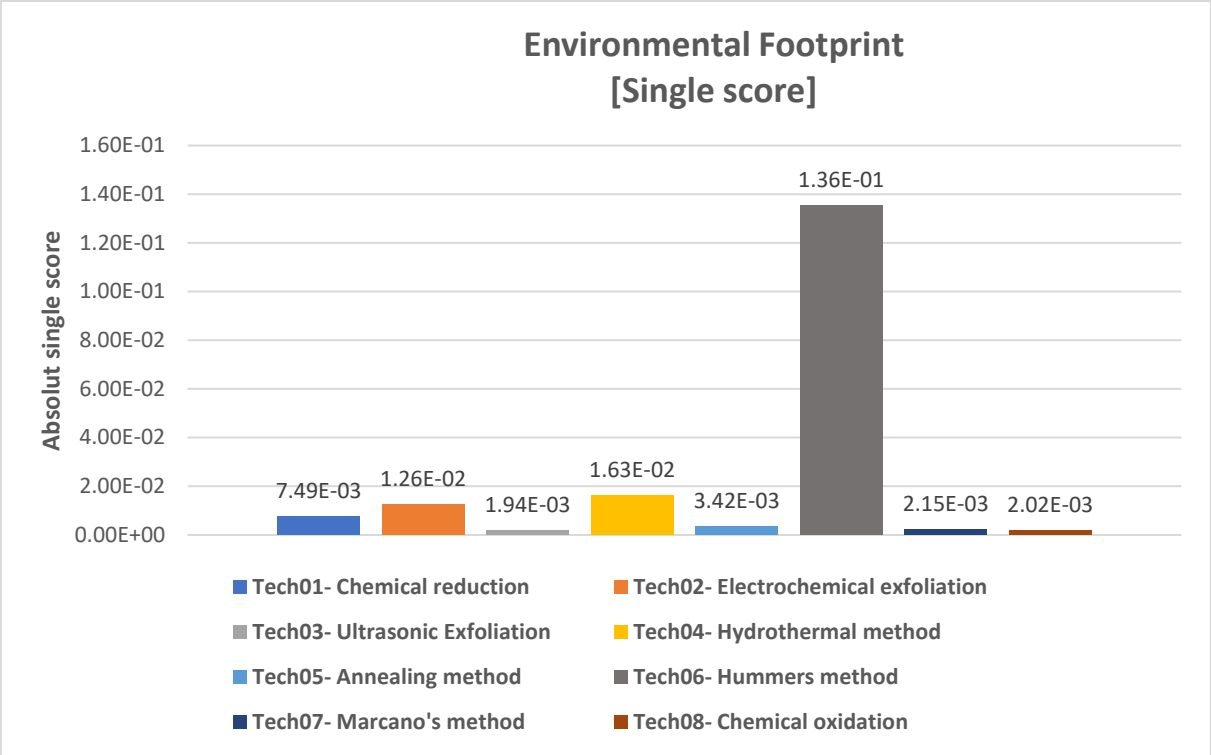

Figure 32: Environmental Footprint [Single Score]

Overall, these findings of PEF underscore the importance of considering energy consumption and process sustainability when choosing graphene production methods to minimize Environmental Footprints. Efforts to adopt more efficient and eco-friendly techniques, as demonstrated by Marcano's method and chemical oxidation, can lead to significant reductions in environmental impacts associated with graphene production.

The below figure compares embodied energies per unit mass of materials, highlighting the varying energy requirements for graphene production across different methods. The Hummers method, known for its high energy consumption, exhibits exceptionally high embodied energy values of 19881.39 MJ/kg. In contrast, ultrasonic and electrochemical exfoliation methods demonstrate relatively lower energy demands, ranging from 284 MJ/kg to 1850 MJ/kg, as detailed in Appendix A.9. Understanding these disparities in energy consumption is crucial for identifying and adopting more efficient production pathways that can minimize Environmental Footprints.

Aluminium, magnesium, and titanium light alloys have high embodied energy values; titanium is nearly 800 MJ/kg. Precious metals have even higher values. Polymers typically cluster around 100 MJ/kg, which is less than light alloys but considerably more than steels and cast irons, which range between 20 and 40 MJ/kg. Technical ceramics like aluminium nitride also have high embodied energies, whereas materials such as glass, cement, brick, and concrete have

much lower values. Composites exhibit a wide range of values, with high-performance composites like carbon-fibre-reinforced polymers (CFRP) at the top, well above most metals. On the other end of the spectrum, materials like paper, plywood, and timber are comparable to other construction industry materials [38].

The figure illustrates that reduced graphene oxide (rGO) produced through the Hummers method, nitrogen-doped reduced graphene oxide (N-rGO) via the Hydrothermal method, and graphene produced through chemical reduction and electrochemical exfoliation methods have the highest embodied energy compared to CFRP materials. While, Ultrasonication, Annealing method, Marcano's method, and Chemical oxidation have lower than CFRP. Graphene composites are a viable alternative to CFRP due to their comparable properties.

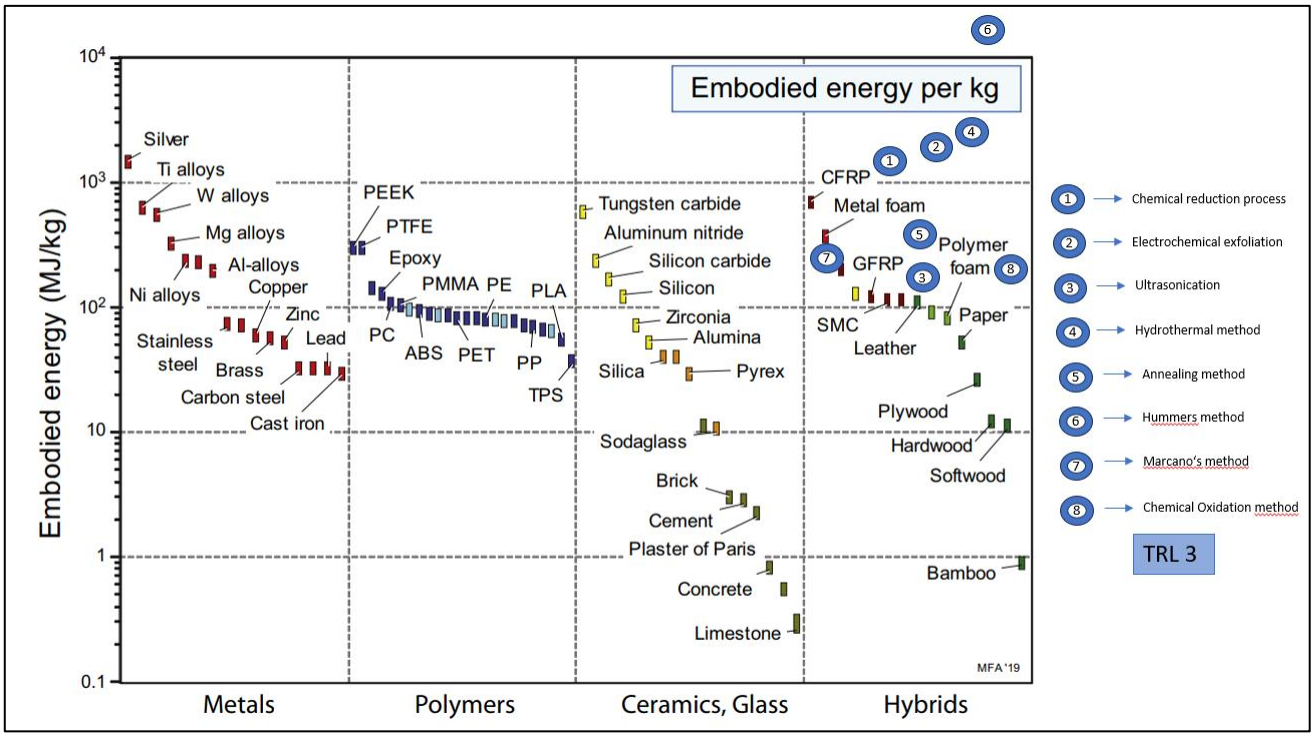

Figure 33: A bar chart of the embodied energies of materials per unit mass [49]

The environmental impact of eight production routes for Graphene, N-rGO, and rGO have been studied using life cycle assessment methodology, as highlighted in the tables, providing valuable insights into the key factors influencing the Environmental Footprint of these processes. Across various methods such as chemical reduction, electrochemical exfoliation, ultrasonic exfoliation, annealing, hydrothermal, Hummers, Marcano's and chemical oxidation methods, distinct impact indicators emerge as significant contributors to environmental concerns.

Starting the Chemical Reduction process exhibits substantial impacts on Resource use, fossils, and Land use, with notable values of 2.07E+03 MJ and 8.14E+02 Pt, respectively. This is primarily attributed to the high energy consumption during production, derived mainly from fossil fuel sources. Additionally, Climate Change - total and Ecotoxicity, freshwater - total are significantly influenced by inputs like phosphoric acid, hydrazine, and hydrogen peroxide, highlighting their critical role in driving environmental impacts. In contrast, Electrochemical Exfoliation demonstrates pronounced impacts on Resource use, fossils, and Land use, exceeding the impact observed in the Chemical Reduction process. This heightened impact is primarily attributed to increased energy consumption, largely derived from fossil fuel sources within the energy grid background data. The process also poses concerns for Ecotoxicity, freshwater - total due to chemical usage and subsequent disposal implications. Similarly, the Ultrasonic Exfoliation method presents relatively lower environmental impacts compared to other processes, mainly due to its reduced energy consumption. Resource use, fossils, and Land use are primary impact indicators, emphasizing the energy-intensive nature of graphite mining and processing in this method.

In the Hydrothermal method, Ecotoxicity, freshwater-total, Resource use, fossils, and Land use contribute significantly to the Environmental Footprint, underscoring the intensive resource requirements associated with high-pressure, high-temperature water synthesis. Additionally, the reliance on ethanol production, a process known for its high energy intensity and reliance on fossil fuels, further amplifies the Climate Change - total and Ecotoxicity, freshwater - total. The Annealing method has lower energy consumption compared to the hydrothermal method and consequently has a reduced impact on the Environmental Footprint compared to the hydrothermal method. This method shows notable impacts on Ecotoxicity, freshwater–total, Land use, and Resource use-fossils. Climate change-total impact is also a critical concern due to the high electricity consumption and energy intensity associated with N-rGO production with both methods.

The Hummers method demonstrated the highest environmental impact among the analyzed methods, particularly in Resource use, fossils, Ecotoxicity, freshwater-total, and Climate change-total, with substantial impact values. This method involves the oxidation of graphite using various chemicals like sulfuric acid, resulting in emissions of NO_x, hydrazine, and other pollutants. Hydrazine, in particular, poses a significant concern due to its contribution to Climate change - total. The production process requires considerable land and energy to produce reduced graphene oxide. Sulfuric acid and hydrazine are significant contributors to Ecotoxicity. freshwater - total. In contrast, Marcano's method showed a more moderate

environmental impact than the Hummers method. While Resource use, fossils, Ecotoxicity, freshwater - total remain significant impact indicators, the modified approach reduces the reliance on high-energy inputs like phosphoric acid, thus lowering the overall Environmental Footprint. The adoption of Marcano's method as an alternative to the Hummers method is justified by its improved environmental performance. The chemical oxidation and thermal reduction method also raised concerns, especially regarding Ecotoxicity, freshwater-total, and Resource use - fossils. Sulfuric acid emerged as a primary driver of Climate Change - total impact, necessitating careful management to minimize emissions and environmental repercussions. Additionally, using chemicals like hydrogen peroxide and potassium permanganate contributes to Ecotoxicity, freshwater - total, underscoring the toxicity-intensive nature of these chemical processes.

The figure below depicts the Environmental Footprint of an electricity grid for Functional unit 1 kWh, with background data identifying four key impact indicators that influence the environmental footprint of electricity consumption. From the analysis provided earlier, it is clear that electricity consumption represents a prominent hotspot that significantly contributes to an increased Environmental Footprint across different production methods. The underlying data from the electricity grid is instrumental in shaping this impact, notably influencing key

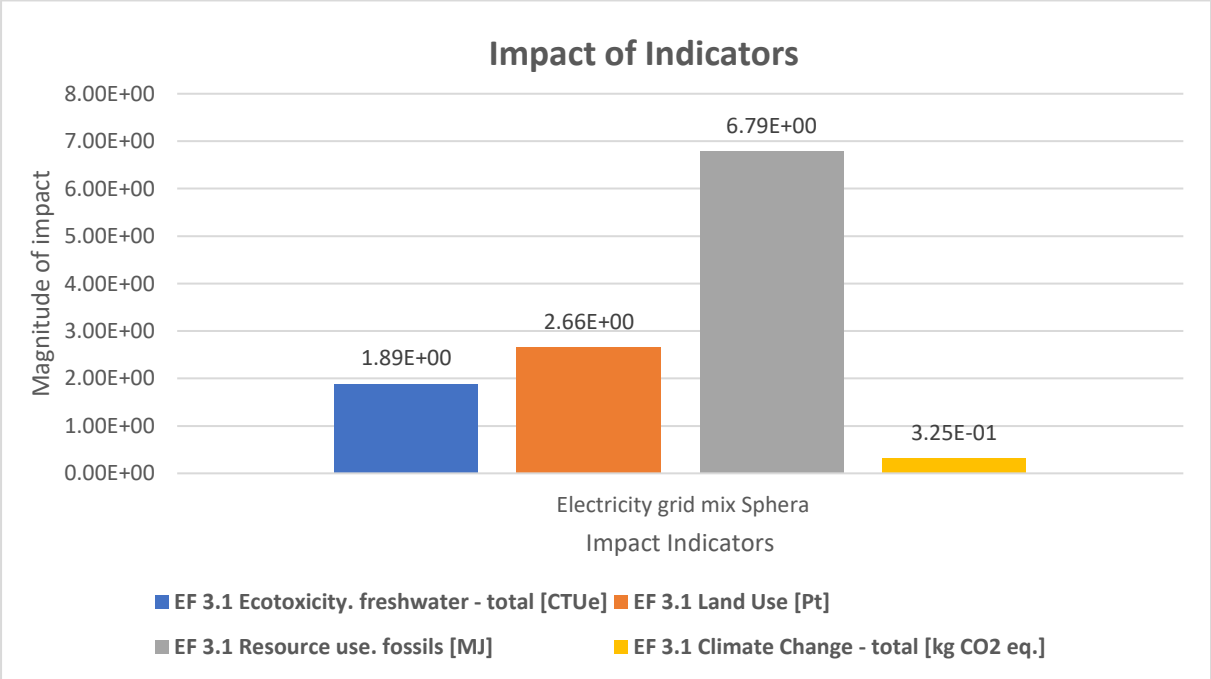

Figure 34: Environmental Footprint of Electricity grid [44]

environmental indicators, including Ecotoxicity, freshwater - total, Resource use - fossils, Land Use, and Climate Change – total.

With an impact magnitude of 1.89E+00 CTUe for Ecotoxicity, freshwater – total, the production of electricity, often reliant on fossil fuels within the energy grid, leads to the release of pollutants and contaminants into freshwater systems. The combustion of fossil fuels emits pollutants that can contaminate water bodies, adversely affecting freshwater ecosystems and biodiversity, thereby increasing the Ecotoxicity, freshwater - total indicator. The extraction and utilization of fossil fuels for electricity generation significantly contribute to the depletion of natural resources, impacting Resource use – fossils with the highest impact value of 6.79E+00 MJ, as shown in the graph. Fossil fuel extraction involves energy-intensive processes such as mining and drilling, which further deplete non-renewable resources.

Additionally, with an impact of 2.66E+00 Pt, Land use is affected by electricity production, which requires infrastructure like power plants, transmission lines, and substations, often necessitating land use. The development and maintenance of these facilities contribute to land use impacts, which impact natural habitats and ecosystems.

Furthermore, Climate Change - total, with an impact of 3.25E-01 kg CO_2 eq., is influenced by the combustion of fossil fuels for electricity generation. Climate change is a result of this process of atmospheric emission of greenhouse gases, such as carbon dioxide (CO_2). The entire impact of these emissions on global warming and other climate parameters is measured as Climate change- total.

To address the environmental impact associated with electricity consumption and mitigate issues related to Ecotoxicity, freshwater - total, Resource use - fossils, Land Use, and Climate change - total, a shift towards renewable energy sources such as solar, wind, hydroelectric, and geothermal energy is recommended. These sources generate electricity with minimal greenhouse gas emissions, reducing the Climate change - total impact and decreasing reliance on fossil fuels. Additionally, implementing energy efficiency measures across industries and households can reduce overall electricity demand. This includes upgrading to energy-efficient appliances, improving insulation, and optimizing industrial processes to decrease Resource use - fossils. Embracing green building standards and technologies is also crucial for reducing energy consumption in buildings, utilizing passive design strategies, energy-efficient lighting, and efficient Heating, Ventilation, and Air Conditioning (HVAC) systems, while integrating renewable energy systems like rooftop solar panels.

Furthermore, encouraging responsible land use practices, such as siting power plants on degraded or non-prime agricultural land and implementing reforestation or land restoration projects, can minimize the environmental impact of electricity infrastructure development. Establishing laws and regulations that provide incentives for adopting energy efficiency and renewable energy initiatives is essential. Some examples of these are carbon pricing systems, tax incentives, subsidies, and strict environmental requirements for power generation.

This study conducted on various graphene production methods reveals significant disparities in environmental impact, mainly driven by energy consumption and chemical usage. The Hummers method stands out with the highest environmental impact due to its substantial energy requirements and chemical usage, leading to significant emissions and resource depletion. In contrast, approaches like Marcano's method show improved environmental performance by reducing reliance on high-energy inputs like phosphoric acid. Chemical oxidation and thermal reduction methods also raise concerns, especially regarding Ecotoxicity, freshwater - total and Resource use, fossils.

Electricity consumption emerges as a critical factor contributing to Environmental Footprints across different production methods, notably influencing indicators such as Ecotoxicity, freshwater-total impact, Resource use, Land use, and Climate change-total. The reliance on fossil fuels for electricity generation leads to pollution and resource depletion, emphasizing the need to shift towards renewable energy sources and adopt energy efficiency measures to mitigate these impacts.

In summary, to minimize the Environmental Footprint associated with graphene production, it is essential to prioritize energy efficiency of the production process, reduce chemical usage, and transition towards renewable energy sources. Adopting sustainable production methods like Marcano's method, Annealing method, Ultrasonic Exfoliation can significantly improve environmental performance, while broader initiatives such as green building standards and responsible land use practices can further mitigate environmental impacts associated with electricity generation.

6. Conclusion

This study has conducted a comprehensive analysis of energy consumption and its impact on the Environmental Footprint throughout the production of graphene materials. By interpreting the relationship between energy consumption and environmental impact within various production processes, this research has identified substantial variations in environmental impacts among different methods. The objective of identifying graphene material inventories across different production methods using a literature review and pattern analysis with distinct functional units has been achieved. The findings reveal that the examined production processes, predominantly conducted at a laboratory scale with high energy consumption, significantly influence the Environmental Footprint. The insights gained from this study will contribute to developing a robust graphene database, which is essential for in-depth analyses of graphene composite and polymer applications and for assessing associated environmental impacts. The study also aimed to calculate the PEF absolute single score for each graphene production process using the EF 3.1 impact indicator method, providing valuable metrics for environmental assessment.

However, several limitations in this research highlight areas for future investigation and improvement. A notable constraint is the scarcity of comprehensive emission data associated with graphene production processes, particularly regarding land, water, air and soil impacts. Additionally, more research is needed on waste utilization to mitigate environmental impacts effectively. Collaboration with authors of relevant publications and enhanced data collection efforts along with graphene production will be crucial for addressing these limitations and refining modeling and calculation methodologies. Another critical area for future exploration is the determination of energy consumption hotspots within graphene production processes. Understanding the stages with the highest energy demands will facilitate targeted efforts to optimize energy usage and reduce Environmental Footprints. Future research endeavors should identify alternative chemicals and process modifications to minimize energy-intensive operations and enhance sustainability. Despite inherent uncertainties, this study underscores the importance of continued research and development to refine graphene production processes and minimize Environmental Footprints. By addressing research limitations and leveraging opportunities for process optimization highlighted in this study, researchers can advance toward more sustainable practices in graphene manufacturing. Collaborative efforts and ongoing investigation will be instrumental in realizing the full potential of graphene technologies while mitigating their environmental impacts responsibly. This study sets the stage for future research

endeavors to delve deeper into energy efficiency and environmental sustainability within graphene production, ultimately contributing to the advancement of cleaner and greener materials technologies.

Appendix A

This section introduces the inventory of graphene production at the laboratory scale

A.1 Chemical reduction process [26]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	0.6624	kg	Literature [26]
Graphite	0.003	kg	Literature [26]
H_3PO_4 (phosphoric acid)	0.0676	kg	Literature [26]
Hydrazine	0.89	kg	Literature [26]
H_2O (Water)	5.8	kg	Literature [26]
H_2O_2 (hydrogen peroxide)	0.003	kg	Literature [26]
$KMnO_4$ (potassium permanganate)	0.018	kg	Literature [26]
Energy Consumption	1100	MJ/Kg	Literature [26]
Output per FU			
Graphene	1	kg	Literature [26]

Note:

1. As per the Author's response, due to a lack of data Author did not consider any emissions because they did not contribute to impact categories.

2. The inventory data utilized in the literature is provided here, with measurements originally reported in grams (g) and milliliters (ml). However, for the study, these units were converted linearly to kilograms (kg).

A.2 Electrochemical Exfoliation process [8]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
Potassium hydroxide	1.12	kg	Literature [8]
Graphite	3.91	kg	Literature [8]
H_2O (Water)	670	kg	Literature [8]
Energy Consumption	1850	MJ/Kg	Literature [8]
Output per FU			
Graphene	1	kg	Literature [8]

Note: The inventory data utilized in the literature is provided here, with measurements originally reported in grams (g). However, for the study, these units were converted linearly to kilograms (kg).

A.3 Ultrasonic Exfoliation process [39]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	0.63	kg	Literature [39]
Graphite	1.05	kg	Literature [39]
H_2O (Water)	21.05	kg	Literature [39]
Energy Consumption	384	MJ/Kg	Literature [39]
Output per FU			
Graphene	1	kg	Literature [39]
Liquid waste	20.600	kg	Literature [39]

A.4 Hydrothermal method [2]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	15.625	kg	Literature [2]
Graphite	0.63	kg	Literature [2]
Deionized water	398.78	kg	Literature [2]
$KMnO_4$ (potassium permanganate)	1.98	kg	Literature [2]
H_2O_2 (hydrogen peroxide)	1.46	kg	Literature [2]
Urea	1.25	kg	Literature [2]
Ethanol	102	kg	Literature [2]
Energy Consumption	2386.80	MJ/Kg	Literature [2]
Output per FU			
N-rGO	1	kg	Literature [2]

A.5 Annealing method [2]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	31.25	kg	Literature [2]
Graphite	1.25	kg	Literature [2]
Deionized water	497.57	kg	Literature [2]
$KMnO_4$ (potassium permanganate)	3.95	kg	Literature [2]
Ammonium nitrate	2.50	kg	Literature [2]

H_2O_2 (hydrogen peroxide)	2.93	kg	Literature [2]
Ethanol	204	kg	Literature [2]
Energy Consumption	502.59	MJ/Kg	Literature [2]
Output per FU			
N-rGO	1	kg	Literature [2]

A.6 Hummers method [16]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	62.56	kg	Literature [16]
Graphite	1.00	kg	Literature [16]
$NaNO_3$ (Sodium nitrate)	0.75	kg	Literature [16]
Deionized water	10,666	kg	Literature [16]
$KMnO_4$ (potassium permanganate)	5.00	kg	Literature [16]
HCL (hydrochloric acid)	9.47	kg	Literature [16]
H_2O_2 (hydrogen peroxide)	5.80	kg	Literature [16]
hydrazine	12.36	kg	Literature [16]
Ammonia, Liquid	47.19	kg	Literature [16]
Energy Consumption	19881.39	MJ/Kg	Literature [16]
Output per FU			
rGO	1	kg	Literature [16]

A.7 Marcano's method [16]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	12.69	kg	Literature [16]
Graphite	0.96	kg	Literature [16]
H_3PO_4 (Phosphoric acid)	215.87	kg	Literature [16]
Deionized water	4884.8	kg	Literature [16]
$KMnO_4$ (potassium permanganate)	5.73	kg	Literature [16]
HCL (hydrochloric acid)	146.59	kg	Literature [16]
H_2O_2 (hydrogen peroxide)	1.39	kg	Literature [16]
hydrazine	0.02	kg	Literature [16]
Diethyl Ether	45.49	kg	Literature [16]
Ethanol	100.65	kg	Literature [16]
Methanol	880	kg	Literature [16]
Energy Consumption	315.03	MJ/Kg	Literature [16]
Output per FU			
rGO	1	kg	Literature [16]

A.8 Chemical oxidation process [8]

Inputs per FU			
Material name	**Quantity**	**Unit**	**Data origin**
H_2SO_4 (Sulfuric acid)	54.77	kg	Literature [8]
Graphite	1.19	kg	Literature [8]
$NaNO_3$ (sodium nitrate)	0.60	kg	Literature [8]
Water	372.41	kg	Literature [8]
$KMnO_4$ (potassium permanganate)	3.57	kg	Literature [8]
$Ca(OH)_2$ (Calcium hydroxide)	38.08	kg	Literature [8]
H_2O_2 (hydrogen peroxide)	2.07	kg	Literature [8]
Argon	14	kg	Literature [8]
Energy Consumption	296.70	MJ/Kg	Literature [8]
Output per FU			
rGO	1	kg	Literature [8]

Note: The inventory data utilized in the literature is provided here, with measurements originally reported in grams (g). However, for the study, these units were converted linearly to kilograms (kg).

A.9 Energy Consumption of Graphene Production

Method	Quantity	Energy Consumption (MJ/kg)
Chemical reduction of graphite oxide [26]	1 kg of Graphene	1100
Electrochemical exfoliation of graphite (KOH 15V) [8]	1 kg of Graphene	1850
Ultrasonic Exfoliation [39]	1 kg of Graphene	284
Hydrothermal method [2]	1 kg of N-rGO	2386.80
Annealing method [2]	1 kg of N-rGO	502.59
Hummers Method [16]	1 kg of rGO	19881.39
Marcano's Method [16]	1 kg of rGO	315.03
Chemical oxidation and thermal reduction [8]	1 kg of rGO	296.70

Note: The inventory data utilized in the literature is provided here, with some measurements originally reported in kWh. However, for the study, these units were converted linearly to MJ.

A.9 Dataset Source Information

Material name	Nation	Source	Data origin
H_2SO_4 (Sulfuric acid)	RER (Rest of Europe)	Sphera	Literature [44]
Graphite	RER (Rest of Europe)	Ecoinvent	Literature [45]
$NaNO_3$ (sodium nitrate)	RER (Rest of Europe)	Ecoinvent	Literature [45]
Water (Desalinated)	RER (Rest of Europe)	Sphera	Literature [44]
$KMnO_4$ (potassium permanganate)	GLO (Global)	Ecoinvent	Literature [45]
$Ca(OH)_2$ (Calcium hydroxide)	DE (Germany)	Sphera	Literature [44]
H_2O_2 (hydrogen peroxide)	DE (Germany)	Sphera	Literature [44]
Argon	DE (Germany)	Sphera	Literature [44]
Tap water	RER (Rest of Europe)	Sphera	Literature [44]
H_3PO_4 (phosphoric acid)	US (United States)	Sphera	Literature [44]
Hydrazine	RER (Rest of Europe)	Ecoinvent	Literature [45]
Potassium hydroxide	US (United States)	Sphera	Literature [44]
HCL (hydrochloric acid)	DE (Germany)	Sphera	Literature [44]
Ammonia, Liquid	RNA (Rest of North America)	Sphera	Literature [44]
Diethyl Ether	RoW (Rest of World)	Ecoinvent	Literature [45]
Methanol	RNA (Rest of North America)	Sphera	Literature [44]
Urea	DE (Germany)	Sphera	Literature [44]
Ethanol	GB (United Kingdom)	Sphera	Literature [44]
Ammonium nitrate	DE (Germany)	Sphera	Literature [44]
Electricity	RER (Rest of Europe)	Sphera	Literature [44]

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" I herewith declare that I have composed the present thesis myself and without use of any other than the cited sources and aids. Sentences or parts of sentences quoted literally are marked as such; other references with regard to the statement and scope are indicated by full details of the publications concerned.

In particular, I have not utilized commercial advice. Third parties have neither received direct nor indirect monetary benefits from me for actions which are related to the content of the submitted work. The thesis in the same or similar form has not been submitted to any examination board and has not been published.

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