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GRAPHITE PRODUCTION

20 January 2022

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This is the Submitted Version of the article submitted for publication to Journal of Industrial

Ecology.

Final Published Version of this article can be accessed via the publisher's website (Wiley

Periodicals LLC) and via this link: http://doi.org/10.1111/jiec.13234

TOWARD A LIFE CYCLE INVENTORY FOR GRAPHITE PRODUCTION

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ABSTRACT

Global electrification of mobility and energy storage is driving an unprecedented demand for Lithium-ion batteries (LIBs) for which graphite is one of the major components. Multiple prior studies have attempted to assess the environmental footprint of LIBs by way of life cycle analysis (LCA), and the poor quality of inventory data on the production of graphite (at various purities) has been highlighted consistently. This work reviews the available inventories used in the assessment of natural and synthetic battery-grade graphite production, and demonstrates that some upstream, downstream, and peripheral processes – including important processes associated with mining, calcination and other steps – are often omitted, leading to greatly underestimated impacts. It proposes a new rigid framework for comparing different graphite production routes and a corresponding indicative inventory for synthetic graphite production. This inventory is used to estimate the global warming potential and energy demand of synthetic graphite, yielding results of 20.6 kgCO2-eq/kg and 45.9 MJ/kg, respectively, suggesting that prior literature may have underestimated these results by a factor of two or more. The work concludes by highlighting the need to evaluate a broader suite of applicable impact categories and to fully account for the full suite of by-and co-products in future LCAs.

KEYWORDS

1. INTRODUCTION

The global demand for graphite has tripled in size between 2015 and 2020 and is forecast to reach 1.9 million tonne by 2028. Within this, the market for battery-grade graphite is expected to be US$14 billion by 2025, with a compound annual growth rate of 26% (Andrew Miller, 2020; Benchmark Minerals Intelligence, 2019; Clark, 2018; European Carbon and Graphite Association, 2018; Leyland, 2019). One of the main drivers for these market increases is the increase in demand for electric vehicles (EVs) (Zhang & Fujimori, 2020). Graphite is an essential material used for EV battery anodes and will likely remain as the preferred material for this purpose (Asenbauer et al., 2020; Wurm et al., 2018). Graphite is the third most abundant material (by weight) in the lithium-ion batteries (LIBs) deployed in the EVs; there is more graphite than lithium in a LIB battery. For example, a Tesla Model S requires as much as 54 kg of graphite (Tesla S) (Graphit Kropfmühl GmbH, 2019; Olson et al., 2016).

There is continued focus on the environmental performance of electric vehicles, with life cycle assessment (LCA) being the key methodology used to assess this (Hawkins et al., 2013). A critical component of completing an LCA is the life cycle inventory (LCI), which describes the material, energy, waste and associated environmental flows for a product or service system. Environmental Product Declarations (EPD), described as type III environmental declarations under ISO 14025 and the European Commission's Product Environmental Footprint (PEF) are both used to facilitate broader use of LCA and as tools supporting external communication and public procurement (Del Borghi et al, 2019), and to limit the methodological variability of LCAs (Zamagni et al, 2012). Development of a type III environmental declaration for any product relies of pre-defined product category rules (PCR)

New European regulations require that all EV parts including batteries to be accompanied by a form of environmental declaration from July 2024 (European Battery Alliance, 2020). Despite this, no PCR document currently exist for graphite, although some attempts have been made (Peters & Weil, 2018; Schmuch et al., 2018; Weimer et al., 2019).

A form of PCR was published for high specific energy rechargeable batteries used in mobile applications in 2018 (The Advanced Rechargeable & Lithium Batteries Association, 2018). This document cites a lack of a suitable LCI for graphitic anode production, and instead prescribes the use of an LCI for carbon black as a proxy. Carbon black is a distinctly different product with a substantially less energy intensive pathway, and the use of this LCI would inevitably result in underestimated environmental impacts for these batteries.

When considering an LCI for graphitic anode production, it is worth considering existing LCAs on LIBs. Extensive reviews conducted by Kim et al. (2016) and Peters et al. (2017) revealed that there are four underpinning studies which provide LCIs for the materials used in LIB components: Dunn et al., (2012), Ellingsen et al. (2014), Majeau et al. (2011) and Notter et al. (2010). Of these, Dunn et al. (2015) and Notter et al. (2010) provide more detail on battery-grade graphite production, however the data is incomplete as discussed later in this paper.

This lack of robust data for graphite production means that there is an urgent need for a rigorous and transparent LCI for graphite production, which would facilitate higher quality LCA studies and EPDs/PEFs. In addition, there is an urgent need to develop PCR for graphite, again allowing for higher quality EPDs/PEFs.

In this paper, we research the existing gaps by firstly describing the production pathways for different grades of graphite. This provides a basis for then reviewing existing LCIs and LCAs for graphite production. We then propose a generalised system boundary for the

production of graphite of different grades and propose an LCI for synthetic graphite production, based on readily available data. The extent of underreporting of environmental impacts for battery-grade graphite production is then estimated by assessing this LCI using global warming potential and process energy demand indicators. The paper concludes by reporting on the additional work required to improve the LCIs and the development of a PCR, with a focus on allocation and impact assessment.

2. GRAPHITE PRODUCTION PATHWAYS

Globally, 20-40% of graphite is produced from mining and the remainder is synthesised from coke (European Carbon and Graphite Association, 2018). In LIBs anode market, natural graphite accounts for about 80% of the battery-grade graphitic material requirement (European Carbon and Graphite Association, 2018).

In this section, the two main production pathways for battery-grade graphite currently on the market are documented in detail, followed by a brief description of post-processing steps and emerging production technologies. Table 1 and Figure 1 summarise the manufacturing processes for synthetic and natural graphite and make references to the main markets for different graphite grades which are further discussed in section 6 of this paper.

Table 1. Alignment of the process stages between two graphite manufacturing methods.

Generalized process step / corresponding intermediate products and graphite markets	Natural graphite	Synthetic graphite
Raw materials extraction and transportation / raw materials	Ore containing flake graphite is mined by open pit or underground and then trucked to the plant, which is generally in the vicinity of the mine.	Petroleum oil is pumped from underground by oil rigs and either pumped to the refinery via pipelines or delivered in sea tankers. Transportation distances can be considerable including a large proportion of international import/export of crude oil. Coal is mined by open pit or underground and then trucked to the plant, which is generally in the vicinity of the mine.
Preparation of feed for graphitization, increasing fixed carbon content /	None	Refining including vacuum distillation and coking followed by transportation and calcination of vacuum residue.

(needle) coke and tar pitch		Coal is processed in coking ovens, and tar is further refined to produce tar pitch of acceptable quality.
Graphitization to achieve graphitic structure / (pre-purified) graphite. Markets 1,2&3	None	Transportation and conditioning of the needle coke followed by mixing with tar pitch (if applicable) and high temperature treatment
Beneficiation and regeneration of valuable reagents / 90-98% by weight graphite. Markets 4&5	Multiple stages of attrition milling and flotation
Sometimes involves acid-alkali leaching.
Inclusive of recovery of valuable products (if any)	none
Purification / 99.95% by weight graphite. Market 6	Can be in form of acid/alkali leaching or roasting, halogen roasting, and thermal	None for primary graphite powder, however secondary graphite powder may require some form of purification
Transportation between process stages	Flotation concentrate to purification plant
High purity graphite to post-processing
The two may be undertaken at the same facility	Crude oil to refinery and coal to calcination plant
Green coke for calcination or calcined coke to graphitization
Tar pitch to graphitization
High purity graphite to post-processing
Post-processing (to anode material). Markets 7,8&9	Milling, classification, shaping and coating	Milling and classification may not be required for primary graphite powder
Shaping and coating are generally applied
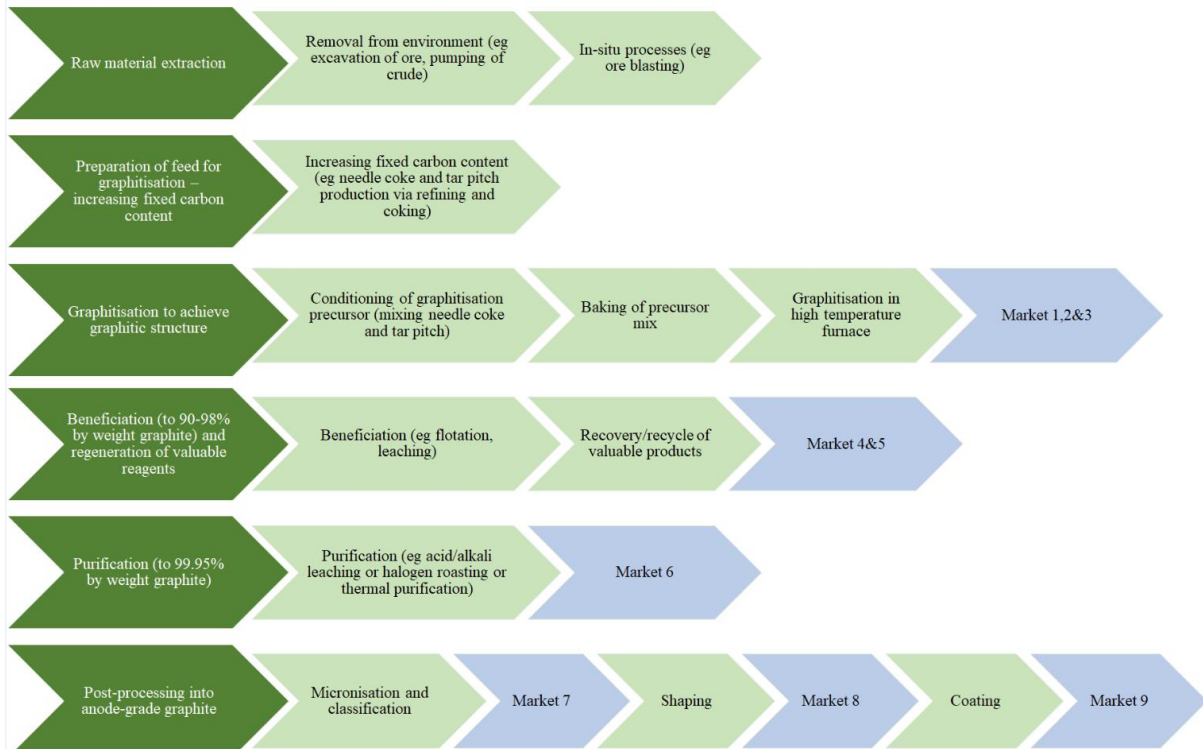| Product conditioning and bagging	Deagglomeration, ultrasonic and magnetic separation of metal impurities, batch mixing, bagging	
Transportation to the battery manufacturer	Anode manufacturing and battery assembly, covered by LCAs on LIBs	

Figure 1. Graphical representation of graphite production and graphite markets.

2.1 NATURAL GRAPHITE

Natural flake graphite is mined in either open pits or underground. The ore may contain between 2-30% by weight of graphitic carbon (Scogings et al., 2015), and development of the deposits containing between 5-20% is most common. Graphite therefore requires beneficiation, especially for use in batteries. The majority of flake graphite production utilizes multiple alternating stages of attrition milling and flotation which can include up to seven milling-floating steps (Jara et al., 2019). Additives, such as pine oil, kerosene or diesel, and sodium silicate, are used in small quantities (hundreds of grams per tonne of output) to increase the efficiency of flotation (911 Metallurgist, n.d.). LCI data for this processing can be garnered from mine feasibility studies and mine operation environmental reports, however these are not usually available in public domain.

Occasionally, further processing to reach 90-98% purity involving acid and/or alkali leaching is used to remove silicate and carbonate impurities (Jara et al., 2019). This leaching is often not required, and the flotation step results in high purity (98% by weight) graphite.

High purity graphite (99.95% by weight) required for battery production, can be achieved through hydrofluoric acid leaching, halogen or alkali roasting, or thermal treatment in an inert atmosphere. Historically, this part of the process has been undertaken in China and industry data is not published; therefore, environmental impacts cannot be adequately quantified. Hydrofluoric acid leaching treatment has been used but the demand for cleaner production is driving a trend away from this chemically intensive treatment.

Most of the energy required for natural graphite production is delivered from heat from the combustion of fossil fuels, with the remaining 20-25% being delivered from electricity.

2.2 SYNTHETIC GRAPHITE

It is understood that conflicting information exists around the feedstock for the synthetic battery-grade graphite manufacturing. Currently, the highest purity graphite is produced from petroleum needle coke, and this process is the most emission and energy intensive due to the complexity of the production chain (Wissler, 2006). As the technology advances, petroleum coke (un-calcined), coal coke and raw coal may become viable alternative feedstocks for the manufacturing of battery-grade graphite. Generalised process stage descriptions in Table 1 and Figure 1 allow for these variations.

The production of synthetic graphite from petroleum involves three distinct processes which are often undertaken in separate facilities, and frequently by different manufacturers. These processes are:

1. Green coke production: Green coke is produced either as an intermediate product from oil refining or via the catalytic cracking of heavy oils with an overall mass yield in the order of 2.5% of crude oil (Dante, 2016). The green coke contains in the order of 5-15% volatiles which are removed by calcination.

2. Calcination: Green coke is calcined to produce needle coke, which has a lower volatile content, and a purity of between 97-99% by weight (Imerys Graphite & Carbon, 2018; WAVE Intl, 2019).

3. Graphitization: The calcined needle coke is conditioned and graphitized at above 2500°C to produce high quality and high purity graphite (Bogacki et al., 2010; Marsh et al., 1997). Conditioning may involve grinding to a specific size, impregnation with a binder, and baking at anywhere between 850-1300°C (Bogacki et al., 2012; Marsh et al., 1997).

Despite these processes being well understood, LCAs often only reference the last process and only in the context of extruded electrodes manufacturing, e.g., for use in steel production and aluminium industry. In this process (Dante, 2016), needle coke is mixed with 15-30% by weight liquid binder (pitch), this precursor is shaped into required form, baked to eliminate volatiles introduced with the binder, and finally graphitized. Repeated baking and cooling steps are usually required, and the overall process takes months to complete. The final form is then machined, and the filings (known as secondary graphite) can be further processed into powders and granules. Secondary graphite is often contaminated, and this process is not necessarily applicable to the manufacturing of graphite used in LIBs.

The manufacture of primary synthetic graphite powder may or may not involve the addition of a binding agent, and therefore baking. Notably, patented methods (for example, (Tian, 2017)) refer to the use of a binding agent whereas methods described in other scientific publications reference direct graphitization of high-carbon precursor in inert atmosphere (Xing et al., 2018). These publications are consistent in indicating that the graphitization to achieve primary powder graphite is significantly faster (6-24 hours). Such short processing time may be attributed to a small sample mass used in these works compared to industrial processes.

Some researchers describe a process by which graphite is obtained from coal and pitch coke (Xing et al., 2018). These materials can undergo coking to produce metallurgical coke with approximately 75% by weight yield (also producing tar pitch at approximately 2-4% by weight of coal feed). Metallurgical coke and anthracite coal can be used for further graphitization with three major limitations: 1) low graphitization yield due to intrinsic carbon structure not suitable for mesophase formation, 2) high level of contamination by metals, and 3) reduced conductivity of final product (GrafTech Int Ltd, 2019). Resulting

graphite has a total carbon content of 88-92% by weight (GrafTech Int Ltd, 2019; Stansberry, 1999) which may not be fully graphitized and therefore, to the best of the authors' knowledge, not suitable for LIB applications without further processing.

Unlike in case of natural graphite, further purification of synthetic graphite derived from needle coke is generally not required due to high graphitization process temperatures. Graphitization is often performed in electrically heated Acheson or Castner furnaces, however up to 25-30% of fossil fuel-derived heat is required for baking (Bogacki, 2010, 2012). This can be used to assess indirect emissions associated with the process.

The direct emissions associated with synthetic graphite production are not well reported. However, (Bogacki et al., 2010, 2012) have conducted a systematic study of direct emissions from baking and graphitization processes. Other direct emissions associated with upstream production activities, including oil and coal mining and processing into needle coke and coal tar pitch respectively, are documented in existing LCA databases.

2.3 POST-PROCESSING

Post-processing is required for high purity natural or synthetic graphite to produce the powder shape and sizes which impart the surface properties needed for use in LIB anodes. This post-processing consists of milling, classification, shaping (spheronization), coating to reduce surface area, and conditioning to ensure safe packaging.

There is no data in the public domain confirming whether coating is always required, and whether sizing and shaping may take place before graphitization of synthetic graphite. Other process variations may exist, for example, the requirement for milling and classification and its intensity can vary depending on the particle size distribution of purified graphite. Further, depending on the shape of the pre-processed graphite, efficiency of spheronization may vary and the pass rate is generally in the order of 40-60% for natural and synthetic graphite, respectively. There is significant IP around coating processes, frequently specific for a battery performance; the coating agents as well as the thermal treatment conditions are not usually disclosed.

The coated material tends to bind together due to the adhesive pitch coating that is applied and subsequently carbonized in the carbon coating process. Therefore, deagglomeration of the particles is required before bagging. Removal of ultrafine metal particles is required, which are from wear of moving mechanical parts in post-purification processing equipment and transfer systems. Other requirements include mixing and ultrasonic screening of multiple final product batches to increase product homogeneity and to reduce static charge between particles prior to bagging. The final product is then packaged for transportation to customers.

2.4 EMERGING PRODUCTION TECHNOLOGIES

A range of processes being developed to produce graphite from other carbon-rich materials including plastic waste and biomass (Banek et al., 2018; Fan et al., 2012; Karim et al., 2019; Ko et al., 2020; Shi et al., 2018; Tran et al., 1997; Xing et al., 2018). However, to date CarbonScape Ltd (Conner et al., 2016) is the only one of these groups who have reported a fully developed process. The scope of this article is limited to commercialised graphite production processes, and therefore these emerging technologies are excluded from the remainder of this work.

3. REVIEW OF EXISTING LCIS AND LCAS

In this study, three underpinning LCI data sources for natural and synthetic graphite manufacturing were reviewed (Dai et al., 2019; Gao et al., 2018; Notter et al., 2010). Dai et al.'s study is an update to (Dunn et al., 2015). This latter reference includes a quality assessment of other synthetic graphite inventories.

A summary of the data reviewed in this work is provided in Table 2 and Figure 2. Figure 3 depicts the scopes of the examined LCA studies, highlighting the limitations of LCI data and corresponding assessment results. Overall, previous graphite production LCAs have different system boundaries, methodologies to account for co-production (allocation), life-cycle impact assessment methods and categories, all of which lead to a variable and incomparable results.

3.1NATURAL GRAPHITE

Described in Figure 3a (Notter et al., 2010) LCI for battery-grade graphite is most commonly used for the assessment of the contribution of graphite manufacturing to the overall battery impact. This study estimates the total energy consumption at 18.4 MWh/tonne and cumulative GWP at 2.15 tonne CO2-eq/tonne. Other categories assessed in this study include Ecoindicator 99 H/A which reflects on toxic effects on human health and ecosystems as well as on resource quality, and abiotic depletion potential which reflects specifically on the use of resources. However, examination of Notter's LCI reveals:

1. Mining of graphite ore is approximated from lime production.

2. Theoretical approximations are made with respect to energy requirements for upgrading to required purity by means of thermal treatment and are based on thermodynamic calculations, with the furnace efficiency not taken into account.

3. Over 90% of the energy (40GJ/t graphite) is included as calorific value of coal; however, coal is not used as a heating source.

4. Post-processing (milling, classification, shaping, and coating) of high-purity graphite to produce battery-grade graphite is omitted or not explicit, thus associated total energy and environmental intensities is underestimated.

Notter's LCI is incorporated in ecoinvent 3.6 and GaBi databases to represent both natural and synthetic graphite manufacturing processes.

An assessment by (Gao et al., 2018) (Figure 3b) is based on a mixture of primary and secondary data for a representative Chinese manufacturing process for natural flake graphite. This study distinguishes between various stages of graphite mining and processing including the final purification step necessary to upgrade graphite to battery-grade purity and post-processing into coated graphite. The total process energy demand is assessed as

31.25MWh/t with over 40% consumed at the post-processing stage and the associated GWP is estimated at 5.3 tonne CO2-eq/tonne graphite assuming manufacturing is in China. This study is the most comprehensive of the currently available studies on natural battery-grade graphite production in terms of energy and greenhouse gas emissions data, however no other impact categories were assessed. Unfortunately, the original inventory is not available publicly.

3.2 SYNTHETIC GRAPHITE

The most recent assessment by (Dai et al., 2019) (Figure 3d) included updated, yet still secondary, data around synthetic graphite manufacturing. Dai et al. includes data around graphite production from petroleum coke and coal tar pitch; however, it does not account for either the upstream activities associated with oil mining, refining and calcination or the post-processing of high purity graphite. The energy inputs were reported as 25 MWh/tonne and emissions contributing to GWP at 4.9 tonne CO2-eq/tonne; and because of the omission of upstream activities, these values are likely underestimated. Dai's data are an extension to the original dataset included in the GREET life cycle analysis model created and reviewed by Argonne National Laboratory (Dunn et al., 2012, 2015) (Figure 3c). Dai's data includes emissions associated with electricity generation which are said to be based on China-average grid emissions; however, it is not explicitly stated why the total energy demand is different in the two studies. Additionally, a calculation using Dai's data indicates that the electricity-related emissions are about 250gCO2/kWh, which suggests manufacturing in South-West regions where hydropower is available (Qinghai, Sichuan, Yunnan) (Li et al., 2017). There are indeed graphite processing facilities in those provinces, however this regional coverage does not represent all synthetic graphite production in China. Major production facilities are located in Jilin, Liaoning, Gansu, and Jiangsu, where electricity-related emissions are higher than 250gCO2/kWh (Li et al., 2017).

Table 2 Data summary for Natural and Synthetic graphite, only GWP and process energy demand is shown.

Reference study	Notter 2010	Gao 2018	Dunn 2015	Dai 2019
type of graphite	natural	natural	synthetic	synthetic
Assumptions	assumptions: lime mining as proxy; added hard coal and electricity to account for extra processing, talks about coal graphitization; calorific value of coal included in energy even though not used for this purpose	based on Chinese research and government publications, not accessible	Assumptions: surpasses other estimates for synthetic; graphitization and baking; oven energy and direct emissions only; ~30% thermal efficiency of Acheson furnace; no emission control	Assumptions: same as Dunn 2015 + indirect emissions; reason for additional energy requirement unknown
Extraction from environment and transportation of raw material / crude oil or graphitic ore	Assuming diesel only: 0.018MJ/kg	27.7MJ/Kg 1.04kgCO2/kg	not included	not included
Preparation of feed for graphitization / needle coke	NA	NA	not included	not included

Graphitization to achieve graphitic structure / pre-processed graphite	NA in this work is assumed that requirements of additional coke and electricity are more applicable for purification and post-processing	NA	22.2 MJ/kg g/ton (0.907tonnes) CO2 440,000 (0.485kg/kg) SOx 64,000 (70.6g/kg) NOx 9,300 (10.25g/kg) PM 4,100 (4.52g/kg)	89.9MJ/kg IPCC 100-yr GHG 4.86kg/kg SOx 79.8 g/kg NOx 13.5 g/kg PM 5.5 g/kg
Beneficiation and regeneration of valuable reagents / 90-98% by weight graphite	Assuming heating by oil and electricity as for natural: 0.21MJ/kg	14.1MJ/kg 0.79kgCO2/kg	NA	NA
Purification / 99.95% by weight graphite	Assuming additional electricity: 3.6MJ/kg	23.8MJ/kg 1.89kgCO2/kg	NA / not included	NA / not included
Post-processing to anode material	Assuming coke embedded energy is used here for heating: 40MJ/kg	46.9MJ/kg 1.6kgCO2/kg	not included	not included
Transportation between process stages	not reported	included, not explicit	not included	not included
Cumulative (total)	43.8MJ/kg IPCC 100-yr GHG 2.15kgCO2/kg	112.5MJ/kg IPCC 100-yr GHG CO2 5.316kgCO2/kg		

Process energy demand, MJ/kg

synthetic (Dai, 2019)	
natural (Gao, 2018)	
natural (Notter, 2010)	

Legend:
- Extraction from environment
- Raw material conditioning
- Graphitization
- Beneficiation
- Purification
- Post-processing to anode material
- Transportation

Axis: 0, 20, 40, 60, 80, 100, 120

GHG (as per IPCC 100-yr horizon), kgCO2-eq/kg

synthetic (Dai, 2019)	
natural (Gao, 2018)	
natural (Notter, 2010)	

Legend:
- Extraction from environment
- Raw material conditioning
- Graphitization
- Beneficiation
- Purification
- Post-processing to anode material
- Transportation
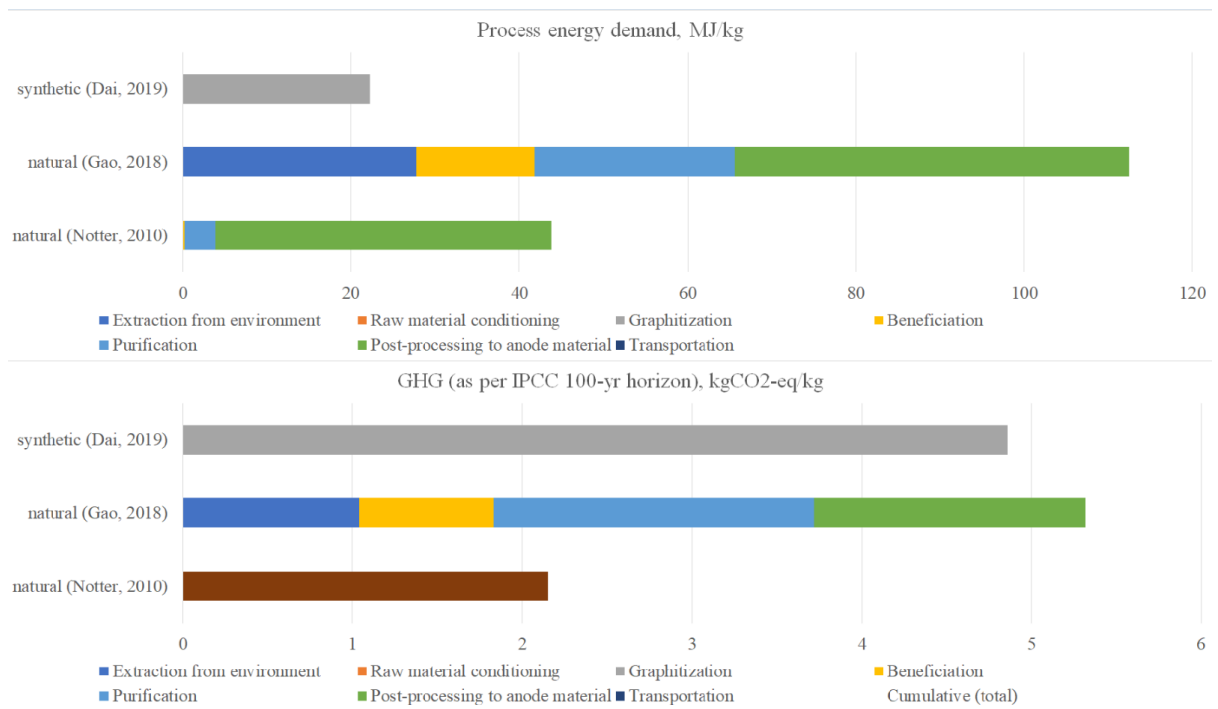- Cumulative (total)

Axis: 0, 1, 2, 3, 4, 5, 6

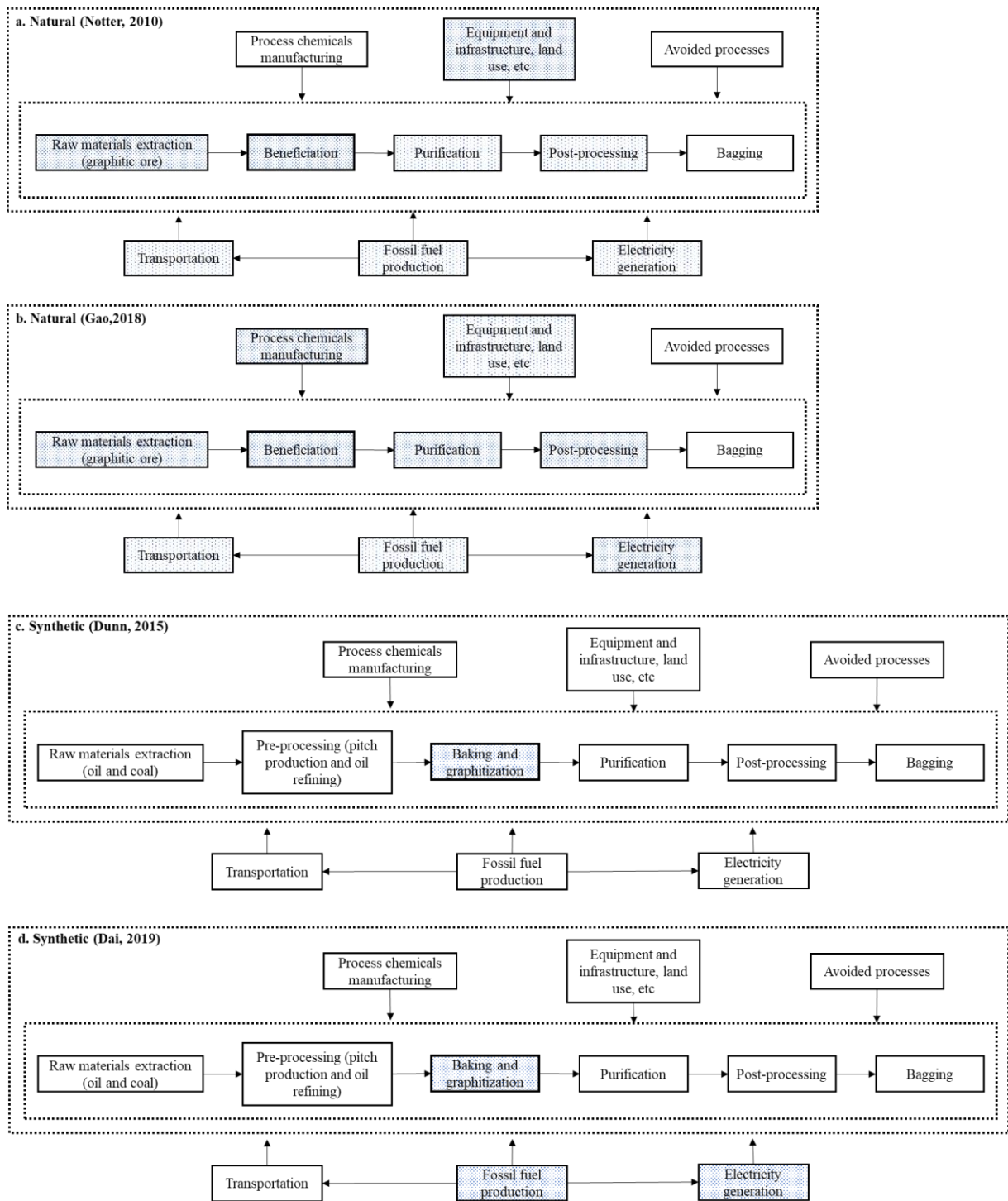Figure 2 Summary of LCA results (process energy demand and GWP) in examined studies.

Figure 3 Depiction of scopes of the examined LCA studies. Shaded areas indicate processes explicitly discussed within the corresponding publication; lightly shaded areas indicate that the description of how the corresponding process was accounted for is not explicit or the assumptions made about the process are questionable, and clear boxes show processes excluded from the assessment.

4. INVENTORY DEVELOPMENT

LCA study requires that functional unit and system boundary are defined in order to ensure the inventory is suitable. We propose the following approach to ensure consistency in the analysis of battery-grade graphite (also consider information in section 6 for alignment of LCI and impact categories).

The primary function of graphite in a LIB is to provide storage area for the lithium ions in a certain atomic ratio, and therefore a mass-based reference unit is considered most appropriate for the basis of LCI development. This approach to the reference unit also enables straightforward comparison of different co- and by-products and various manufacturing chains, and easier integration into future LCA studies.

The possible co-products in the graphite manufacturing chain and the market exits are shown in Table 1 and Figure 1. Only graphitic products are considered; however, the upstream processes also produce multiple marketable products. Transportation is applicable at each process stage.

A generalised system boundary for battery-grade graphite, including co- and by-products which can be applicable to different production routes, is provided in Figure 4. The applicability of the different production systems within this boundary will depend on the product and end-applications. For example, 98-99% pure graphite is a marketable product and can be used as-is in some applications. However, depending on the production method, it may or may not require an inventory for preparation of feed for graphitization, graphitization itself, beneficiation, and purification.
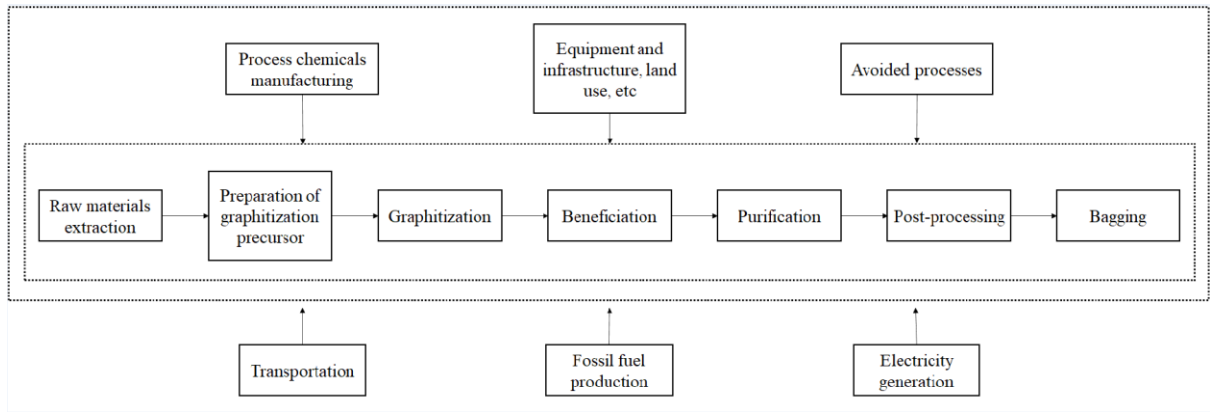
Figure 4 Generalised system boundary for battery-grade graphite production

4.1 INVENTORY FOR SYNTHETIC GRAPHITE

In this section, an alternative LCI for synthetic graphite is proposed based on publicly available data and the generalised system boundary described above.

The following data is required to complete a life cycle inventory for battery-grade synthetic graphite:

1. Coal is a common precursor for the tar pitch which is used for the preparation of graphitization precursor and also for coating of the final spherical graphite. Relevant data include coal mining, coking to produce tar and distillation of tar to produce pitch. In this work energy and emissions associated with the last distillation step are not included. Allocation is based on physical yield of tar pitch.

2. There are four types of petroleum coke produced via fluid and flexi-coking and by delayed coking. Of these, only highly anisotropic coke from delayed cokers is suitable as a precursor for synthetic graphite. Relevant data include crude oil mining, oil transmission to refinery, and refinery operation including delayed coker. Allocation is based on mass yield of green coke.

3. Green coke is further calcined, and in this work composition of calcined coke was calculated based on compositions of green coke and volatiles. Relevant dataset covers operation of calcination unit / plant.

4. Calcined coke is then mixed with tar pitch, baked to reduce volatiles and graphitized. Relevant dataset is commonly included in the databases as synthetic graphite manufacturing process.

5. No reputable sources could be identified for inventories for post-processing including micronizing, shaping and coating, including within commercial LCA databases.

There are research publications which focus on laboratory results for various process stages for both natural and synthetic graphite; however, the consistency of information across these publications is limited. Nevertheless, these studies can provide useful life cycle inventory data. Notably, most of the relevant inventories are available within all major LCA databases which would allow a more accurate and comprehensive analysis of the impacts for various purposes. For demonstration purposes, in this study an indicative life cycle inventory was developed from the publicly available data. Table 3 documents the inventory parameters and the respective sources. Note that any impacts beyond process GWP and process energy demand are not included and transportation between stages is discussed separately. This is to ensure consistency with the available datasets for synthetic graphite (Dunn et al., 2012,2015; Dai et al., 2019).

The following assumptions were made to relate data from different sources:

1. At refinery, 10% by mass conversion of crude to vacuum residue, coke yield from coking unit is 25% by mass. Total allocation of oil mining is therefore 10% (to produce sufficient amount of vac residue); 10% for atmospheric and vacuum distillation units; 25% for coking unit; 2.5% overall from well to coke including transmission.

2. Coal tar pitch yield is 2% of feed coal. Transportation from mine to coking oven (assumed to be based at a standalone coking facility, not at a steel mill) is omitted.

3. Yield of needle coke from calcination is 74%. Transportation from refinery to calcining plant is omitted.

4. Emissions from baking and graphitization are 0.4% and 2% respectively. Transportation from calcining plant to graphitization plant is omitted. Crushing of needle coke and mixing of pitch with needle coke is not included.

5. Resulting graphite is >99.9% pure and is in powder form, further purification is not required. However, micronization, shaping, and coating are required to upgrade to battery-grade.

Table 3 Life cycle inventory for synthetic graphite production

	Amount	unit	Notes/References
Coal inventory			
Output			
hard coal, extracted from mine, washed	1.00E+00	kg	
Input			
Energy	1.26E+00	kWh	(Wang et al., 2018), ~82% electricity, 12% petrol & 6% diesel
Emissions to air			
CO2	8.50E-01	kg	(Wang et al., 2018)
CO2	3.00E-02	kg	
NOx	1.00E-02	kg	
N2O	1.00E-02	kg	
CH4	1.50E-01	kg	
Coal coking inventory (for pitch production)			
Input			
Hard coal, extracted from mine, washed	1.00E+00	tonne	
Energy	4.2+00 (average from 3 references)	GJ	(Gupta, 2014; He et al., 2017; Song et al., 2019), ~3% electricity and balance blast furnace gas (assume similar to syngas)
Output			
Tar pitch	2.40E+01	kg	(He et al., 2017; Song et al., 2019; Stansberry et al., 1999), correlation via coke yield of 75%
Emissions to air			
CO2	2.00E+2 to 4.00E+2	kg	Ge (2016), correlation via coke yield of 75%, assume independent coking facility (not at steel mill)
CO	2.50E-2 to 8.46E-1	kg	(Environmental Protection Agency,

CH4	2.20E-2 to 1.55E-1	kg	1995, Valia, 2019), correlation via coke yield of 75%
NOx	2.60E-2 to 8.21E-1	kg	
Oil mining and transmission inventory			
Input			
Energy	9.37E-01	kWh	(Nimana et al., 2015), correlation via energy density 41.4GJ/m3 and mass density 1010kg/m3, ~8.5% electricity, 7% diesel, 84.5% NG
Output			
Oil crude	1.00E+00	kg	
Emissions to air			
GHG as CO2	9.80E-01	kg	(Gadalla et al., 2006; Nimana et al., 2015) correlation via energy density 41.4GJ/m3 and mass density 1010kg/m3
Oil refining inventory			
Input			
Oil crude	1.00E+00	kg	
Energy atm distillation	2.49E-01	kWh	Szlko (2007), assume 10% electricity and balance syngas
Energy vac distillation	1.46E-01	kWh	Szlko (2007), vac residue is 10% of crude, assume 10% electricity and balance syngas
Energy delayed coking	3.18E-01	kWh	Szlko (2007), coke is 25% of vac residue, assume 5% electricity and balance syngas
Output			
Green coke	2.50E-02	kg	Gabi database on-demand
Emissions to air			
GHG as CO2	2.75E-01	kg	Szlko (2007)
Calcination inventory			
input			
Green coke	1.00E+00	kg	

Energy	4.40E+00	kWh	(Chudnovsky et al., 2009) with heat recovery, assume 10% electricity and balance NG
Output			
Needle coke	7.40E-01	kg	(BS Envi-Tech p Ltd, n.d.)
Emissions to air			
CO_2	1.30E-02	kg	(Rawat & Duara, 1985), calculated based on composition and mass loss during calcination of 26%
CO	1.40E-02		
CH_4	1.55E-01	kg	
Baking inventory			
Input			
Needle coke	8.00E-01	kg	(Stansberry et al., 1999), 80-20 ratio of needle coke to pitch
Coal tar pitch	2.00E-01	kg	
Energy	3.50E+00	kWh	(Bogacki et al., 2012) Natural Gas
Output			
Graphitization precursor	9.96E-01	kg	(Bogacki et al., 2012), assuming 0.4% mass loss on baking
Emissions to air			
CO	2.67E-03	kg	(Bogacki et al., 2012)
CH_4	7.95E-05	kg	
NOx	2.46E-04	kg	
Graphitization inventory			
Input			
Graphitization precursor	1.00E+00	kg	
Energy	8.80E-01	kWh	(Bogacki et al., 2010; Dunn et al., 2012), Natural gas Note that this value is the same as reported in (Dunn et al., 2015)
Output			
>99% pure graphite	9.80E-01	kg	(Bogacki et al., 2012), assuming 2% mass loss on graphitization
Emissions to air			
CO	1.93E-02	kg	(Bogacki et al., 2010)

5. ASSESSMENT RESULTS AND DISCUSSION

As identified previously, analysis of natural graphite production chain conducted by Gao (2018) has clearly demonstrated how significantly underestimated the energy demand and the GWP of the process is when using existing data embedded within the LCA databases. This section aims to demonstrate the extent to which the GWP impact and cumulative energy demand of synthetic graphite are underestimated.

Figure 5 demonstrates the relative process energy demand at each process stage and corresponding GWP intensity. IPCC 100-yr horizon was used to evaluate GWP impact. It is quite clear that energy and emissions associated with the graphitization stage (3.2MJ/kg and 0.243kgCO2-eq/kg), which is often the only part of the process considered in the LCAs, forms a small portion of the overall impact. Graphitization combined with baking of the precursor (16.23MJ/kg and 7.734kgCO2-eq/kg) is another common system boundary used when reporting on graphitization impacts, however even this combination appears to cover less than half of the total. This high-level analysis suggests total GWP and energy demand of approximately 20.6 kg CO2-eq/kg and 45.9 MJ/kg, respectively, thereby demonstrating that the true impacts of synthetic graphite production can be more than twice as high as the values currently reported.
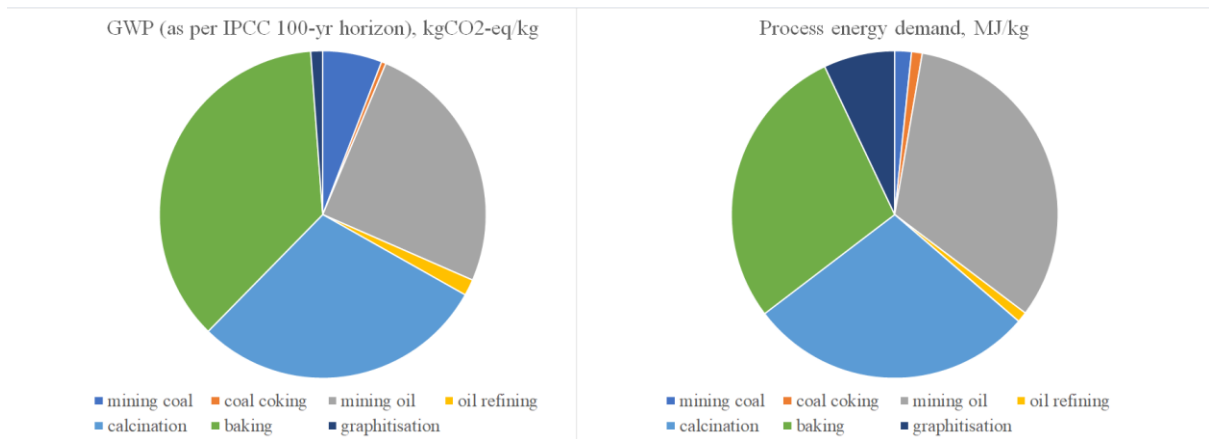
Figure 5 Global warming potential and process energy profile for synthetic graphite
production (numerical results shown in SI).

5.1 A NOTE ON TRANSPORTATION

Information on the complete supply chains for both natural and synthetic graphite is extremely scarce in the public domain. However, some assumptions can be made based on the public information on operation of major players such as Graftech, Imerys, SGL, Phillips66, and Syrah Resources. Combined with the guidelines (Cefic & ECTA, n.d.), the GWP impact associated with the shortest and the longest indicative transportation routes can be evaluated for both synthetic and natural graphite. The aim of this study is again limited to demonstrating the magnitude of the contribution from transportation to the overall impact. Overall, shortest and longest routes for synthetic and natural graphite are in the order of 20000-35000km and 1000-25000km respectively, with the corresponding GWP impact in the order of 0.3-0.35 and 0.015-0.45 kgCO2/kg graphite. Transportation appears to have minimal contribution to the overall GWP associated with natural and synthetic graphite production. It may however have a more substantial impact when financial aspects are considered.

6. FURTHER CONSIDERATIONS FOR THE DEVELOPMENT OF PCR FOR GRAPHITE PRODUCTION

To develop a type III environmental declarations for graphite products requires the development of pre-defined product rules (PCR). The motivation for conducting product environmental declarations is to enable comparison between products which fulfil the same function and to encourage positive market feedbacks for lower environmental impact products by providing transparent environmental information through LCA (Fet and Skaar, 2006, Fet et al., 2009). There are a number of EPD systems which can vary in methodology and PCR requirements, however in recent years attempts have been made to harmonize these approaches to type III environmental declarations (Minkov et al, 2015).

To develop PCR to reflect the graphite value chain and the different graphite products requires an open, participatory consultation of interested parties which could include graphite suppliers, manufacturers, trade associations, purchasers, users, consumers and NGOs or public agencies (ISO 14025). The development for the PCR document includes the definition of the product category, followed by the collection or development of an appropriate LCA and finally the specification of common goal and relevant rules for said product category. These rules can indicate what additional environmental information should reported, the data requirements and other instructions to ensure consistent and scientifically robust LCA.

6.1 ALLOCATION FOR CO- AND BY-PRODUCTS THROUGH THE VALUE CHAIN

As previously shown in Figure 1 and Table 1, the production of battery-grade graphite yields a range of co-products within the product system. Co-products refer to any two or more products coming from the same unit process or product system (ISO 14044). In line with ISO 14044 the burdens attributed to these co-products should be modelled using system expansion; however, it is not possible in case of graphite. In this case the allocation of impacts should be based on physical parameters or economic value. A brief description of the markets identified in section 2 is presented below and can assist in defining appropriate allocation rules. Physical yields from each process stage are highly dependent on production parameters and can vary significantly between producers.

Market 1: extruded graphite electrodes, permanent mould castings for foundries and other extruded/moulded/pressed shapes. Production follows the lengthy manufacturing process (up to 6 months) usually referenced for synthetic graphite. Precursor for electrodes is mostly petroleum needle coke due to coal coke having more impurities which make manufacturing more difficult and result in a lower quality of the final product. Coal coke can be used for foundry moulds and shapes. Ultra-high purity shapes are used in nuclear plants; however, details of purification are not easily available.

Market 2: secondary synthetic graphite powder. A by-product or waste product resulting from machining of extruded graphite electrodes. Generally, fairly inconsistent in quality and is mostly used in recarburizing industry and refractories; however, can also be used in LIBs after additional purification and post-processing. Also used as lubricant, as friction material, in foundries, and as additives.

Market 3: primary synthetic graphite powder. Information is extremely scarce; however, a few patents and research publications suggest that the manufacturing generally follows the

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same process as extruded electrodes, except the process is substantially faster (patents state between 6-24hrs). This is the precursor for graphite anodes for LIBs. Also used as friction material, lubricant in specialist applications, and as additives.

Market 4: large flake graphite, 95-98% purity. Used in refractories and for manufacturing of expanded graphite.

Market 5: fine flake graphite, 95-98% purity. Used as a precursor for manufacturing anode grade powder, in recarburizing industry and in refractories. Also used in foundries, as friction material and additives; can be used as lubricant if sufficiently pure. Used in pencils.

Market 6: high purity flake and synthetic graphite powder, >99.95% purity. Main precursor for anode grade powder for LIBs, used in other batteries/fuel cells and as an additive in battery cathodes. Also used as lubricant, as friction material in specialist applications, and as additives. Ultra-high purity (generally achieved by repeat purification or via a combination of purification methods) used in nuclear plants (synthetic only) and in medical applications.

Market 7: micronized natural and synthetic graphite. Applications of different size fractions include foundries, friction products, batteries, carbon brushes, conductive additives, lubricants, pencils, and powder metallurgy.

Market 8: spherical graphite. This is applicable for both, synthetic and natural graphite, and the main product is predominantly consumed by the battery market with small portion used in fuel cells and for lubrication. Waste stream from the process (between 30-70%, combined of out-of-spec fines) is used up by recarburizing industry.

Market 9: coated battery-grade graphite powder. Coating processes are proprietary and application-specific, and the details are not disclosed including whether coating is always required.

At this stage it is unclear whether any of the by-products from synthetic and natural graphite result in any product substitution on the market. In order to provide a robust framework for any new technologies which may be developed in the future, the scope should consider such by-products and corresponding avoided processes.

6.2 LCIA IMPACT CATEGORIES

Currently available assessments of synthetic and natural graphite production are mostly limited to global warming potential (GWP, as CO2-equivalent emissions) and a broader suite of impact categories must be considered. Preliminary analysis of the processes involved in the manufacturing chain reveal that a complete inventory is likely to include substances contributing to all commonly assessed categories (eg those included in ReCiPe or CML assessment methods) either directly or indirectly, as follows:

- Acidification. Relevant substances emitted during purification (acidic emissions to soil and waterways) and petroleum processing (sulphur and nitrogen oxides emissions to air and water).

- Eutrophication. Relevant substances emitted as result of petroleum processing (phosphorus and nitrogen emissions to air and water).

- Fine particulate matter formation. Relevant substances emitted throughout process chain.

- Raw material extraction contributes to fossil resource scarcity.

- Global warming. Relevant substances emitted throughout process chain.

- Ionizing radiation. Unlikely to contribute directly; however, this category has to be included if nuclear power is used as source of electricity.

- Land use. During raw material extraction and for the establishment of manufacturing facilities.

- Raw material extraction contributes to mineral resource scarcity.

- Photochemical ozone formation. Relevant substances emitted throughout process chain and through the use of heavy machinery.

- Stratospheric ozone depletion. Unlikely to contribute directly; however, this category has to be included to account for background processes.

- Toxicity. Relevant substances emitted during petroleum processing and during purification as a result of using chemicals.

- Water consumption. Significant amount of water is used during natural graphite beneficiation. This category has to be included if hydropower is used as source of electricity.

A screening study is required to confirm which of these categories are most applicable and which assessment methods are most appropriate.

7. CONCLUSIONS

Lack of quality data for the graphite production chain is frequently highlighted in the examined existing LCAs for battery-grade graphite. In the current state of the market, development of PCR for all raw materials including graphite has become critical, particularly considering the new regulations proposed for batteries by the European Commission which will require carbon footprinting, circular economy measures and improved supply chain transparency (European Battery Alliance, 2020). The supply chain is undergoing major changes with centralisation and vertical integration; these changes will inevitably lead to profound impacts on the sustainability.

Graphite is supplied to a multitude of markets, each demanding different grades, and this supply can be achieved via both natural and synthetic graphite, each with distinctly different production methods. In this work, a framework is proposed for comparison of similar quality products on a life cycle basis to inform future work and to aid the development of PCR. During detailed examination, it was found that the existing LCA studies are based on incomplete inventories for battery-grade graphite. However scarce, information exists in the public domain and in major LCA databases to ensure no omissions or misrepresentations of the production chain.

Comparison of two underpinning existing LCAs for natural graphite demonstrate wide variation in results (e.g. 2.2-5.3 kg CO_2eq./kg) and indicate that the true CO2 emissions and process energy can be more than double the values currently reported. No quality inventory exists for synthetic graphite; therefore, this study proposed a more complete preliminary inventory based on published research data. Similarly, to the case for natural graphite, this new inventory suggests that GHG emissions and energy consumption for

synthetic graphite may be more than double the values reported in prior work, with total impacts of approximately 20.6 kg CO_2eq./kg and 45.9 MJ/kg, respectively.

Finally, a major gap in existing literature is that the current assessments are largely limited to GWP and process energy; however, both production chains result in a variety of emissions to air, land and water, contributing to a wide range of potential environmental stressors. Therefore, a broader range of indicators is applicable and must be considered in future LCAs and when PCR are developed.

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