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Energy, greenhouse gas, and water life cycle analysis of synthetic graphite anode production in the United States

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This study presents a comprehensive life cycle analysis of potential synthetic graphite battery anode material (BAM) production in the U.S. based on industrial-scale data. The analysis focuses on three impacts: greenhouse gas (GHG) emissions, total energy use, and water consumption. We also conducted sensitivity analyses to evaluate the effect of variation in process parameters and energy sources used for synthetic graphite BAM production on its life cycle GHG emissions. A detailed supply chain analysis of graphite BAM in the U.S. was also undertaken, along with a study of its associated GHG emissions. The results show GHG emissions of 29.7 kg CO_2-eq. per kg BAM, total energy use of 580 MJ kg^{-1} BAM, and water consumption of 121 L kg^{-1} BAM for the baseline condition. The graphitization step is a major process hotspot, contributing to over 74% of all impacts. This is attributed to the energy and material input requirements for this step, particularly through the use of crucibles. Across the entire synthetic graphite production process, electricity is the primary contributor, followed by crucibles used in graphite block production, and then calcined petroleum coke. Sensitivity analyses indicate that improvement in micronization yield, reuse of crucibles, and use of low-carbon nuclear energy can significantly reduce GHG emissions of potential domestic graphite production (by ~70%). Supply chain analysis identified major graphite BAM sources in the U.S. and showed that the U.S. has a competitive advantage in domestic production of synthetic graphite BAM in terms of reduced life cycle GHG emissions compared to present-day imported sources (by ~20%).

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Environmental significance

This study addresses the impact of U.S. production of synthetic graphite (SG) battery anode material (BAM) from an environmental perspective. With growing demand for lithium-ion batteries (LIBs), there is a need for a consistent supply of graphite, a key component in LIB anodes. The availability of raw material (petroleum coke) in the U.S. offers a consistent and reliable source for domestic production of SG BAM. This research provides a life cycle analysis of prospective domestic SG BAM production, identifying major process hotspots in greenhouse gas emissions, energy use, and water consumption. The influence of processing variables in reducing environmental impacts through sensitivity analyses is also explored.

1 Introduction

The transportation sector is the largest contributor to the United States' total greenhouse gas (GHG) emissions (~30% share in 2022), with light-duty vehicles accounting for over half of this share.[1] These emissions are mostly related to fossil fuel (primarily gasoline) combustion during vehicle operation and can be reduced through the use of alternative powertrain-based vehicles, such as electric vehicles (EVs). The market share of EVs in U.S. light-duty vehicle sales has increased over the past decade.[2,3] These vehicles are mainly powered by lithium-ion batteries (LIBs) that are charged from the electrical grid. However, most LIB constituent materials are imported to the U.S.[4] and are considered critical for a variety of reasons, including their necessity for use in both LIBs and other end-user technologies meant for non-vehicle applications.[5]

Graphite is one such critical material – it is used in LIB anodes[6] and comprises ~15–25% of LIB pack weight, depending on the cathode chemistry employed.[7] Although other alternative anode materials have been explored, graphite is the most widely used commercial LIB anode due to its demonstrated cycling stability and electrical conductivity.[8] Graphite anodes are made using either natural or synthetic graphite. Natural graphite (NG) occurs mostly as a flaky mineral deposit in rocks that are further processed into high-purity graphite[9] while synthetic graphite (SG) is produced from calcined petroleum coke *via* high-temperature processing.[10] Though NG has lower

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production costs, SG is preferred for battery anode material (BAM) applications due to its better thermal stability, higher quality and purity, and lower coefficient of expansion.[11] Currently, the U.S. does not produce NG[12] and imports ~35% of its total SG demand for all applications.[13] The growing demand for EVs and an increasing focus on critical material production within the U.S. is expected to result in a considerable expansion of synthetic graphite BAM production within the country,[14,15] especially given the domestic availability of feedstocks (petroleum coke) for its production. Such production is an important step from the standpoint of national energy security, as it can help the U.S. become self-reliant in meeting its graphite demand through a robust, in-house supply chain. However, graphite BAM production is highly energy-intensive, with its constituent processes (mainly baking and graphitization) consuming large amounts of thermal and electrical energy.[16] Hence, it is also important to understand the environmental impacts of producing graphite (as SG) in the U.S., as such an analysis can help to provide insights that enable the sustainable production of these materials.

Life cycle analysis (LCA) is a widely used tool to evaluate the environmental impacts of different materials and technologies, including various LIB-related critical materials.[17–19] Yet even though the production of SG anode *via* the Acheson process[20] dates back to the late 1800s, with several process improvements and modifications over the years,[21] there are only a few LCA studies in the literature that account for the environmental impacts of graphite anode production. Some investigations have provided primary life cycle inventory (LCI) data of graphite BAM production (NG and/or SG) and conducted its LCA.[16,22,23] The initial LCA studies[24–26] have simplified the graphite production process, often through using surrogate processes from other sectors (*e.g.*, carbon anode baking process used in the aluminum industry) for impact estimations. However, these initial studies do not consider the additional processing steps necessary to achieve the high purity levels for graphite BAM, meaning that their impact results underestimate the life cycle impacts of SG BAM production. Dunn *et al.*[22] provide comprehensive LCI data for SG BAM production, combining the baking step with an updated LCI for the highly energy-intensive graphitization step. Also, while the typical feedstock for SG BAM production is green petroleum coke that is subsequently calcined,[16] Dunn *et al.*[22] consider using calcined petroleum coke as the initial feedstock. This means that, like prior LCA studies on this topic, Dunn *et al.*[22] underestimate the material and energy inputs for SG BAM production, and thereby, its life cycle impacts. Iyer and Kelly[27] updated the LCI of SG BAM production by considering green petroleum coke as the initial feedstock that undergoes the calcination step before the baking process. Surovtseva *et al.*[16] have identified data quality gaps in previously developed LCIs for graphite BAM production. They use data from multiple literature sources to conduct the LCA of SG BAM production and report life cycle greenhouse gas (GHG) emission and energy consumption impacts of 13.8 kg CO_2-eq. per kg graphite and 45.9 MJ kg^{-1} graphite, respectively – with both results being greater than those reported in prior studies. In a recent study by Carrère *et al.*,[28] the authors have suggested that prior LCA studies considered the Acheson block production route for SG BAM production, while the current industry standard is the Acheson powder production route. They indicate that the impacts of graphite crucible use, electricity consumption during graphitization, the micronization process yield, and other process-related emissions are significantly underestimated in previous LCA studies on this topic. The study[28] provides detailed LCI data from an industrial process and reports life cycle GHG emissions of 42.2 kg CO_2-eq. per kg graphite, which is the highest impact among all studies published to date. Overall, the life cycle GHG emissions for SG BAM production vary between 5.0 and 42.0 kg CO_2-eq. per kg graphite in the reviewed literature, with this large variation stemming mainly from data quality and process-level factors. A recent study by Wang *et al.*[29] reported detailed processing steps, precursor feedstocks, and electricity consumption for Chinese SG production across 12 operational and 22 upcoming plants, as well as an example LCI of SG production using data from these plants. They report plant-specific carbon footprint to vary between 6.8 and 12.9 kg CO_2/kg SG, with an average footprint of 9 kg CO_2/kg SG.

Although a detailed analysis of SG BAM production using state-of-the-art industrial-scale production processes is available in the literature,[28] a similar study with a U.S.-based focus that investigates the impacts of SG BAM production as a function of supply chain and/or production location has not been undertaken to date. Here, we seek to address both of these gaps. The main objective of this study is to conduct an LCA of SG BAM production in the U.S. based on current industrial-scale data, focusing on three life cycle impacts: GHG emissions, energy use, and water consumption. Additionally, we analyze the United States' graphite supply chain to understand how both domestic and imported graphite affect the life cycle GHG emissions of graphite anodes used in EVs in the U.S. Different production scenarios and sensitivity analyses are performed to understand the influence of specific production parameters on the GHG emissions of graphite BAM production. The novelty of this study is that we examined the US context for synthetic graphite BAM production and considered manufacturing variability issues. The results from this study can be used to inform policymakers, BAM manufacturers, and future studies focused on SG production and applications about the challenges and opportunities in its manufacturing from an environmental impact perspective.

2 Life cycle analysis (LCA) methodology

2.1 Goal and scope

This study has two goals: (a) to conduct an LCA of SG BAM production to determine the hotspots in its life cycle environmental impacts (GHG emissions, energy use, and water consumption), and (b) to determine the life cycle GHG emission impacts of graphite supply chains in the U.S. For the first goal, the system boundary for SG BAM production includes a cradle-to-gate analysis, starting with the processing of calcined

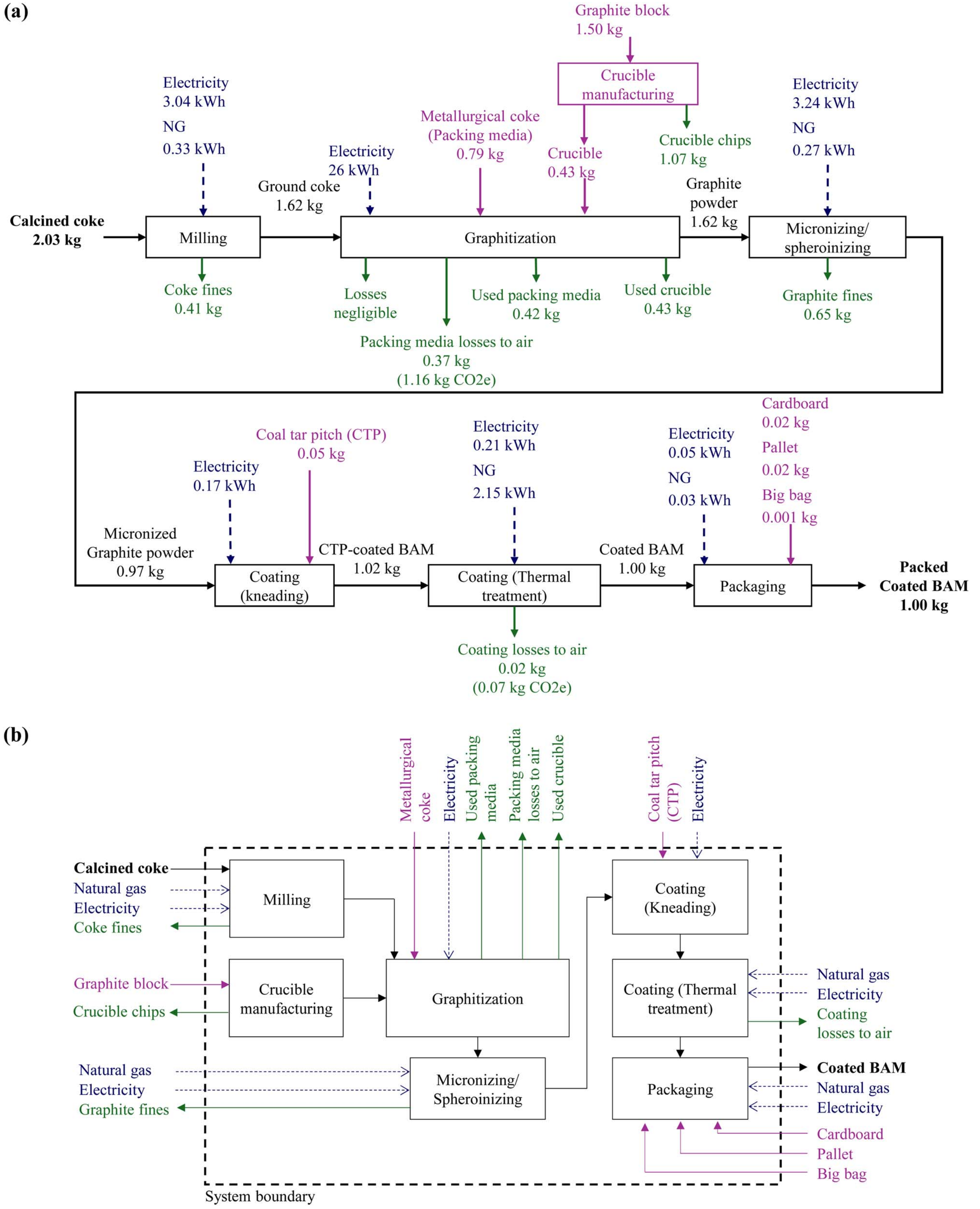

Fig. 1 (a) Process flow diagram of SG BAM production, with values adapted Carrère *et al.*[28] (refer to Table S3 in the SI for details). (b) System boundary used for the analysis of GHG emissions and fossil energy and water consumption. The blue, magenta, and green fonts indicate energy sources, material sources, and material outputs, respectively.

petroleum coke and finishing with the production of packed coated BAM. The functional unit used in this study is 1 kg of SG BAM meant for use in LIBs.

We have used Argonne National Laboratory (ANL)'s Research and Development Greenhouse Gases, Regulated Emissions, and Energy Use in Technologies 2023 model[30] (hereafter referred to as "R&D GREET") to perform the life cycle impact analysis of SG BAM production in this work, using the LCI data shown in Fig. 1(a) and Table S3 of the SI. R&D GREET is a widely used tool to determine the life cycle environmental impacts of fuels, materials, and technologies. It provides these impacts on various categories, including GHG emissions, nitrous oxide (NO_X) emissions, sulfur oxide (SO_X) emissions, particulate matter (PM_{10} and $PM_{2.5}$) emissions, and water consumption. In this study, we focus solely on three impacts: GHG emissions, energy use, and water consumption. All the impact burdens of the production phase are allocated entirely to SG BAM production, as all other outputs are considered residues. Based on our discussion with Tokai COBEX[31] – a commercial graphite producer, these residues are either emitted to the air or consist of solid waste that cannot be recycled back or recovered into the process.

2.2 System boundary and process description

Fig. 1(b) shows the system boundary of SG BAM production for this LCA study. SG BAM production starts with the processing of calcined petroleum coke. Calcined coke is generally prepared by heat treatment of green petroleum coke above 1000 °C to remove moisture, volatile matter, and impurities.[32] Detailed information on the material and energy requirements for the production of calcined petroleum coke are provided in Table S2 of the SI. Calcined petroleum coke is milled to a fine powder and then sent for graphitization. In the graphitization process, ground coke is loaded into graphite crucibles and covered by a packing media, usually metallurgical coke. Packing media are electrically conductive and facilitate the transfer of electricity around crucibles for heating. During graphitization, some of the packing media material can oxidize, so makeup metallurgical coke is needed for each new batch. The crucibles loaded with ground coke are sent into an Acheson electrical furnace. The furnace is heated *via* electricity to ∼3000 °C and held at that temperature for ∼2–3 weeks to obtain graphite powder.[28] Graphite powder is further micronized to reduce the size to the micrometer range, with the process involving ∼40% loss of graphite as fine powder. The remaining micronized graphite powder is coated with coal tar pitch (CTP), such that CTP accounts for 5–10% of the mixture's weight. CTP acts as a binder and helps improve electrochemical properties. CTP is traditionally used as a binder in roofing tiles, asphalt pavements, and in aluminum industry for electrode manufacturing.[33,34] Once coated, the mixture is heat-treated at ∼1000 °C to remove impurities and volatiles. The resultant coated SG BAM is sent for packaging.

Detailed system input and output flows of mass, energy, and emissions, normalized for the production of 1 kg of SG BAM, are also shown in Fig. 1(a) and also in Table S3 of the SI. Data for these flows was adapted based on details from Carrere *et al.*[28] and communications with Tokai COBEX,[31] who sourced the material and energy flows from production facilities in France and Japan, with guidance from industry experts. Process parameters, such as the number of uses of a crucible (during graphitization) and the yields of intermediate processes, were discussed with experts. Carrere *et al.*[28] assume that graphitization crucibles are used only once and then disposed of. However, there could be possibilities of using the same crucible several times. Nevertheless, our baseline estimations assume a single use per crucible and a micronization yield of 60%, as reported by Carrere *et al.*[28] Additional estimations, which explore variations in these parameters (*i.e.*, the use of crucibles during graphitization and changes in micronization yield), are detailed in Section 3.4.1 in the sensitivity analysis. The baseline inventory for all the material and energy inputs used in SG BAM production is based on R&D GREET.[30] While the LCI for SG BAM production in this study is based on details from a facility in France, to assess the environmental impacts of its production in the U.S., we adjusted the GHG footprint of the electricity used in various processes to that for the U.S. average electricity grid mix (0.4 kg CO_2-eq. per kWh).[30]

Fig. 2 shows the material and energy flows for the production of a graphite block – the feedstock to crucible production in Fig. 1(a). The inventory for graphite block production was provided by Tokai COBEX.[31] Briefly, calcined petroleum coke is used as the feedstock, which is mixed with CTP and metallurgical coke (packing media). The mixture is baked and graphitized at high temperature to obtain a graphitized block. The obtained graphite blocks are then machined to manufacture crucibles (Fig. 1(a)), with ∼71% of the block lost as machined waste. It is important to note that the basis for the process flow diagram in Fig. 2 is of 1 kg of graphitized block, which differs from the basis used in Fig. 1(a), which is of 1 kg of packed coated BAM.

2.3 Non-combustion emissions from SG production

During the graphitization and thermal coating processes, a fraction of the carbon in metallurgical coke and CTP vaporizes as CO_2, resulting in non-combustion process CO_2 emissions. According to Tokai COBEX,[31] 47% of the metallurgical coke input in the graphitization process vaporizes and is released into the atmosphere. Baking processes include post-burning treatments to reduce carbon compounds such as CH_4 to CO_2. Even though the exhaust gas can contain traces of VOCs and other carbon compounds, due to a lack of precise measurements, all emissions are assumed to be CO_2. Given that metallurgical coke contains 85% carbon, and considering the stoichiometric conversion of carbon to CO_2, it is estimated that approximately 1.2 kg CO_2-eq. per kg of SG BAM is generated. In the coating process, volatile compounds comprise 45 wt% of CTP (35% is volatile carbon and the remaining 10% comprises hydrogen, oxygen, and other compounds). These volatile compounds are lost to the atmosphere during the thermal coating process and generate non-combustion emissions. Based on the volatile carbon content and stoichiometry of these

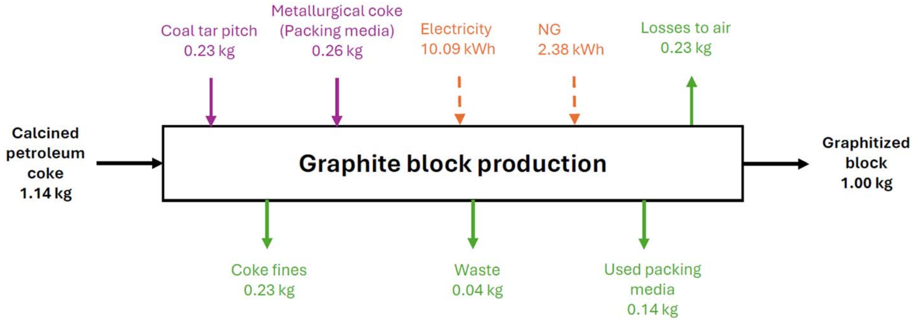

Fig. 2 Process flow diagram of graphite block production.

compounds, ~0.1 kg CO_2-eq. per kg BAM is released. These non-combustion emissions from graphitization and thermal coating processes are accounted for in our life cycle GHG emission results.

2.4 Sensitivity analysis

Due to some expected variabilities in processes such as micronization, and crucible production and use, sensitivity analyses are conducted to understand the influence of these variabilities on the overall life cycle GHG emissions of SG BAM production. In the baseline scenario, 71% of the graphite block utilized in crucible production is lost in the form of chips.[28] However, it is reasonable to consider that future optimization of crucible production might identify opportunities to reduce graphite block material loss. The first sensitivity case models crucible losses from 20% to 71% at intervals of 10%. For the micronization step, the process has a 60% yield in the base case scenario.[28] However, if improvements in the micronization process are achieved, the loss of graphite powder as fines can potentially be reduced. Therefore, a second sensitivity analysis evaluates the effect of changes in the micronization yield from 60% to 90% at intervals of 10%. A third sensitivity case investigates the synergies between micronization yield and crucible uses. Micronization yield is varied between 60% and 90% at intervals of 5%, and the number of uses of a single crucible is varied between one and eight at intervals of one use.

One of the potential benefits of using LIBs as an energy source for vehicle mobility is the possibility of using low-GHG electricity sources for their production and charging, thus enabling reductions in the life cycle GHG emissions of the concerned vehicle. Low-GHG electricity can also be used for producing different LIB constituents, including SG BAM. For the baseline scenario, the average U.S. electricity grid mix is considered as the electricity source. We also consider alternative scenarios where this grid electricity usage across different SG BAM production stages is replaced by either nuclear energy or average Chinese grid electricity. Nuclear energy is considered among the lowest GHG-intensive electricity sources, while China is studied here as it currently dominates the global production of SG BAM.

2.5 Supply chain analysis of graphite used in the United States

In addition to our primary goal (LCA of SG BAM production in the United States), we also conducted the supply chain analysis of graphite used in EV battery anodes to understand the life cycle impacts of graphite resources available in the U.S. The analysis considers 2022 as the reference year for impact estimations, using data from previously published studies.[4,35] The total graphite demand for EV LIBs in the U.S. is separated into that for domestically produced and imported EV LIBs, with the graphite for both sources split by the type of graphite used - SG and NG. Domestic EV LIBs based on SG BAM are further split into domestically produced SG and imported SG. The major source countries for imported SG are also identified, based on U.S. trade data.[36]

In 2022, the United States imported 131.3 thousand tonnes (kt) of EV LIBs.[37] Based on an average graphite content of 0.3 kg per kg of battery,[38] it is estimated that 34.8 kt of graphite was sourced for imported LIBs. Domestic EV battery production is estimated at 50 GWh in 2022.[14] Considering an average graphite content of 1.2 kg per GWh of battery cells,[39] it is estimated that 51.6 kt of graphite was used in domestic EV battery production. Therefore, the total graphite usage in the United States in 2022 is estimated at 86.4 kt. This amount supports an EV battery demand of 72.0 GWh, which is only 2% higher than the 70.6 GWh value reported by the International Energy Agency.[40]

Recent statistics indicate that on an overall basis, ~43% of the graphite used in global LIB anodes is NG, while the remaining share (57%) is SG.[41] Applying these shares yields a total usage of 15.0 kt of NG and 19.8 kt of SG in the United States, respectively, from imported EV LIB anodes. Additionally, 22.2 kt of NG and 29.4 kt of SG are used for domestically produced EV LIB anodes.

Estimates for SG production in the U.S. are obtained from Gohlke *et al.*,[14] who have determined that ~5 GWh of total anode material is produced domestically, of which 86% corresponds to graphite anodes. Each GWh contains ~1.0 kt of graphite,[42] resulting in 4.5 kt of domestically produced graphite. This graphite is synthetic, as the United States does not currently produce natural graphite.[12]

For domestic batteries, China dominates the SG imports to the U.S. (~50% share), followed by Mexico (11%), Spain (9%),

Table 1 GHG emissions per kWh for different countries

Grid source	GHG emission (g CO_2-eq. per kWh)
USA	440
Canada	124
China	700
Japan	602
Mexico	465
Madagascar	1019
Spain	360
RoW	549
Nuclear	3

Table 2 Supply chain share of natural and synthetic graphite in the U.S

Graphite source	Percentage share
Natural imported	43.00%
China	46.69%
Mexico	10.75%
Canada	10.15%
Madagascar	10.00%
Other	22.40%
Synthetic imported	51.85%
China	58.19%
Mexico	6.19%
Spain	6.98%
Japan	4.95%
Other	23.70%
Synthetic domestic	5.15%

and Japan (~6%), with the remaining share supplied by a wide range of nations.[36] Per USGS,[43] NG is supplied to the United States mainly by China (33% share of supply), Mexico (18%), Canada (17%), Madagascar (10%), and the residual share from other countries.

The distribution of NG and SG used in EV LIBs imported to the U.S. is based on global statistics reported by Benchmark Mineral Intelligence.[44] These statistics indicate that ~67% of the global NG supply used in LIB anodes is sourced from China, with notable contributions from Madagascar (10%), Brazil (7%), Mozambique (6%), and other nations (10% total), respectively. In the case of SG, China again dominates its supply share (69%), followed by India (8%), Spain (4%), and Japan (4%), with the remaining share coming from other countries.

After constructing the supply chain for graphite BAM to the United States, we calculated the weighted life cycle impact of BAM supply to the U.S. As part of this exercise, we also compare the life cycle GHG emissions of graphite production by type (NG and SG) from different sources (U.S. domestic and imported). We use R&D GREET model for this analysis. To account for regional differences in the life cycle impacts of graphite BAM production, we consider the GHG intensity of the electric grid mix across different locations from which the U.S. sources BAM supply. While specific electric grid mixes are used for individual BAM production locations (*e.g.*, China), for "other" countries, we use the average global electric grid mix as representing the electricity used for BAM production. GHG emissions per kWh for different electricity grids used in this study are shown in Table 1 based on R&D GREET.[30] The supply chain share of natural and synthetic graphite in the U.S. is given in Table 2.

3 Results and discussion

3.1 Greenhouse gas emissions of synthetic graphite BAM production

A cradle-to-gate LCA of SG BAM production *via* the Acheson powder route in the U.S., considering baseline conditions, shows the overall life cycle GHG emission to be 29.7 kg CO_2-eq. per kg BAM (Fig. 3). The graphitization stage is the major hotspot (Fig. 3(a)), contributing ~80% of the life cycle GHG emissions of SG BAM production. The graphitization process is both energy- and material-intensive. From the energy perspective, high-temperature processing (at ~3000 °C) for 2–3 weeks is needed during this stage, using electricity for heating. From the material perspective, the single-use lifetime of the crucible,

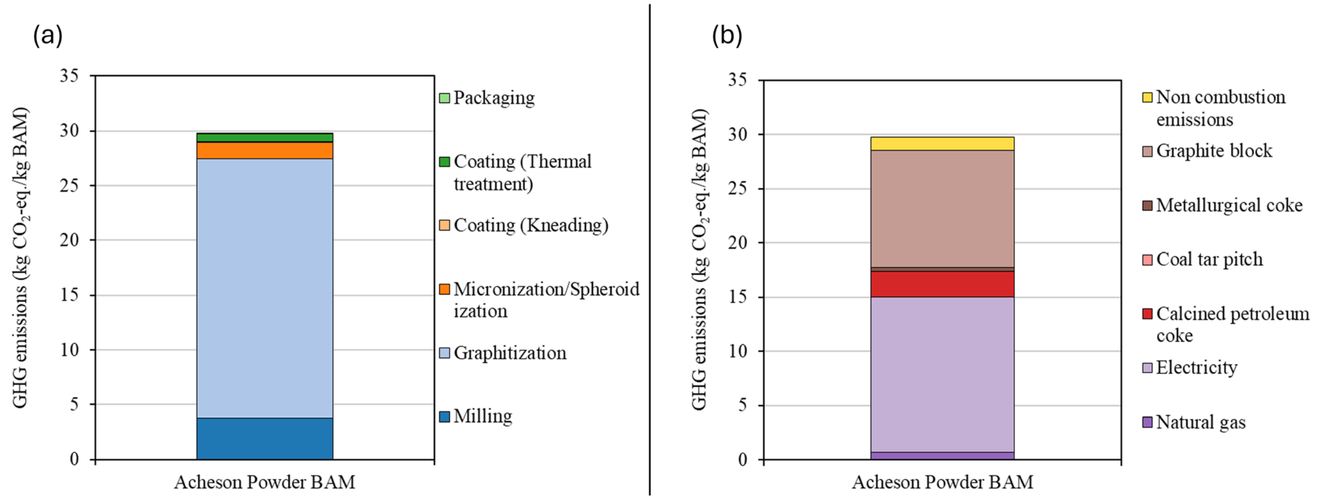

Fig. 3 Greenhouse gas emissions breakdown of synthetic graphite BAM production: (a) per process stage, (b) per input/output flow.

considerable material losses associated with crucible production from graphite block, and the use of metallurgical coke as packing media that generates non-combustion CO_2 emissions, explain the significant role of graphitization as an environmental hotspot. The milling process has the second-highest impact contribution after graphitization among all the processing stages, contributing ∼13% to total GHG emissions. The majority of this contribution comes from calcined coke feedstock, followed by the electricity used during the size reduction of calcined petroleum coke.

In terms of contributions from different material and energy inputs and emission outputs, electricity is the major contributor to the life cycle GHG emissions of SG BAM production (∼48% share) (Fig. 3(b)). Around 80% of the total electricity consumption for SG BAM production occurs during the graphitization process. The graphite block is the second largest contributor to GHG emissions of SG BAM production (>35% share; Fig. 3(b)). Graphite blocks are machined into crucibles, which are then used to encase calcined coke during the graphitization process. Graphite blocks are also made of calcined petroleum coke and require high energy during processing. Furthermore, over 70% of the weight of the initial graphite block is lost as machined chips during crucible manufacturing. Since crucible chips have lower economic value than BAM,[28] we have allocated 100% of the burden of graphite block and crucible production to SG BAM. Crucible chips are generally not reused for manufacturing due to concerns about impurities and non-uniform crystalline structure that can impact their performance.[45] In our base case, crucibles are used only once. If they can be reused several times before being discarded as suggested by some manufacturers,[46] this could reduce their environmental impact. Crucibles may get oxidized during high-temperature processing after multiple uses, which can prevent them from being reused for graphitization. Within graphite block production, both the petroleum coke feedstock and electricity are the major hotspots. Non-combustion emissions come from packing media losses to air during graphitization and coating losses to air during the coating process.

The overall trends in our results are in line with prior literature. For instance, Carrère *et al.*[28] report graphitization as the major hotspot in the SG BAM production life cycle. In their study, they identified energy as the major contributor, followed by crucibles. The GHG emissions value in their study is higher than this study, which may be due to the differences in their assumed material and energy sources. The carbon footprint value reported by Wang *et al.* is lower than this study.[29] Their lower footprint is due to comparatively less energy consumption for manufacturing graphite crucibles, less electricity consumption for the graphitization process, and higher conversion efficiencies for the micronization/spheroidization process. However, Wang *et al.* do not include a complete LCI for any single SG production plant, possibly due to the proprietary nature of their data.[29] Previous studies[16,22,27] do not consider the use of graphite block and packing media in their analysis, nor do they consider post-processing of graphitized products *via* micronization, coating, and packaging processes. Because of these reasons, their results underestimate the life cycle GHG emissions and energy consumption. For example, Surovtseva *et al.*[16] reported GHG emission to be 13.8 kg CO_2-eq. per kg graphite BAM, which is ∼54% less than the base case value reported in this study.

Life cycle GHG emissions for NG BAM production range between 5-10 kg CO_2-eq. per kg graphite BAM[23,30,47] and are much lower than those for SG BAM production. However, SG BAM is a preferred choice for EV applications over NG BAM due to its superior properties, such as longer cycle life, higher purity, and consistent quality.[11] The largest opportunity for U.S. domestic production would be SG BAM, since the U.S. currently does not produce NG but has leverage in SG production capabilities due to the domestic availability of raw material (green petroleum coke) for its production. Other alternatives for SG BAM manufacturing processes, such as electrochemical methods, are also considered in the literature.[48] The life cycle GHG emissions of SG BAM production through such an alternative method are reported to be 0.7 kg CO_2-eq. per kg BAM,[48] which is significantly lower than other studies. However, such alternative methods are still in their infancy and may not be comparable to existing commercial processes. Apart from graphite BAM production from virgin materials, some studies have also looked at the environmental impacts of recycling

Table 3 Comparison of life cycle GHG emissions between different studies

S.No.	System boundary	Functional unit	GHG emissions (kg CO_2-eq. per kg)	Reference
1	Final stage up to coated and packed BAM	1 kg SG BAM	30.48	This study
2	Final stage up to post treatment of SG (does not include coating and packing into BAM)	1 kg SG	6.8 to 12.9	Wang *et al.*[29]
3	Final stage up to coated and packed BAM	1 kg SG BAM	42.2	Carrere *et al.*[28]
4	Final stage up to the graphitization step	1 kg SG	13.8	Surovtseva *et al.*[16]
5	Final stage up to the graphitization step	1 kg SG	8.76	GREET 2023 (ref. 30)
6	Final stage up to coated BAM	1 kg NG BAM	9.62	Engels *et al.*[23]
7	Final stage up to coated BAM	1 kg NG BAM	6.39	GREET 2023 (ref. 30)
8	Final stage up to post processed BAM	1 kg NG BAM	5.32	Gao *et al.*[47]
9	Final stage up to graphitization step	1 kg SG	0.7	Kulkarni *et al.*[48]
10	Recycling of graphite from spent LIB	1 kg graphite recovered	0.53–9.76	Rey *et al.*[49]
11	Recycling of graphite from spent LIB	1 kg graphite recovered	0.27–3.53	Natarajan *et al.*[50]

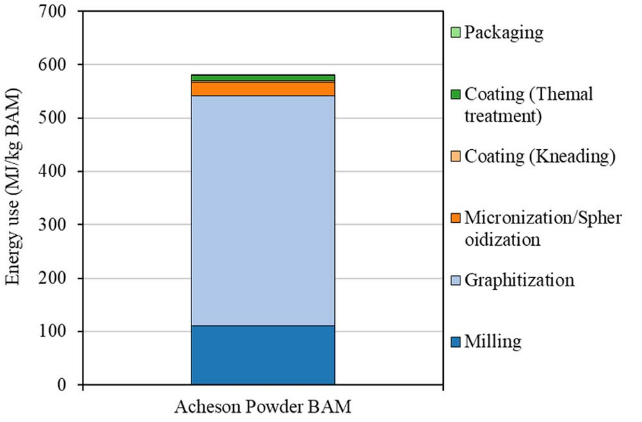

Fig. 4 Total energy demand breakdown of synthetic BAM production per process stage.

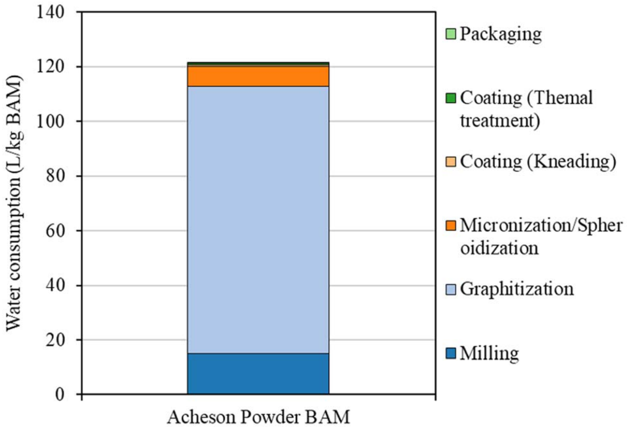

Fig. 5 Water consumption breakdown of synthetic BAM production per process stage.

graphite from spent LIB anodes through direct recycling of LIBs.[49,50] These studies report life cycle GHG emissions of 0.27–9.76 kg CO_2-eq. per kg graphite recovered. However, concerns remain about the quality of graphite recovered through such direct recycling due to contamination with metals, lithium salts, and binders.[51] Many other recycling methods, such as chemical and thermal, are being studied, but concerns still remain on the performance and stability of the recycled anode compared to that of virgin material.[52] Apart from the performance and quality issue, other challenges still remain in terms of recycling infrastructure setup, process scale-up, cost, and environmental concerns from the use of harsh chemicals.[52] Table 3 provides a summary of LCA studies on graphite production for anode applications.

3.2 Total energy use

Energy use is an important life cycle environmental impact metric as it helps to: (a) determine the cumulative total energy used in the upstream processing during manufacturing; and (b) identify potential areas to lower energy use and replace conventional energy sources with lower-GHG emitting energy sources to reduce the overall environmental impact. In this study, the life cycle energy usage is estimated to be 580 MJ kg^{-1} BAM produced (Fig. 4). We see a similar trend in the results for energy use as that seen for GHG emissions (Fig. 4). The graphitization stage requires the most energy (>70% share), followed by the milling stage (>18%) (Fig. 4). Within graphitization, over 50% of the required energy is for the graphite block, while electricity accounts for > 43% of its energy contributions. Within the graphite block, around 69% of its required energy is for graphitization. For the milling process, more than 78% of the energy is embedded in the calcined petroleum coke feedstock.

An emergy-based study on SG BAM production also suggests that electricity is one of the major energy inputs.[53] In that study, the electricity required in graphitization and petroleum coke feedstock had similar emergy contributions (~28% each). Emergy refers to the total direct and indirect energy spent to make a product or service.[54] Emergy accounts for both nature-based (such as sunlight) and society-made transformations, such as electricity.[55] Therefore, while the emergy-based values are not directly comparable to the values used in our study, they do suggest similar conclusions as our analysis.[53] Newer methods to reduce graphitization temperature and lower energy use with catalytic graphitization processes are also being studied in the literature.[56] However, they are not yet commercialized.

3.3 Water consumption

Water is another essential natural resource critical for the manufacturing sector. Understanding water consumption throughout the life cycle of a product can provide valuable information on project planning, such as identifying locations for new manufacturing facilities. R&D GREET defines water consumption as the amount of make-up water added to processes to compensate for losses due to evaporation and leakage.[57] Therefore, based on our analysis, the amount of make-up water used for producing 1 kg of SG BAM is 121 Liters (Fig. 5). Similar to energy use and GHG emission impacts, the graphitization stage is the major contributor (~80% share) to the life cycle water consumption of SG BAM production (Fig. 5). Of this, ~60% contribution is associated with electricity generation, while the remaining is mainly from the graphite block, primarily due to its use of electricity for block production. Thus, most of the water consumption impacts of SG BAM production are embedded in electricity generation.

3.4 Sensitivity analysis

3.4.1 Crucible losses, micronization yield, and crucible uses. The impact of micronization yield and crucible resource efficiency is also considered for the sensitivity analysis. Under real conditions, there might be opportunities to improve micronization yield, reduce the crucible losses during machining, and increase the number of times of use of the graphite crucibles during graphitization, all of which can improve the process efficiency and product yield, and at the

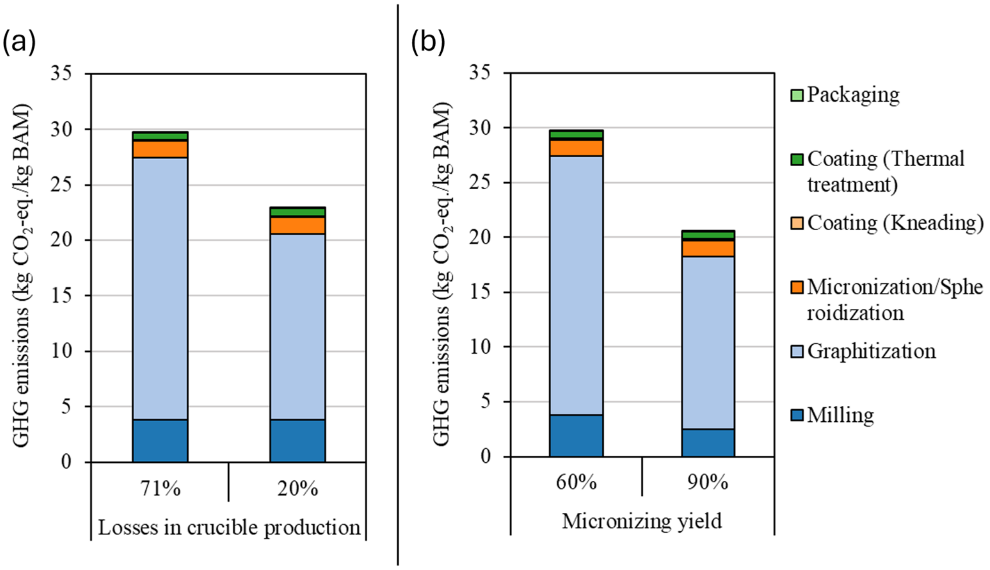

Fig. 6 Greenhouse gas emissions comparison for producing synthetic graphite BAM when varying: (a) the losses in the graphite crucible production, (b) the micronization yield of graphitized powder.

same time reduce environmental impacts. However, reducing crucible losses and increasing the number of crucible uses effectively target the same parameter: crucible resource efficiency. In this sensitivity analysis, crucible losses are varied between 20% and 71%, where 71% is the crucible loss in the base case, while micronization yields are varied between 60% and 90%, where 60% is used in the base case. The results of the sensitivity analyses for crucible machining losses and micronization yield are shown in Fig. 6(a and b), respectively. Reducing the machining loss during crucible production from 71% to 20% can reduce life cycle GHG emissions of SG BAM production by over 23%, while increasing the micronization yield from 60% to 90% can lower these GHG emissions by over 30% - all on per-kg SG BAM production.

The combined effect of varying the number of crucible uses and the micronization yield is also considered in the sensitivity analysis, as shown in Fig. 7. It shows that the life cycle GHG emissions of SG BAM production are highly sensitive to micronization yield at a lower number of crucible uses, as seen by the increasing change in contours with an increase in the micronization yield on the *y*-axis (Fig. 7). This indicates a greater opportunity to reduce GHG emissions. In contrast, the comparative advantage of reducing GHG emissions is lower for graphite crucible uses by more than three times (*i.e.*, when the crucible is used more than 4 times), as seen by the rate of change in contours/colors on both axes (Fig. 7). The combined effect of increasing micronization yield to 90% and using the same crucible eight times can reduce the life cycle GHG

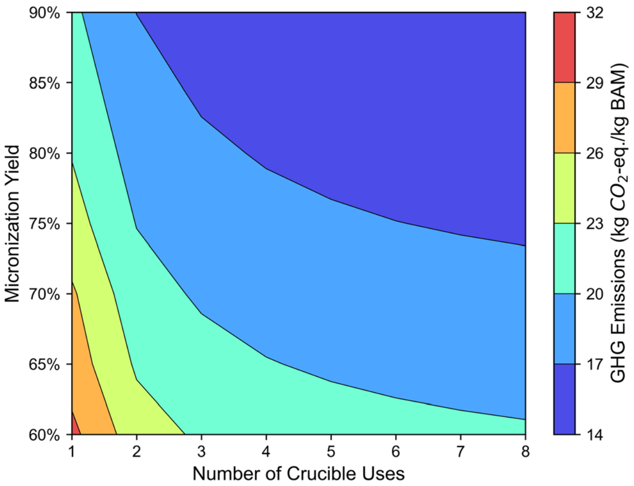

Fig. 7 Surface plot showing the combined effect of improving micronization yield and the number of crucible uses on GHG emissions.

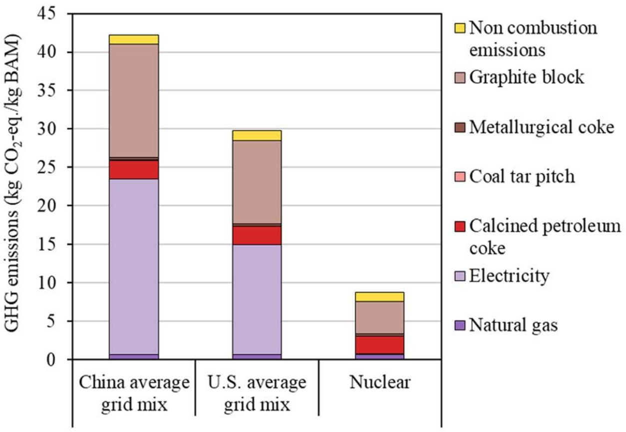

Fig. 8 Greenhouse gas emissions comparison for producing synthetic graphite BAM when using China grid electricity, U.S. average grid electricity, and nuclear energy.

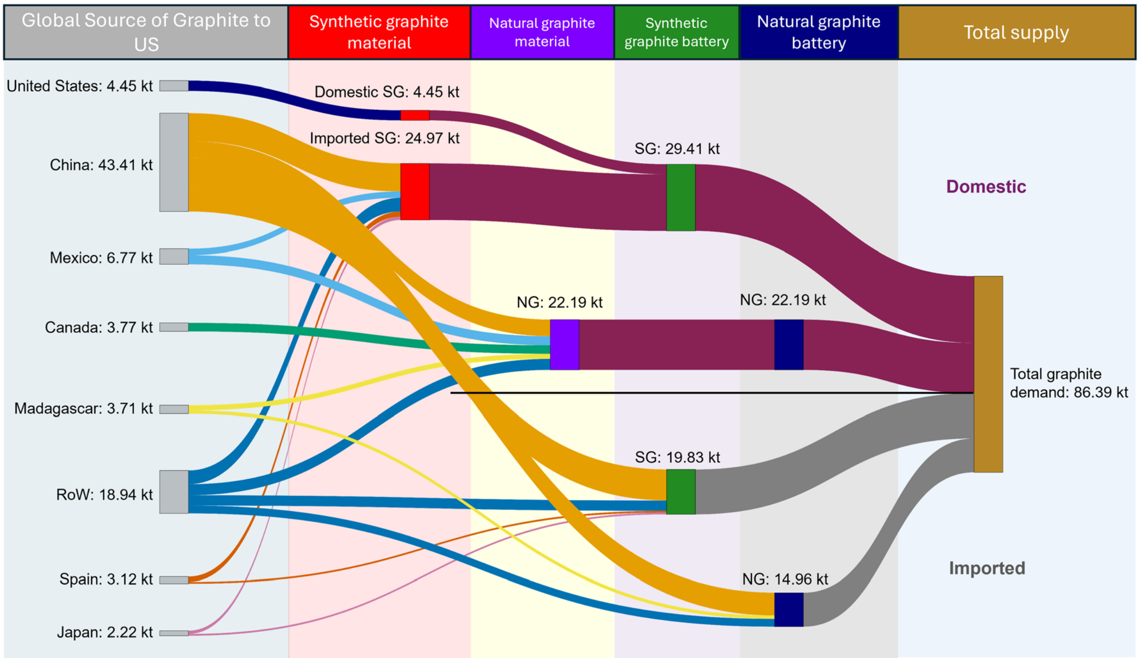

Fig. 9 Graphite supply chain contribution for U.S. EV batteries (NG refers to natural graphite and SG refers to synthetic graphite).

emissions by over half of that in the baseline scenario (14.3 kg CO_2-eq. per kg BAM). Note that using a crucible eight times is equivalent to a single-use crucible with a 9% loss factor during production.

3.4.2 Electricity source. To understand the impact of different energy sources on the life cycle GHG emissions of SG BAM production, we conducted scenario analysis based on three geographies: (a) U.S. electricity grid mix (as the base case); (b) China's electricity grid, as most of the global SG BAM production currently occurs in China[35] and we anticipate this production to have higher GHG emissions than the U.S., and (c) a 100% nuclear energy grid as a low GHG-emitting energy source. Life cycle GHG emissions of SG BAM production are higher in China (42.2 kg CO_2-eq. per kg BAM) than in the U.S. (by > 40%; Fig. 8). For comparison, the Chinese electricity grid is ~60% more GHG-intensive than the U.S. average grid mix[30] due to greater contributions from coal-based power plants. In contrast, nuclear energy has a very low GHG footprint and has a much lower GHG intensity than the U.S. average grid mix (by ~99%).[30] Hence, the use of Chinese grid electricity increases the GHG emission impacts from both the graphitization stage as well as graphite block production stage. When using low-GHG-emitting nuclear electricity, the life cycle GHG emissions of SG BAM production reduce by ~70% (8.8 kg CO_2-eq. per kg BAM) compared to the base case scenario with the U.S. average grid mix (Fig. 8). This shows that the U.S. has a domestic opportunity to produce SG BAM with lower environmental impacts that help establish a robust supply chain, even with the current use of grid electricity (base case). Further GHG emission reductions could be achieved by exclusively using nuclear electricity sources for SG BAM production, through co-locating with or contracting electricity from a nuclear power plant.

3.5 Supply chain analysis

To understand the impact of the graphite supply chain and potential environmental opportunities for U.S. domestic production, we also performed a supply chain analysis for SG BAM production. The total graphite BAM demand in the U.S. is ~86.4 kt. The U.S. currently does not produce any NG and is also heavily reliant on imported SG.[43] As seen in Fig. 9, most of the graphite used as BAM in the U.S. is imported. Around 5% of SG used in BAM is produced in the U.S., while the remaining 95% share is imported. In total, the U.S. graphite BAM supply comprises 57% SG and 43% NG.[41] China contributes more than

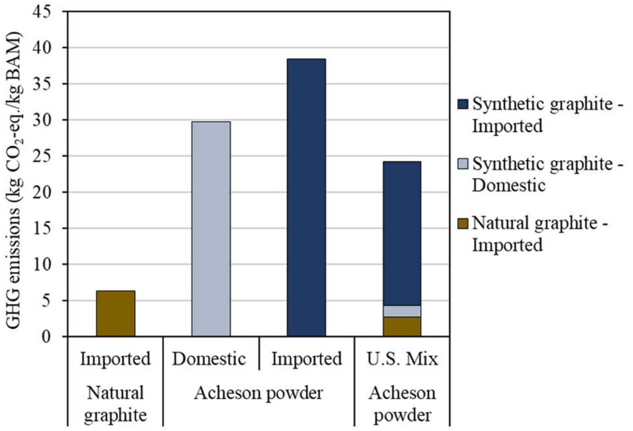

Fig. 10 Greenhouse gas emissions associated with different sources of graphite BAM. The marine blue and golden bars represent the weighted average contributions of all the global sources (see Fig. 9) to the supply of imported synthetic graphite and natural graphite, respectively.

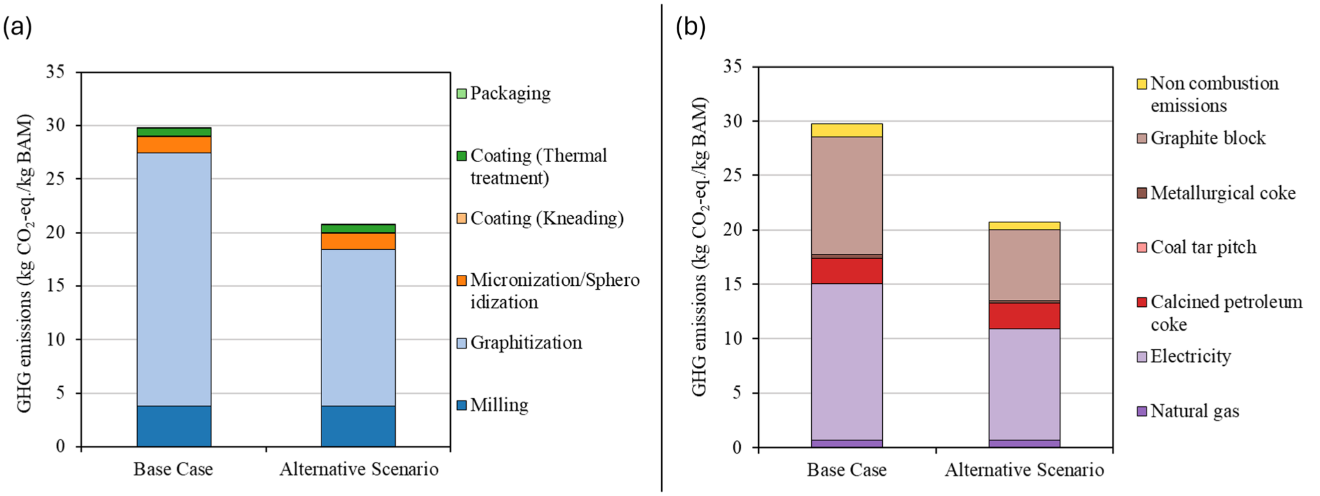

Fig. 11 Impact of switching graphitization and micronization/spheroidization processing steps on greenhouse gas emissions of SG BAM production (a) per process stage, (b) per input/output flow.

50% of the total BAM supply to the U.S., followed by Mexico (8%). Both SG and NG imports in the U.S. for BAM application are either imported as graphite material that is later processed for battery applications or directly as graphite BAM.

Based on the supply chain contributions from Fig. 9, we compared the environmental impacts of graphite BAM from different origins (Fig. 10). Imported NG has the lowest life cycle GHG emissions (6.4 kg CO_2-eq. per kg BAM), being ∼80% less than that of U.S. domestic SG production, while the life cycle GHG emissions of imported SG BAM are more than ∼20% of that for U.S.-based production (38.5 kg CO_2-eq. per kg BAM). Due to the important contributions of NG in the U.S. graphite supply chain and its associated GHG emissions, the average life cycle GHG emission impact of the U.S. average graphite BAM is 24.2 kg CO_2-eq. per kg BAM, which is ∼20% lower than that of the base case U.S. SG BAM production. Note that we assume here that all NG and SG BAM imports are processed outside of the U.S., even though that may not always be true, as seen in Fig. 9, because the data is not available at a sufficiently granular level to determine the level of treatment for such imports. Overall, the data suggests that there are opportunities to further reduce the environmental impacts of U.S. EV LIBs through U.S.-based production of SG BAM. Importing more natural graphite can reduce environmental impact. However, synthetic graphite is increasingly preferred as the anode material for EV applications.[11] The raw materials for producing synthetic graphite are domestically available in the US, while there is no domestic source for natural graphite. Therefore, there is more focus on domestic synthetic graphite production in the US to ensure its consistent supply.

3.6 Potential alternative scenario for process improvement

Graphitization is the greatest contributor to the environmental impact of SG BAM production. We considered a hypothetical scenario in which micronization/spheroidization is conducted prior to graphitization (see Fig. 1(a)). The energy required to micronize/spheroidize milled feedstock is assumed to be the same as the energy required to micronize/spheroidize the graphitized feedstock, since the material input is the same in both cases (assuming the same material yield). We also linearly interpolate the energy required to graphitize micronized/spheroidized feedstock from the energy required to graphitize milled feedstock. However, it should be noted that this scenario needs to be further studied experimentally to determine the impact of switching process order on the properties of the final product. A simplified LCA analysis shows that performing micronization/spheroidization before graphitization can significantly reduce (∼30%) GHG emissions (Fig. 11(a and b)). This is primarily due to a reduction in the amount of feedstock for the graphitization process, which reduces energy use and crucible requirements.

4 Conclusions

A life cycle analysis (LCA) of potential synthetic graphite (SG) battery anode material (BAM) production in the U.S. was performed using industrial-scale data for the Acheson powder production route. SG BAM production impacts were evaluated on three counts: greenhouse gas (GHG) emissions, energy use, and water consumption. Sensitivity analyses of SG BAM production were also performed, varying the micronization yield and crucible production efficiency, while different energy sources were also considered for background data in the LCA. We also examined the supply chain of graphite BAM in the U.S. and assessed its GHG emissions. The results show that U.S.-based SG production could be advantageous for LIB anode applications from the environmental viewpoint due to its lower life cycle environmental impacts (GHG emissions and energy use) than those of SG BAM production from imported sources. The analysis also highlights opportunities to further reduce these life cycle impacts by integrating graphite processing with low GHG-emitting energy sources, since energy (mostly electricity) is the major impact contributor. Our sensitivity analyses, considering improvements in micronization yield and crucible uses, show significant GHG emission reductions (by over 50%) over the baseline U.S.-based domestic production scenario. A supply chain analysis showed that the U.S. is heavily reliant on imported graphite. Domestically produced SG BAM has twin advantages of lower environmental impact and reduced risk of supply disruption compared to existing SG imports. Future studies could consider additional supply chain analyses, including raw material availability for graphite production in the U.S. There are also possibilities for process improvements by considering the impacts of switching the graphitization and micronization stages and catalytic graphitization of the feedstock to reduce overall energy consumption.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

The authors confirm that the data supporting the findings of this study are available within the article. Supplementary information: life cycle inventory data for calcined petroleum coke and synthetic graphite battery anode material production are provided in the SI. See DOI: https://doi.org/10.1039/d5va00171d.

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