Environmental Science & Technology

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Graphite Flows in the U.S.: Insights into a Key Ingredient of Energy Transition

Jinrui Zhang, Chao Liang, and Jennifer B. Dunn*

Cite This: *Environ. Sci. Technol.* 2023, 57, 3402−3414

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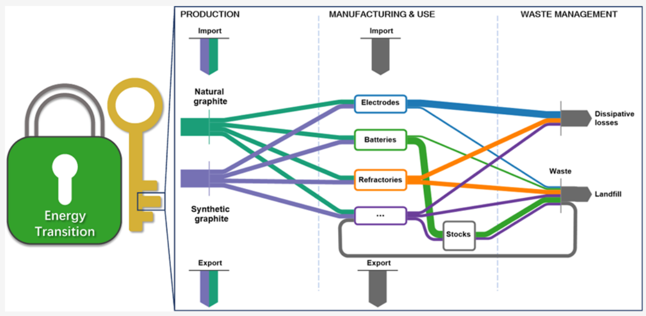
ABSTRACT: Demand for graphite will grow with expanding use of lithium-ion batteries in the United States. Much graphite is imported, raising supply chain risks. It is therefore imperative to characterize graphite's sources and sinks. Accordingly, we present the first material flow analysis for natural and synthetic graphite in the U.S. The analysis (for 2018) begins with processed graphite trade and includes graphite production, graphite product trade, manufacturing of end products, end product use, and waste management. It considers 11 end-use applications for graphite, two waste management stages, and three recycling pathways. In 2018, 354 thousand tonnes (kt) of processed graphite were consumed in the U.S., including 60 kt natural graphite and 294 kt synthetic graphite. 145 kt of graphite were traded. Refractories and foundries consumed 56% of natural graphite; 42% of synthetic graphite went into making graphite electrodes. Batteries accounted for 10 and 5% of natural and synthetic graphite consumption, respectively; 78% of total graphite used dissipated into the environment; 22% reached the waste disposal stage of which 71% was landfilled and 29% was recycled; and 59 kt of graphite accumulated in in-use stocks. Recycling more graphite and producing graphite from lignin would favorably influence today's supply chain.



KEYWORDS: *natural graphite, synthetic graphite, material flow analysis, lithium-ion batteries, recycling, lignin*

1. INTRODUCTION

Given graphite's versatility and unique properties, it plays a critical role in various industrial and technology applications.[1,2] An allotrope of carbon, graphite, consists of tightly arranged carbon atoms in a hexagonal structure. The hexagonal rings form sheets that are held loosely together and can slide over each other.[3] Graphite is soft, chemically inert, and has high lubricity, stiffness, and thermal conductivity.[1,4] Besides metal, graphite is the only material that can conduct electricity.[5] These notable physical and chemical properties position graphite as a critical industrial mineral resource.[1]

1.1. Graphite Types and Their Properties. Broadly, there are two types of graphite: natural and synthetic. Table 1 compares the properties and uses of synthetic and natural graphite. Natural graphite is mined from rock. Depending on natural graphite's crystallinity, grain size, morphology, and reserve location, it is generally classified as amorphous, flake (crystalline), or lump and chip (crystalline).[1,4] Natural graphite is an indispensable material for refractories, brake linings, and steelmaking.[1,4] Calcining petroleum coke and coal tar pitch is the typical raw material of producing synthetic (or artificial) graphite.[6,7] Synthetic graphite is produced from the graphitization process with a high purity of 99.9%. Synthetic graphite electrodes are nonreplaceable in electric arc furnaces (EAF).[8,9] Compared to natural graphite, synthetic graphite generally has a lower density, lower electrical conductivity, and slightly higher porosity.[1] It is more expensive. Additional important end uses for both natural and synthetic graphite include batteries, carbon brushes, conductive materials, fuel cells, and lubricants.[1,10]

1.2. Graphite Demand Is Expected to Grow. The worldwide demand for natural and synthetic graphite, including graphite produced in the U.S., is expected to increase to support global greenhouse gas (GHG) emissions reductions in non-hydrocarbon energy provision, sustainable mobility, steel production, and digitization.[16,17] In energy storage systems, graphite usage in lithium-ion batteries (LIB), stationary batteries, lead-acid batteries, and fuel cells[16] is expected to increase five-fold by 2050 under a scenario that limits global warming to two degrees.[18] Graphite is also used in other core energy transition products: silicon for solar panels and rotor blades and electric brushes for wind turbines.[16] Graphite is a key ingredient in sustainable mobility approaches. It is a key component in LIBs and in composite light weighting materials

Received: November 17, 2022
Revised: February 2, 2023
Accepted: February 3, 2023
Published: February 15, 2023

Table 1. Comparison between Natural and Synthetic Graphite[1]

type	natural graphite: amorphous	natural graphite: flake	natural graphite: lump and chip	synthetic graphite
crystallinity	microcrystalline	crystalline	crystalline	microcrystalline to crystalline (less crystalline than natural graphite)
form	earthy to compact microcrystalline aggregates; grain size <4 μm	well-developed crystal platelets; grain size 40 μm−4 cm	interlocking aggregates of coarse crystals; powders to 10 cm pieces	2 μm powders to 2 cm pieces
product grade (% graphite)	60−90	75−97	90−99.9	99.95
prices ($/metric ton)	600−800	1150−2000	1700−2070	7000−20,000
main uses	refractories, foundries, paint, coatings, and batteries	refractories, brake linings, lubricants, batteries, and expandable graphite applications	carbon brushes, brake linings, and lubricants	batteries, carbon brushes, graphite electrodes, nuclear moderator rods (porosity unsuitable for refractories and foundries)
recycling	recycling of spent refractories to new refractories and roadbed materials through pre-sorting, crushing, screening, magnetic separation, and color separation.[11,12]			
	recycling of graphite from spent lithium-ion batteries by physical approach (direct separation after crushing and separation after artificial splitting) and chemical approach (pyrometallurgy and hydrometallurgy).[13,14]			
	recycling of broken electrodes as recarburiser.[15]			

that improve fuel economy. It is also used in high-performance brakes and charging stations.[16] Specialty graphite is used in semiconductor production powering digitalization.[16,19] Graphite demand is also increasing for other advanced technologies including aerospace applications, graphene, pebble-bed nuclear reactors, vanadium redox batteries, ceramic armor tiles, and electro-consolidation.[17,19] Natural graphite and synthetic graphite can both contribute to the clean energy transition. Accordingly, both types of graphite are crucial to the U.S. economy.

The battery industry is expected to be the largest demand driver for graphite.[18,20] According to a World Bank forecast,[18] graphite demand in 2050 for energy storage batteries, primarily LIB, will be five times higher than the total natural graphite produced in 2018 under a scenario that limits climate change to two degrees. Global production of both natural graphite and synthetic graphite is expected to double in 2028 compared to 2018 mainly because of the battery industry's rapid growth.[8] Currently, in the U.S., LIBs generally contain synthetic graphite supplemented with natural graphite. Synthetic graphite is dominant because it has a more desirable consistency, quality, and properties.[21] Globally, however, around 80% of the graphite in LIBs is natural graphite.[16,22] Synthetic graphite's relative cost (two−ten times higher than natural graphite) explains this difference.

1.3. The Graphite Supply Chain Is International. In 2018, global natural graphite and synthetic graphite production were 950 thousand metric tonnes (kt) and 1460 kt, respectively.[16] Currently, there is no natural graphite mining in the United States. Turkey, China, and Brazil have the largest natural graphite reserves, accounting for 78% of the world's total.[23] China dominates graphite production. It is the largest natural graphite-producing country[4] and produced around 630 kt (68%) of natural graphite in 2018.[16,24] This dominance is likely to continue over the next 10 years.[16] China, as the largest synthetic graphite producer, produced around 780 kt (54%) in 2018.[16] China also produces the largest amount of synthetic graphite electrodes globally. In 2018, China produced 370 kt of synthetic graphite electrodes, 50% of global production.[8]

Global demand growth for graphite, particularly in the energy sector, coupled with the mineral's limited and concentrated supply puts the supply chain of natural graphite at risk.[25] Reflecting these constraints, the European Union (EU) listed natural graphite as a critical raw material in 2020.[26] In 2018, the EU consumed 118 kt of natural graphite. Only 2 kt of this graphite was produced in the EU. The region consumed 297 kt of synthetic graphite, 268 kt of which was produced domestically.[16,27−29] The U.S. also considers natural graphite to be a critical and strategic mineral.[1] Like the EU, the U.S. does not produce natural graphite domestically. 60−80% of synthetic graphite consumed in the U.S. was produced domestically.[24] However, higher greenhouse gas (GHG) emissions[7,30,31] and higher costs[16] of synthetic graphite compared to natural graphite inhibit its competitive advantages. In addition, because it has high porosity, synthetic graphite is not suitable for refractories and foundries.[1] Given the importance of graphite to critical industries and applications in the U.S., it is necessary to understand how to reduce supply chain risks and environmental effects, especiallyGHG emissions.

1.4. Material Flow Analysis of Graphite: State of Knowledge. To address this need, we have developed a detailed and thorough material flow analysis (MFA) of graphite from cradle to grave for the U.S. in 2018. In general, MFAs systematically assess all flows and stocks of target materials in a spatially defined system over time.[32] They connect the resources, the intermediates, and final sinks of target materials to show pathways for sustainable utilization of resources.[32] A sink accommodates the output of a system, which is the opposite of a source.[33]

Previous MFAs trace the supply chains and uses of rare earth elements, metals, and plastics[34−36] but only a handful of earlier MFAs relate to graphite. Olivetti et al.[37] tracked global materials used in LIB for automobiles, electronics, and grid storage and raised the dominance of China in the natural graphite supply chain as a concern. Mayyas et al.[38] assessed the current raw materials supply chain for worldwide automotive LIB production to identify the potential economic and environmental impacts of recycling. They pointed out that the supply of natural graphite might be constrained by concentrated production in China and that lack of regulations and infrastructure for recycling LIBs limited recycling's ability to alleviate graphite supply chain constraints. Song et al.[39] combined a critical raw materials evaluation with MFA to investigate raw materials supply for LIB and waste management in China. They found a two-fold increase in graphite consumption in LIBs between 2013 and 2016. They noted that waste graphite was produced in large quantities and that recycling it could limit resource loss. Matos et al.[40] and Ciacci et

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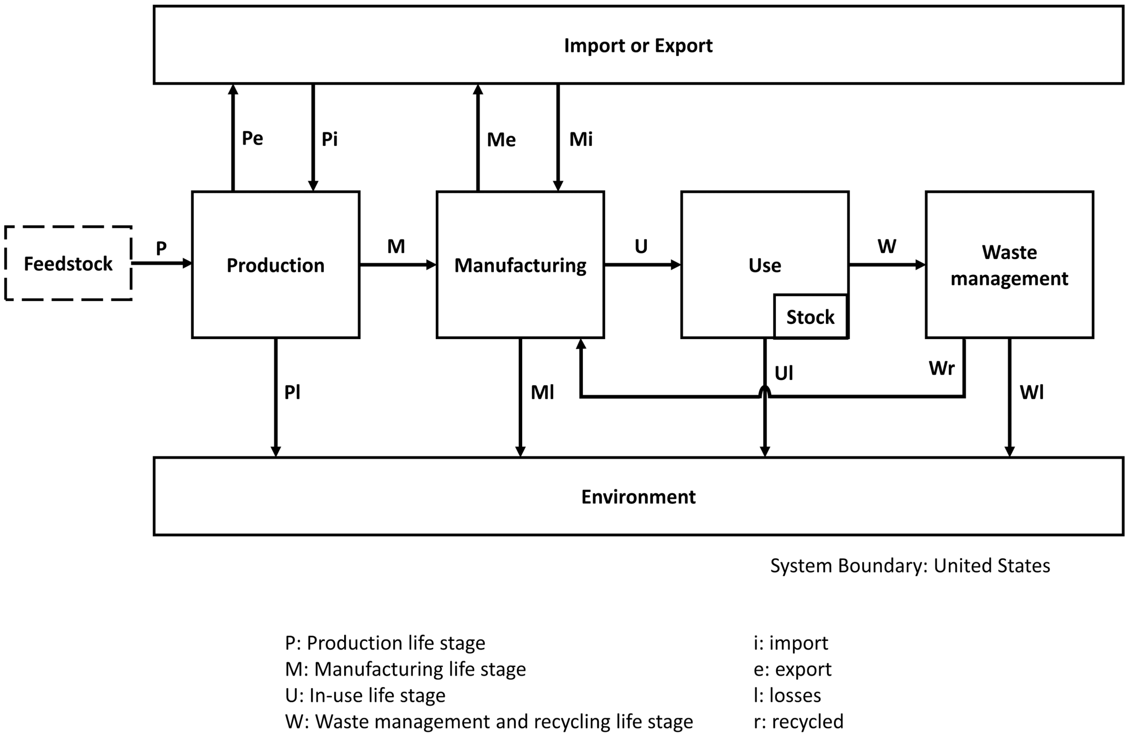

Figure 1. System boundary for natural and synthetic graphite material flow analysis in the United States.

al.[41] conducted MFAs on five battery-related materials, including natural graphite, from 2012 to 2016 for the European Union (EU) to develop overall knowledge of relevant supply chains from extraction to waste management. They found that industry in the EU was 99% dependent on imported natural graphite. The EU graphite recycling rate was low, only 8% in 2016. Dunn et al.[42] performed an MFA of global raw materials demand of LIB for electric vehicles and pointed out the potential circularity of recycling natural graphite in 2040. Rui et al.[4] conducted an MFA of natural graphite in China from 2001 to 2018. They reported extensive natural graphite domestic use and export. The recycling rate was only 0.7−5.7% between 2001 and 2018. Despite these contributions, the graphite-related MFA literature lacks a comprehensive assessment of all uses of graphite beyond LIB. No existing MFA includes synthetic graphite. Furthermore, no study provides a complete overview of natural and synthetic graphite in various end uses from a whole life cycle perspective for the U.S., which is among the many countries with ambitious energy storage targets that will tax the graphite supply chain.

To fill this gap, we provide a detailed and thorough MFA for natural and synthetic graphite in the United States for 2018, the most recent year with relevant data available from the U.S. Geological Survey, a critical data source.[24] The MFA includes processed graphite trade, production, graphite product trade, manufacturing, use, and waste management. The MFA in this study is static, as is typical for systems with relatively limited data[32,43,44] and relatively common for minerals used in LIB.[45−47] From detailed insights into dominant graphite uses and waste management practices in the U.S., we provide policy makers and stakeholders a system-level overview to identify challenges and opportunities for improving graphite resource efficiency. We supplement this analysis with an overview of graphite's criticality. The urgency of understanding graphite criticality and flows in the U.S. has grown with the passage of the Inflation Reduction Act of 2022,[48] which incentivizes the use of domestically-sourced and recycled graphite. Using the insights from this MFA, we identify the challenges and opportunities in each stage of the graphite supply chain in the U.S.

2. METHODS AND MATERIALS

2.1. System Boundary. We track the natural and synthetic graphite flows in the U.S. in 2018 with a classic MFA framework.[34] In this framework, the life cycle of graphite consists of four stages (Figure 1): Production, Manufacturing, Use, and Waste Management. The U.S. did not mine natural graphite domestically in 2018 but did produce synthetic graphite. Pet coke and coal tar pitch are feedstocks for synthetic graphite production in the U.S. 0.99 tonnes of pet coke and 0.24 tonnes of coal tar pitch (around an 80−20 ratio) are needed to produce one tonne of synthetic graphite.[7] The feed flows of pet coke and coal tar pitch for synthetic graphite production are excluded in this study. The import and export graphite flows in the production stage represent natural and synthetic graphite raw material flows. The U.S. imports and exports many products that contain graphite which we include as flows from the manufacturing stage. Some of the graphite produced in 2018 stays in use, which is reflected by the "stock" element of the use stage. All four stages exhibit graphite losses to the environment.

2.2. Assessment of Stocks and Flows. The mass balance principle applies in all stages and flows in the entire system to account for the graphite flows and stocks.[32] We calculated graphite flows ($\dot{x}_i$) from the mass flow of graphite-containing goods ($\dot{m}_i$) and their graphite concentrations (c_i) with eq (1).

$$\dot{x}_i = \dot{m}_i \times c_i \tag{1}$$

where $i = 1,\cdots,k$ refers to a graphite-containing good.

The graphite stock in the use phase of a certain year T ($S(T)$) is calculated as all of the incoming flows ($\dot{x}_{\text{in},i}\,(t)$) minus outlet flows ($\dot{x}_{\text{out},i}\,(t)$) from previous years as in eq (2). The outlet

https://doi.org/10.1021/acs.est.2c08655
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flows ($\dot{x}_{out,i}(t)$) are calculated with eq (3), a delay model.[4,49] It is equal to incoming flows ($\dot{x}_{in,i}(t - L_i)$) of a previous year ($t - L_i$) considering the life spans (L_i) of graphite products. As first defined by Ayres,[50] there are only two fates for materials in material flows—"dissipative loss and recycling or reuse." Based on this convention, we adopt the term dissipation to characterize the unrecoverable graphite. Fates of dissipated graphite include combustion, oxidation, sublimation, and wear. The remaining graphite that has recycling potential becomes waste, which is either landfilled or recycled. Most graphite products have a short life span and reach the end-of-life stage in 1−3 years.[4] Importantly, some graphite products wear out easily. These products dissipate to the environment during the use stage. Examples of these products include electrodes, lubricants, and pencils. It is important to deduct the dissipation of each graphite product when calculating the income flows of graphite to stocks. The average life span and dissipation rate of each graphite product are summarized in the Supporting Information (see Table S5).

$$S(T) = \sum_{t=1}^{T} \sum_{i=1}^{k} (\dot{x}_{in,i}(t) - \dot{x}_{out,i}(t)) \tag{2}$$

$$\dot{x}_{out,i}(t) = \dot{x}_{in,i}(t - L_i) \tag{3}$$

2.3. Criticality Assessment. The material criticality assessment aims to identify possible supply disruptions and the vulnerability of the U.S. to this disruption.[51] Graphite (natural and synthetic) is on the U.S. Geological Survey's 2022 list of critical minerals[52] based on the criticality assessment method developed by Nassar et al. 2020.[53] To gain insights beyond the U.S. Geological Survey's analysis, which does not distinguish between natural and synthetic graphite, we used other methods to validate the criticality of each type of graphite. Specifically, we used the methodology for critical raw materials assessment developed by European Commission research group,[54] adopting U.S.-relevant parameters. According to Schrijvers et al.,[51] who reviewed raw material criticality assessments during 2008−2018, the most widely used index to capture the diversity of producing or supplying countries is Herfindahl−Hirschman Index (HHI) combined with Worldwide Governance Indicators (WGI). The HHI is calculated as the sum of squared market share of each participant. It is highly responsive to the asymmetry of market shares and reflects the shares for every firm in the market.[55] The EU approach uses HHI to represent for country concentration and WGI to represent country governance.[54] The EU approach also includes recycling and substitution potentials. In the EU method, economic importance (EI) is a measure of a material's importance to economy in related end-use and supply risk (SR) is a factor reflecting the impact of disruption in supply of the material.[39] The EI, SR, HHI, and import reliance (IR) are calculated in eqs (4−7), respectively.

$$EI = \sum_{s} (A_s \cdot Q_s) \cdot SI_{EI} \tag{4}$$

where EI is the economic importance; s is sector; A_s is the share of graphite used in the sector; Q_s is the sector's gross value added; and SI_{EI} is the substitution index related to EI.

$$SR = \left[(HHI_{WGI,t})_{GS} \cdot \frac{IR}{2} + (HHI_{WGI,t})_{US\ sourcing} \cdot \left(1 - \frac{IR}{2}\right) \right] \cdot (1 - EoL_{RIR}) \cdot SI_{SR} \tag{5}$$

$$(HHI_{WGI,t})_{GS\ or\ US\ sourcing} = \sum (S_a)^2 WGI_a t_a \tag{6}$$

where SR is the supply risk; HHI is the Herfindahl−Hirschman index; WGI is the scaled Worldwide Governance Indicator; t is the trade relationship between importing and exporting countries; GS is the global supply mix; US sourcing is the supply mix to the U.S.; IR is the import reliance; EoL_{RIR} is the end-of-life recycling input rate; SI_{SR} is the substitution index related to SR; the S_a is the share of country a in supply of graphite.

$$\text{import reliance} = \frac{\text{import} - \text{export}}{\text{domestic production} + \text{import} - \text{export}} \tag{7}$$

2.4. Summary of Data. The data characterizing graphite flows in manufacturing, use, and trade of graphite-containing products are very limited. For example, the U.S. Geological Survey[24] withholds considerable amounts of these data to protect company proprietary information. Therefore, this study included various data sources including the U.S. Geological Survey,[24] the United States International Trade Commission (USITC) database,[56] industrial reports, and peer-reviewed literature to fill gaps (details in the Supporting Information).

We included 11 categories of end-use graphite applications:

- Graphite electrodes which are used in electric arc furnaces that melt scrap and other raw materials.[8,9]
- Refractories that consume magnesia-carbon and alumina-carbon refractory materials to produce steel.[19,57]
- Crucibles, binders, and ingot molds and ladles used at foundries.
- Four battery categories: primary batteries (single-use batteries), secondary batteries (rechargeable batteries), lithium-ion battery-powered mobile devices and cordless tools, and automotive lithium-ion batteries.
- Friction-reducing products in vehicles such as brake linings and clutch facings.
- Carbon products that include carbon brushes, components for electrical purposes, and graphite bearings.
- Solid lubricants, which typically contain graphite, as it is noninflammable and has high thermal stability, chemical inertness, and thermal conductivity.[58]
- Recarburising in steel production via electric arc furnace to increase the carbon content of steel.[59]
- Graphite shapes are purified and micronized graphite is added to metal power mixtures to fabricate sintered parts.[19] These graphite products provide lubrication and mechanical strength.[19]
- Rubber that contains graphite fillers to improve the properties of rubber materials. Properties include mechanical reinforcement, electrical or thermal conductivity improvement, and ease of processing.[60]
- Other products that include pencils and pencil leads, industrial diamonds, carbon bike frames, and paints are other graphite-containing products.

The detailed data and assumptions for graphite-containing product trade, the amount of graphite in various products,

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Environ. Sci. Technol. 2023, 57, 3402−3414

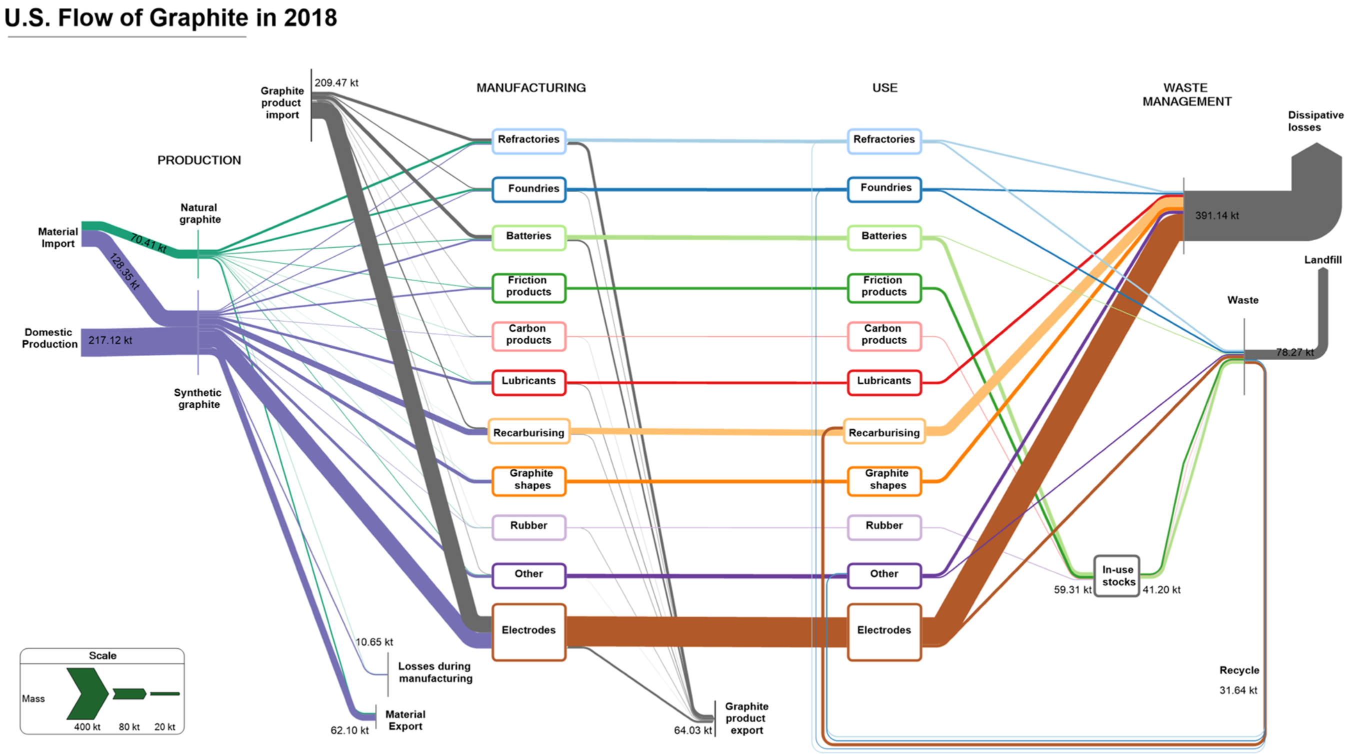

Figure 2. Reconciled material flow of graphite in the United States in 2018. Drawn using E!Sankey software. Natural graphite material is in green, synthetic graphite material is in blue, and graphite in graphite-containing products is in gray. After the manufacturing stage, graphite is mixed in applications and there is no distinction between natural and synthetic graphite. Dissipation includes combustion, oxidation, sublimation, and wear. Graphite that dissipates is unrecoverable.

products' recycling rates, in-use stocks, and waste management are summarized in the Supporting Information.

2.5. Waste Management of Graphite in the U.S. Graphite products are generally short-lived with useful lives of one to three years except batteries, which last longer.[4] Many graphite-containing products wear out and dissipate to the environment during their use phase.[4,11] For example, electrodes are consumed continuously (dissipation) and intermittently (broken). We treat graphite electrodes that are consumed continuously as dissipating. We treat graphite consumed intermittently as moving to the waste stage and assume that the breakage rate is 10%.[61] It was optimistically assumed that all of the broken electrodes were recycled as recarburizers in this study. The U.S. Geological Survey[24] has indicated that the market for recycled refractory graphite material is expanding. However, there are no data that estimate the quantity of recycled refractory graphite. Most recycled refractory waste is recycled as low-value roadbed materials, and the high-value recycling for refractory raw material is limited to 7% worldwide.[11,12] Therefore, in this study, we assumed that 7% of the refractory and foundry waste was recycled as high-value raw materials and 23% into roadbed materials.[4,62,63] Although LIB recycling exists in the U.S., it is cathode materials-focused and metals-focused.[64,65] Accordingly, we assumed no graphite in LIBs is recycled in the near term. Based on our optimistic assumptions, up to 29% of waste stage graphite could be recycled. The detailed assumptions and data used for waste management can be found in the Supporting Information.

2.6. Data Uncertainty and Data Reconciliation. We quantified data uncertainty with data quality indicator scores based on five dimensions: geographical representativeness, temporal representativeness, completeness, other correlations, and source reliability.[43,66,67] Once we assigned these scores, we quantified the data uncertainty as coefficients of variation (CV). Using multiple data sources introduces inconsistencies and contradictions. We reconciled these with the STAN MFA software.[68] STAN is widely applied in MFA for data reconciliation and aims to minimize the weighted necessary adjustment.[32] The data and its corresponding CV were adjusted during data reconciliation as detailed in the Supporting Information.

3. RESULTS

3.1. Characterization of Graphite Material Flows in the U.S. Figure 2 summarizes the reconciled material flows of graphite in the U.S. in 2018. Flows are expressed as thousand tonnes of graphite. Overall, in 2018, 354 kt of processed graphite was consumed in the U.S. Of this total, 60 and 294 kt were natural and synthetic graphite, respectively. Figure 2 (left) presents the origin of graphite used in the U.S. including domestic production and imports. Some of this graphite is directly exported. There was no domestic production of natural graphite. Two-thirds of synthetic graphite is produced domestically. The manufacturing phase of the MFA (Figure 2, middle) reveals that a significant amount of graphite is imported. The mass flow of imports is equivalent to the mass of domestically produced synthetic graphite. Compared to the amount of graphite imports, the U.S. exports a relatively small amount of processed graphite material (62 kt) and products (64 kt). In the waste management phase (Figure 2, right) a majority (391 kt) of graphite was dissipated and was lost to the environment. Only 110 kt of graphite entered the waste stage; 80 kt of this flow was landfilled and 30 kt was recycled. The

https://doi.org/10.1021/acs.est.2c08655
Environ. Sci. Technol. 2023, 57, 3402−3414

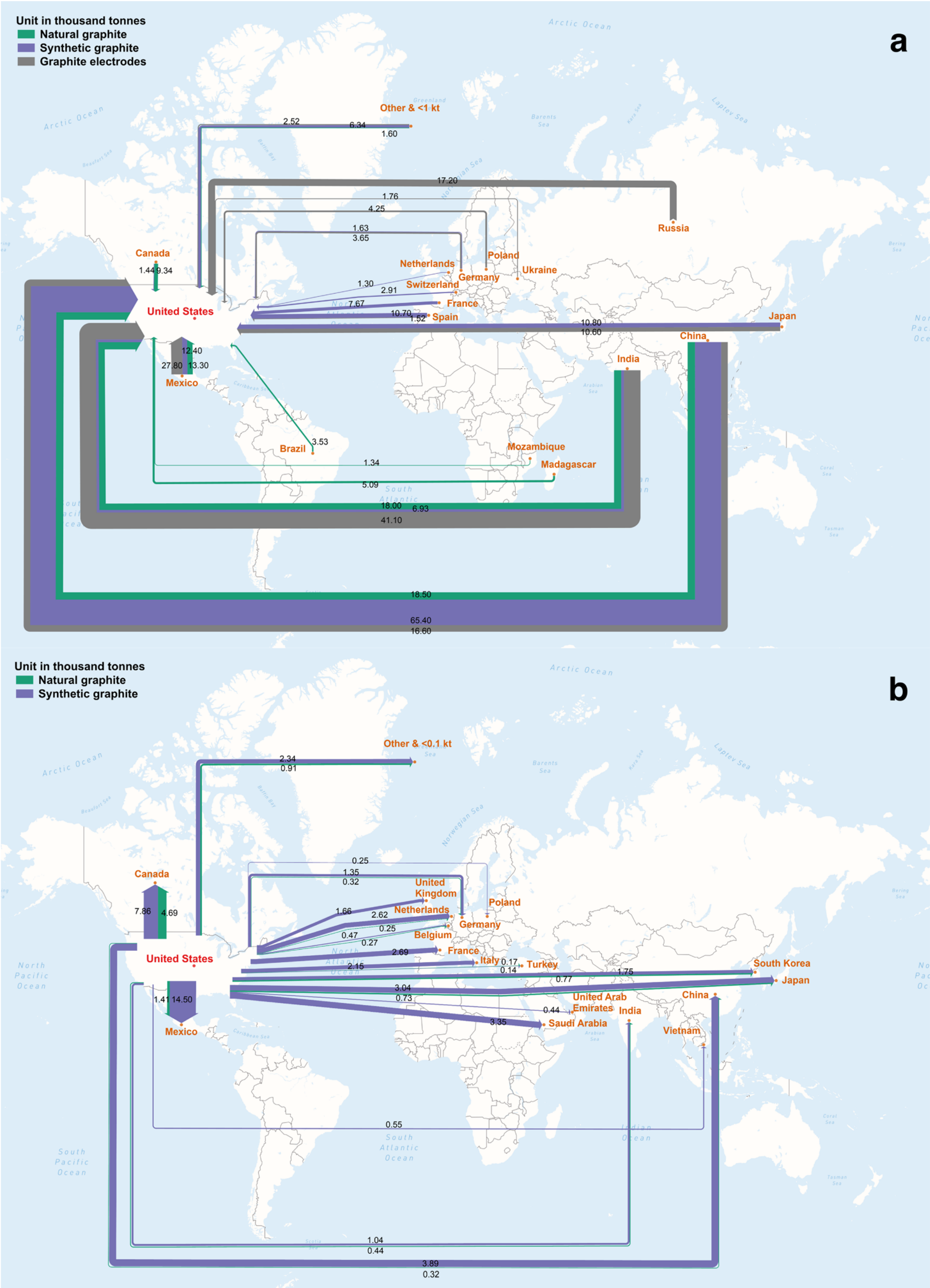

Figure 3. Graphite trade of United States in 2018. (a) Sources of natural graphite, synthetic graphite, and graphite electrode imports. (b) Destinations of natural graphite, synthetic graphite exports. Map image credited to OpenStreetMap under the Open Data Commons Open Database License (ODbL).[70]

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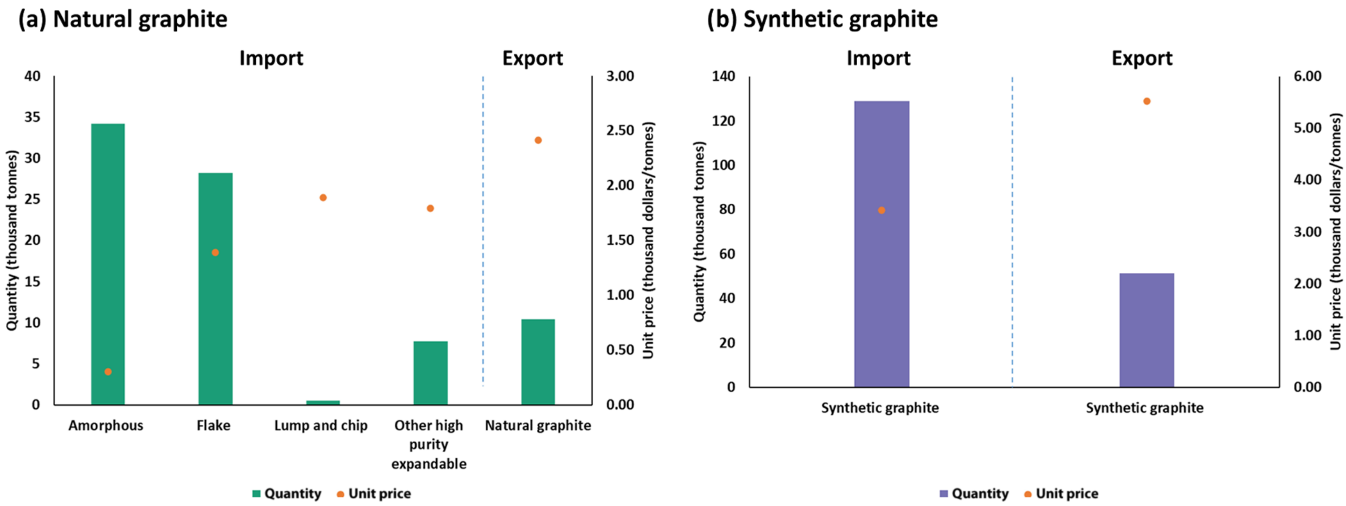

Figure 4. Total import and export quantities and unit prices of natural graphite and synthetic graphite for the United States in 2018.

recycling rate is therefore 6% of total consumption, which is low but on par with the EU and China.

In the manufacturing stage, almost all end-use applications consume natural and synthetic graphite. The major applications differ, however, for the two graphite types based on their properties, selling prices, and availability. Refractories and foundries consumed the largest share (58%) of natural graphite. Given the highly porous nature of synthetic graphite, which is not suitable for refractories and foundries, less than 30% of graphite consumed in these products is synthetic. Therefore, refractories and foundries are highly dependent on imported natural graphite. In the case of synthetic graphite, electrodes were the dominant end use (43%). Electrodes in EAFs consist solely of synthetic graphite. In response to the clean energy transition,[18] batteries were one of the main applications for natural (10%) and synthetic (5%) graphite in the U.S. Notably, the share of natural graphite (30%) was lower than of synthetic graphite (70%) in batteries produced in the U.S. In global battery production excluding the U.S., the share of natural graphite can be as high as 86% because it is less expensive.[16,22] Additional applications consumed between 1 and 8% and 1−15% of natural and synthetic graphite, respectively.

Compared to the manufacturing stage, the use stage exhibits similar consumption patterns. Electrodes were the dominant end-use application accounting for 48% of total graphite consumption. Recarburising and batteries consumed 15 and 7% of graphite, respectively. This result highlights that, beyond graphite use in steel production and metallurgical processes, batteries are a major market for graphite. The other applications, including refractories, foundries, friction products, carbon products, lubricants, graphite shapes, rubber, and other, accounted for between 0.6 and 7% (33% total) of graphite consumption. 13% of graphite flows in the use stage in 2018 accumulated in in-use stock.

As noted earlier, most graphite products have a short life span. They are easily worn or dissipated. Consequently, most graphite flows (78% of total graphite in the use stage in 2018) dissipated. We note that electrodes and recarburizers accounted for 54 and 19% of total dissipation losses, respectively. Only 22% of graphite at the end-of-life went to the waste stage where it has the potential to be recycled. In the waste stage, 19, 18, and 0.4% of graphite flows came from the in-use stock from 2014, 2016, and 2009, respectively. The overwhelming majority (71%) of waste graphite ends up in landfills. Of the 29% that is recycled, 74% is from broken electrodes that are recycled and used in recarburizing. One important conclusion from this analysis is that there is very limited opportunity at present to recover spent graphite through recycling. Even if lithium-ion battery recycling were common today in the U.S. and routinely recovered graphite, recovering all of the graphite in batteries would only add 21 kt of recycled graphite, which amounts to only 4% of total consumption. We discuss future trends in recovering graphite from spent batteries in the Discussion section.

3.2. Graphite and Graphite Products Trade. Graphite is a critical ingredient for steelmaking, a foundational industry in the U.S., and LIBs, a growing part of the country's decarbonization strategy. Understanding the origin of graphite, both natural and synthetic, and the dominant graphite use category in the U.S. (electrodes) is therefore pivotal to understanding supply chain risk. Accordingly, Figure 3 tracks import and export flows of natural graphite and synthetic graphite in 2018. Graphite electrodes are also included because they account for 63% of graphite in graphite product imports. While we can characterize imports by country for natural and synthetic graphite and electrodes, export data for the latter are lacking. Most natural graphite was imported from China (26%), India (25%), Mexico (19%), and Canada (13%). Siri Lanka was the only source for lump and chip natural graphite imports, accounting for 0.8% of total natural graphite imports. Synthetic graphite was mostly imported from China (51%), Mexico (10%), Japan (8%), and Spain (8%). Graphite electrodes were mostly imported from India (32%), Mexico (22%), Russia (14%), and China (13%). China's role as a dominant provider of graphite and graphite electrodes to the U.S. is unsurprising given that China is the world's largest graphite producer. However, graphite imports in 2018 were relatively geographically diverse. Imports from India and Mexico diversified the supply chain and reduce risks. Notably, Mexico has a free trade agreement with the U.S. and therefore can benefit from incentives in the Inflation Reduction Act to supply critical minerals to the U.S. Interestingly, although the U.S. does not domestically mine natural graphite as of 2018, the country exported 10 kt of natural graphite to Canada (45%), Mexico (14%), the Republic of Korea (7%), and Japan (7%). Synthetic graphite exports were bound for Mexico (28%), Canada (15%), China (7%), and Saudi Arabia (7%). Exports of graphite from the U.S. to Mexico

https://doi.org/10.1021/acs.est.2c08655
Environ. Sci. Technol. 2023, 57, 3402−3414

and China stand out because the U.S. imports a large amount of graphite from these countries.

Total quantities of natural and synthetic graphite imports and exports along with corresponding unit prices are shown in Figure 4. In 2018, graphite export prices exceeded import prices by 1.3−8 times for natural graphite and by a factor of 1.6 for synthetic graphite. We can therefore conclude that graphite exports from the U.S. have been refined to improve properties and value. Advanced refining technology is a key to the future growth of graphite industry and increases the profitability of graphite in the U.S.[17] Examples of graphite product trade can be found in the Supporting Information.

3.3. Criticality Assessment Results. The criticality assessment results of natural and synthetic graphite for the U.S. in 2018 are shown in Figure 5. The thresholds of EI = 2.8 and SR = 1, which determine the criticality of materials, are based on European Commission 2017.[69] Both natural and synthetic graphite are in the critical area. This result for the U.S. is unique because previous assessments combine the two graphite types despite their unique applications, properties, and sources. The SR of natural graphite is high mainly due to the highly concentrated supply from China, a World Governance Indicator for China that indicates instability, and, overall, a high import reliance for the provision of natural graphite. Comparatively, the SR of synthetic graphite is just above the threshold mainly because of high domestic production and limited import reliance. The EI of natural graphite is just above the threshold because natural graphite is mainly used in refractories and foundries and the gross value added for graphite in these applications is low. By contrast, the high EI of synthetic graphite is attributed to the relatively high gross value added of dominant end-use in electrodes. The SR and EI of all types of graphite (natural and synthetic) are similar to synthetic graphite because synthetic graphite is the main type of graphite used in the U.S. The results of criticality of all types of graphite are consistent with the U.S. Critical Minerals List.[52] The detailed calculations and parameters are in the Supporting Information.

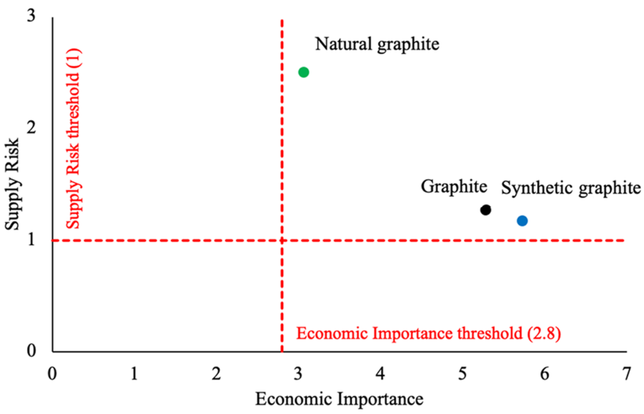

Figure 5. Criticality assessment results of natural and synthetic graphite for the United States in 2018.

3.4. Data Uncertainty and Challenges. The data and its CV were both adjusted during data reconciliation (detailed calculations are in the Supporting Information). The "Other" category in the manufacturing stage has the highest CV. This category is calculated as the difference between the total flow of graphite and all of the graphite flows excluding those in the Other category. The end-use categories of synthetic graphite (except for flows of graphite in electrodes which are based on high-quality data) have high CVs because the data on which they are based does not represent the U.S. very well geographically (world vs U.S.). Because the graphite waste and recycling flows are estimated based on indirect data and experts' opinions, some of these flows have high CVs. As 89% of CVs are under 10%, the data uncertainly would not significantly affect overall material flow analysis results of graphite as well as environmental and supply risk mitigation opportunities in this study. To reduce data uncertainty, efforts should be made to share end-use and recycling data in future graphite MFA studies. This need is especially great for synthetic graphite because it is the main graphite type used in the U.S. Data related to its production, use, and end-of-life in the U.S. are very limited.

4. DISCUSSION

The MFA we have developed highlights four challenges and opportunities for graphite use in the U.S. as demand for graphite grows.

4.1. Synthetic Graphite, Costly and GHG-Intensive, Dominates Graphite Use in the U.S. Eighty-three percent of graphite used in the U.S. is synthetic (Figure 2). Compared to natural graphite, synthetic graphite is more expensive (Table 1). We consider this cost differential in the context of LIBs, one of the major drivers of graphite demand. The higher cost of synthetic graphite could directly influence the cost competitiveness of U.S.-produced electric vehicles. As mentioned, in spite of its higher price, in the U.S. synthetic graphite is more commonly used in LIBs (70% in 2018 per Figure 2) than natural graphite.[21] Synthetic graphite is widely available in the U.S. and it offers batch-to-batch property consistency. If we assume that synthetic graphite is double the cost of natural graphite, an electric vehicle (EV) with synthetic graphite in its battery will cost \$430 more than an EV with a battery containing natural graphite.[21] This difference may be an underestimate because synthetic graphite can cost up to ten times more than natural graphite.

In addition to being more expensive, synthetic graphite is more GHG-intensive than natural graphite. Natural graphite production normally includes mining, beneficiation, purification, postprocessing, and transportation.[31] The cumulative GHG emissions of natural graphite production range from 2.3 to 7.8 kg CO_2e per kg graphite (kg CO_2e/kg).[30,71−73] Based on the imported natural graphite flows in Figure 2, 162−549 kt CO_2e emissions are associated with imported natural graphite.

Synthetic graphite used in LIBs is produced from high-temperature processing of fossil fuel feedstocks. This process begins with green coke production from oil refining or catalytic cracking of heavy oils. This coke is calcined, graphitized, purified, processed, and transported to its point of use.[31] The GHG emissions of graphitization alone are in the range of 0.5−4.9 kg CO_2e/kg.[7,74] Total GHG emissions of synthetic graphite production can be as high as 20.6 kg CO_2e/kg.[31] The GHG emissions associated with domestically produced and imported synthetic graphite in Figure 2 are 7117 kt CO_2e GHG. Taken together, the GHG emissions associated with producing the graphite used in the U.S. in 2018 per the mass flows in Figure 2 is 9000 t CO_2e.

Overall, natural graphite has 62−89% lower GHG emissions than synthetic graphite. Coupled with its lower cost, if it weren't for natural graphite's supply chain constraints it would be a better choice for decarbonization technologies in terms of GHG emissions and costs. If natural graphite replaced synthetic graphite in battery production in 2018 in the U.S., the GHG mitigation potential could be 178−256 kt CO_2e. Cost reductions could range from 14 to 65 million US$ (based on prices in Figure 4). As renewables increasingly penetrate the grid and electricity production is less GHG-intensive, the GHG emissions of producing both types of graphite—driven in large part by electricity consumption—will gradually decline.[16]

Beyond GHG emissions, graphite production incurs other environmental burdens. Examples include fine particulate pollution, water consumption, toxic substance usage, mineral resource consumption, and fossil fuel depletion.[1,21,31] These effects should be incorporated more fully into life cycle assessments (LCA) of graphite to aid in efforts to mitigate the environmental effects of the clean energy transition.

4.2. Synthetic Graphite Dominance May Mask Energy Security Risks of Graphite Use in the U.S. The SR of synthetic graphite in the U.S. is relatively low, but if its supply is disrupted, its high EI indicates the economy will be significantly affected. Given this somewhat uncertain situation, it is somewhat surprising that the U.S. produces enough graphite to play a major role in international synthetic graphite trade. In fact, in 2018, the U.S. accounted for around 8−20% of global synthetic graphite exports[16,17,21] while the import reliance of processed natural and synthetic graphite raw material for the U.S. was 39%. However, when accounting for trade in graphite-containing products, the total import reliance for graphite increases to 57%. These statistics highlight that although large amounts of synthetic graphite are produced domestically, graphite products (manufacturing stage in Figure 2) in the U.S. still mainly depend on imports.

Looking at two case studies can help clarify this insight. First, the dominant force behind increasing graphite demand in the near future is likely to be the manufacturing of LIBs.[17,18] For example, Tesla is one of the dominant LIB producers in the U.S. At its Gigafactory in Nevada, it produced roughly 20 GWh of batteries in 2018, consuming about 10 kt of graphite. This mass is 50% of total graphite consumed in battery manufacturing in the U.S. in 2018 (Figure 2).[21,75] Notably, of the graphite-containing end-use products the U.S. imports, most are primary batteries, lead-acid batteries, and lithium-ion batteries. On the other hand, exports are mainly high-value-added electric vehicles. Given the higher cost of U.S. domestic synthetic graphite compared to natural graphite, EVs manufactured in the U.S. could suffer a competitive disadvantage if they use this more expensive form of graphite. (EV cost, however, is determined by many factors.) The import reliance of graphite in batteries is 46%.

A second case study is graphite's imperative role in decarbonizing steel. Steel production approaches that use graphite electrodes (electric arc furnaces and hydrogen direct reduction) are less GHG-intensive than basic oxygen furnace steel manufacturing technology. While hydrogen direct reduction technology remains under development,[76] GHG emissions from electric arc furnace-based steel manufacturing are 70% lower than steel manufacturing with basic oxygen furnaces.[77,78] In 2018, producing graphite electrodes consumed 49% of graphite in the U.S. As demand for low-carbon steel increases, more graphite may be consumed in electrode production. The import reliance of graphite in electrodes is 48%. So, the nation's ability to increase electric arc furnace steelmaking may depend on secure domestic graphite sources. Given the potential to reduce CO_2e emissions by 74 million tonnes (based on 2018 flows in Figure 2), reducing the import dependence of electrodes is essential.

One way to reduce this import dependence and remove graphite from the list of critical minerals is to increase mining for natural graphite in the U.S. Recently in the U.S., three companies were developing and evaluating natural graphite projects. These include the Coosa Graphite project in Alabama, the Graphite Creek project in Alaska, and the Chedic Graphite project in Nevada.[17,21] However, these projects will take longer than 10 years to start actual production. The reserves in these projects are limited compared to anticipated graphite demand.[21] While China will remain a dominant graphite producer, India, Mexico, Brazil, and Africa are all likely to be important graphite producers and exporters.[37] The supply of natural graphite will gradually diversify which could alleviate supply chain pressures in the long term.

4.3. Opportunities to Recycle Graphite Should Be Expanded. Figure 2 highlights the amount of graphite lost to dissipation at the end of life. This graphite is not available for recycling. Of graphite that does not dissipate, 71% goes to landfill at end of life even under the optimistic estimates of graphite recycling that we assumed. In fact, against the flow of graphite consumed in the U.S. in Figure 2, the flow of recycled graphite is vanishingly small. Opportunities to increase recycling lie in recovering end-of-life batteries and electrodes and extracting the graphite they contain for recycling and reuse.

As more LIBs are produced, there is a growing opportunity to recover the graphite they contain.[18] In 2018, 29 kt of graphite from batteries came into in-use stock in the U.S. At the same time, only 22 kt from batteries left in-use stocks for end-of-life disposal. LIBs have a relatively long life span. Their in-use stock will rise with increases in their manufacturing. Eventually, this in-use stock will reach end-of-life. The projected flow of end-of-life graphite in the U.S. will be in the range of 40−95 kt in 2030.[79] This expected flow may be slowed by secondary use of automotive LIBs, which often retain 70−80% of their maximum capacity for years,[80] in other low-end stationary energy storage applications.[4,18] It is important to recycle the graphite in these batteries (automotive or stationary) rather than follow today's predominant practice of landfilling it.

Graphite recycling from batteries has the potential to be both environmentally responsible and profitable. While much of LIB recycling technology development has emphasized cathode materials, a few recent studies aim to recycle graphite from the anode.[80,81] Notably, recycling spent LIB graphite could have a co-benefit of recovering cathode metals (particularly Li) entrained in it.[80] According to Rey et al.,[25] nine recycling methods to recycle graphite from spent LIB have estimated GHG emissions of 0.5−9.8 kg CO_2e/kg graphite recovered. These emissions are comparable to GHG emissions from natural graphite production. This comparable result raises an important point. Technology developers must use LCA and other methods to evaluate the environmental performance of graphite recycling compared to conventional production to develop processes that offer environmental benefits. These LCAs should consider property differences between virgin and recycled graphite. For example, recycled anode graphite from LIBs exfoliates to graphene more readily than other graphite forms,[82] which can increase the profitability of LIB recycling.

It is also important to consider approaches to reducing graphite consumption. One such example is the better management of electrodes. Approximately 24 kt of electrodes were broken and recycled as recarburizers in the U.S. In 2018. Through advanced production of steel and adoption of best operation practices for electric arc furnaces that follow

https://doi.org/10.1021/acs.est.2c08655
Environ. Sci. Technol. 2023, 57, 3402−3414

appropriate electrode placement procedures,[9,83] the breakage rate of electrodes could drop from 10 to 5%. The corresponding decrease in graphite demand for electrodes would be around 12 kt, which is equivalent to the 2018 consumption of synthetic graphite demand for domestically produced batteries. Notably, 77% of recycled graphite from refractories and foundries is used as low-value roadbed materials. To achieve a reduction in graphite demand, recycling technology should be engineered to produce high-value recycled graphite rather than low-value uses. Such advances will provide economic incentives for recycling and capture the full value of graphite.[11,12]

The challenges associated with graphite recycling and the relatively low cost of this mineral combine to dampen the motivation to recycle graphite in the U.S. As a result, practical, scalable, and profitable recycling routes must be developed to reduce the dependence of U.S. on graphite imports and the environmental effects of graphite production.

4.4. Alternative Graphite Feedstocks Should Be Pursued. In some applications, it is not yet possible to look for alternatives to graphite. For example, there is no available alternative for natural graphite in refractories, brake linings, and steelmaking.[1,4] To date, no other material can match the properties of synthetic graphite that are essential for use in graphite electrodes.[8,9] It is possible, however, to use other materials in place of graphite in batteries. Graphite anodes in LIBs could be replaced by other carbon materials such as hard carbon and nanocarbon,[84,85] silicon-based materials,[86] or metallic materials.[87] For rapidly emerging new battery technologies,[18] the anode materials for solid-state batteries and zinc-air batteries are lithium and zinc, respectively. However, many factors, such as high costs, production scale-up, and safety requirements still impede the widespread market deployment of these new battery technologies. Accordingly, graphite is expected to be the major LIB anode material for at least the next decade.[88]

As described, production of both natural and synthetic graphite is extractive and fossil-fuel based. Furthermore, there are many challenges in meeting the increasing graphite demand with recycling. Therefore, the need for alternative graphite feedstocks, including lignin,[89,90] that may be less environmentally burdensome, cheap, and without supply constraints is essential and urgent. Lignin is a low-cost and abundant carbon source.[91] It can be found in lignocellulosic biomass, such as wood and switchgrass. It is also found in agricultural residue like wheat straw and corn stover. In 2010, there were up to 50 million tonnes of lignin isolated as a byproduct from pulping processes and the production of cellulosic fuels.[92] The value of lignin has been explored for several applications, such as a concrete additive, animal feed additive, and phenolic resins.[90] Besides these applications, graphitization is a pathway from lignin to graphite.[91,93,94] Lignin-derived graphite can potentially substitute for natural and synthetic graphite in several high-tech applications, especially as an anode material in batteries.[89] There is already scalable commercial production of lignin-derived graphite. In 2019, Stora Enso, a pulp and paper producer in Finland, invested 10 million euros in a pilot plant to produce lignin-derived graphite as a replacement for conventional graphite in lithium-ion batteries.[95] According to Hermansson et al.[96] and Moretti et al.,[97] the GHG emissions of dry lignin extraction range from 0.2 to 0.6 kg CO_2-e/kg. If the lower end of GHG emissions (0.5 kg CO_2e/kg)[7] of the graphitization process is applied for lignin-derived graphite production, the lignin-derived graphite has the potential to reduce GHG emissions by 52−86% compared to natural graphite and by 95−97% compared to synthetic graphite. The total GHG mitigation potential in the U.S. in 2018 could be 6821−7375 kt CO_2e, if all imported and domestic produced natural and synthetic graphite in Figure 2 were replaced by lignin-derived graphite. Lignin has the potential to be a low-GHG, domestic, renewable source of graphite as evidenced by the growing body of research focusing on various lignin-derived materials used in the energy storage field.[90]

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c08655.

Graphite flow data (xlsx)

Graphite trade data (xlsx)

Uncertainty analysis (Tables S1−S3 and Figure S1); summary of data sources (Table S4); trade flow details (processed graphite, electrodes, refractories, foundries, batteries, friction products, carbon products, lubricants, recarburizing, graphite shapes, rubber, and other); examples of graphite product trade (Figure S4); in-use stock, waste, and recycle (Table S5); battery life span sensitivity analysis (Figures S2 and S3); data limitations; material flow of graphite (Figure S5); and data reconciliation (Figure S6) (PDF)

AUTHOR INFORMATION

Corresponding Author

Jennifer B. Dunn − *Department of Chemical and Biological Engineering, Northwestern University, Evanston, Illinois 60208, United States; Northwestern-Argonne Institute of Science and Engineering, Evanston, Illinois 60208, United States; Center for Engineering Sustainability and Resilience, Northwestern University, Evanston, Illinois 60208, United States;* orcid.org/0000-0002-2065-5106; Email: jennifer.dunn1@northwestern.edu

Authors

Jinrui Zhang − *Department of Chemical and Biological Engineering, Northwestern University, Evanston, Illinois 60208, United States;* Present Address: Institute of Energy, Peking University, 5 Yiheyuan Road, Beijing 100871, China; orcid.org/0000-0002-9468-070X

Chao Liang − *Institute for Sustainability and Energy at Northwestern, Northwestern University, Evanston, Illinois 60208, United States;* orcid.org/0000-0002-3307-7712

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.2c08655

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation Future Manufacturing Program (NSF CMMI-2037026).

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https://doi.org/10.1021/acs.est.2c08655
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