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TOPICAL REVIEW

Graphene Roadmap Briefs (No. 4): innovation prospects for Li-ion batteries

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Keywords: graphene and related materials, innovation roadmap, industrialization, lithium ion battery (LIB), Si anode

Abstract

The present issue of Graphene Roadmap Briefs compiles results on the status and prospects of graphene and related materials (GRM) in Li-ion batteries (LIB) applications. It explores both the technical feasibility and market perspective based on two dedicated innovation interface investigations (3I) carried out in 2017 and 2022, respectively. Each consisted of extensive desk research, expert consultations and an interactive roadmap workshop to aggregate the contemporary innovation prospects for the emerging value chain. The combined results provide us with the unique opportunity to also track key developments in LIB technology and changes in GRM perception in that sector over the course of time. In essence, GRM diffusion as a battery electrode material will likely evolve around high-quality materials that offer superior performance, and not simply around low-cost production of arbitrary GRM types. Si-based LIB anodes constitute a particularly promising application areas for GRMs in LIB. Despite remaining challenges in both R&D and industrial scalability, the increasing commercial interest in Si-based LIB anodes may constitute a driver for overall GRM commercialization, as their functionality is already confirmed.

About: Graphene Roadmap Briefs

Graphene Roadmap Briefs highlight key innovation areas impacted by graphene and related 2D materials (GRMs) as well as overarching aspects of GRM innovation status and prospects. The series bases on the evolving technology and innovation roadmap process initiated by the European Graphene Flagship initiative. It covers crucial innovation trends beyond fundamental scientific discovery and applied research on GRM utilization opportunities.

List of acronyms

GRMs	Graphene and related 2D materials
TIR	Technology and innovation roadmap
3I	Innovation interface investigation
G	Graphene
GO	Graphene oxide
rGO	Reduced graphene oxide
GNP	Graphene nanoplatelet
LIB	Lithium-ion battery
C2P	Cell-to-pack
SEI	Solid electrolyte interface
TRL	Technology readiness level
OEM	Original equipment manufacturer
KPI	Key performance indicator
CVD	Chemical vapor deposition

The list of abbreviations and acronyms excludes proper names, common use (such as 2D), metric system units, chemical symbols, and isolated introductions (for terms most common as acronym such as CNT).

1. Introduction

The practical isolation of graphene in 2004 [1] sparked considerable expectations in terms of scientific discovery, technological application potential, and economic value. The translation of a novel material into widespread economic impact is a process that takes substantial time spans [2] and present market developments appear to lag behind initial expectations [3]. Nevertheless, GRM promise further technology improvements in various sectors and applications [4], particularly including electrochemical energy storage [5].

Rechargeable battery technology is currently considered a key technology for the European Union [6]. Today, the lithium-ion battery (LIB) represent the electrochemical storage technology of choice with highest energy densities commercially available. Hence, LIB constitute an enabler for various types of battery applications that require different performance characteristics such as consumer electronics, electric vehicles (EVs) and stationary energy storage systems [7]. Due to improvements in battery technology and cost reduction, the share of battery-powered equipment and applications has increased tremendously during the last years [8]. For instance, the global battery EV (BEV) market increased more than ten times from 2017 to 2023. Unit sales rose from 0.77 million to 9.5 million in only six years, leading to a market share of 18% for BEV among global car sales in 2023 [9]. Battery production capacities of several 100 GWh p.a. have already been announced or built up, in particular to serve the growing battery-electric mobility demand [10].

To further develop and improve the LIB technology, research is being conducted into new battery technologies and materials that are more powerful, more sustainable or cheaper. Historically, the reduction in cell costs and enhancement of performance have resulted in the emergence of a range of suitable cathode materials, including Ni-rich/low-Co mixed metal Li, Ni, Mg, Co oxides (NMC) and lithium iron phosphate (LFP) [11, 12]. In contrast, fewer developments have taken place on the anode side in recent years. In essence, graphite has been the state of the art for the anode in LIB ever since their initial commercialization in the 1990s [13]. The mechanism of Li-storage in graphite is based on Li-intercalation between its carbon monolayers. Here, the de-/lithiation process induces rather small structural volume change of about 10% during cycling [14]. Today, an increasing interest in silicon-based anodes for advanced LIB designs can be observed [11, 15]. Promises and challenges of Si incorporation in LIB anodes are widely discussed [13, 16]. Roadmaps of cell manufacturers and OEMs suggest the broader industrial diffusion of silicon as an anode material in the next few years until 2030 [17]. The utilization of advanced materials, particularly those comprising silicon as a dominant active material, represents a significant and emerging trend in the field of anode performance enhancement [15, 18].

The combination of carbon materials, graphite and silicon in Si/G/C composites improves electrochemical performance by combining the high capacity of silicon with the cycling stability of graphite [19] and the conductivity of carbon materials. Carbon materials such as carbon nanotubes (CNTs) and rGO increase electrical conductivity and buffer volume changes in silicon, which improves mechanical stability and cycle stability [20]. At the same time, they promote a uniform distribution of the silicon particles and optimize the contact surfaces, thus maximizing the capacity of the composite, therefor creating a system with improved capacity, stability and conductivity [20].

In general, the production of SiO_x anode materials is already an established business with high growth rates since the 2020s [21, 22]. Si producers already identified battery materials as a major and rapidly expanding market [21, 23]; therefore, they dedicate significant efforts and resources towards developing specialized solutions. Pure Si-electrodes offer a theoretical specific capacity of 4200 mAh g^{-1} [24] but at the expense of excessive volume expansion. Si particle size may increase by up to 300%–400% [13, 24, 25] during lithiation, inducing substantial mechanical stress on particles, interface, and electrodes. This leads to instabilities of the overall electrode/electrolyte system and an overall poor cycling stability of the respective cells.

In this context, GRM come into play as an option to aid the further improvement of silicon–carbon composite (Si/C) integration, highly determining Si-based anode performance and LIB properties. The unique electronic, mechanical and chemical properties of graphene enable ample possibilities for battery cell enhancement [26–28]. These properties make graphene attractive for achieving both high-energy and high-power LIB [29]. The electrical and physical properties of graphene are well suited to the increasing electrical conductivity, enabling enhanced rates of charge and discharge and greater power density. The robust structure of graphene offers resistance to particle volume changes, thereby improving mechanical stability and prolonging cycling lifetime. The large surface area of graphene further enhances lithium-ion storage, thus increasing the energy density. For instance, it has been proposed as cathode additive to improve the electronic conductivity [30], as an interlayer between components, or as part of a current collector [27]. Interest surges regarding the exploration of GRM as LIB anode material [15] though, in particular in the context of supporting the performance of Si-based anodes [31, 32]. Actors strive to increase the Si content, but struggle with constraints in the active Si fraction. Issues include Si particle swelling, low first discharge efficiency as well as the low ionic and electronic conductivity of Si [18, 26, 33]. Here, the multi-functional character of GRM promise to address several issues simultaneously and, thus, fundamental breakthroughs in lithium-ion batteries [34]. The right weight ratio between silicon, graphite and carbon materials, as well as carbonaceous additives to form hierarchical structures of composites, are considered as crucial for high performance [20].

In contrast to the silicon materials business, the GRM research and industry community still lacks a unified perspective on whether, when and how GRM may find their entry into battery value chain. Here, roadmapping may provide guidance for both science and industry. The present issue of Graphene Roadmap Briefs focuses silicon-based LIB anodes to explore probable pathways to GRM diffusion in the battery industry, with particular regard to ongoing efforts to establish a competitive battery value chain in Europe. We employ the 3I framework [35] to aggregate relevant expert perspectives along the potential future value chain for GRM-enhanced LIBs (section 2). In particular, our results base on two dedicated 3I roadmap focus investigations carried out in 2017 (section 3) and 2022 (section 5), respectively. We discuss both innovation roadmaps in the light of applicable KPI relevant to the battery-electric supply chain—with particular regard to GRM-enhanced electrode materials to overcome technical challenges for future battery designs. We review and analyze key developments and the progress achieved in the meantime (section 4) to gain a comprehensive understanding of the innovation context and prospects for all stakeholder groups along the emerging value chain for GRM-enhanced LIB anode materials (section 6).

2. Methodology: innovation chain logic

The 3I concept [35] enables the exploration of potential future value chains with a high degree of complexity. Novel materials such as graphene may hold ample promise for diverse applications, but typically the pathways to widespread market diffusion are (a) opaque in the direction of impact, (b) distant in the future, (c) highly specific in application, and (d) usually require complex interactions among multiple stakeholder groups. Thus, we employ 3I focus investigations to elucidate GRM innovation potential for advanced Li-ion batteries.

The concept first calls for rigorous desk research to explore relevant stakeholder groups along with their current innovation status and prospects for their field. The results enable the construction of an innovation chain that approximates future supply-demand relationships. Figure 1 shows the adoption of the generic GRM innovation chain to the LIB context. It recognizes 3 basic innovation spheres from materials supply over component to final product levels:

(1) Anode materials, where GRM may claim a promising niche as additive for advanced anode compositions,
(2) Li-ion batteries as component level industry providing energy storage solutions, and
(3) Automotive and wearables as selected end-use industries providing demand and impulses for upstream industries.

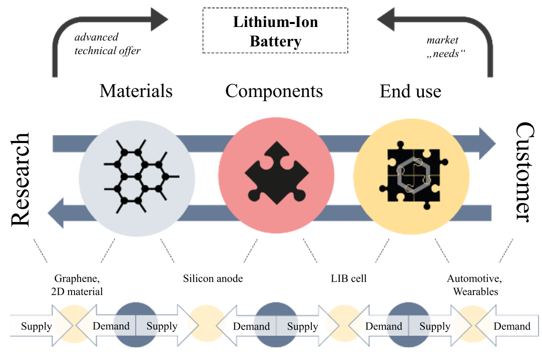


Figure 1. Value chain approach for TIR research following the 3I scheme with the use case ‘Li-ion battery’, adapted from [35]. © The Author(s). Published by IOP Publishing Ltd. CC BY 4.0.

Of course, ample sub-structure (in terms of internal supply chains and diverse stakeholder perspectives) may exist in either innovation sphere. Usually, the information exchange among peers (within each sphere) is much stronger than across spheres, in particular when reliable supply-demand relationships are not yet established. The 3I concept recognizes innovation interfaces as a precursor for future value creation. We identified the development of silicon anodes as a particularly promising pathway for GRM diffusion into the LIB market. The innovation chain (figure 1) consists of two major innovation interfaces (colored yellow): LIB cell producers interact with both (a) upstream suppliers to develop advanced silicon anodes and (b) downstream customers to define future product generations according to demand. From the perspective of emerging GRM suppliers, the former constitutes a direct innovation interface, where they aspire to enter the value chain, while indirectly depending on market impulses provided by end product industries.

At both ends of the innovation chain, the translation of (c) GRM research results into an emerging supply industry and of (d) consumer demand into advanced end products constitute innovation interfaces in their own right (colored grey in figure 1). However, these are relatively trivial in nature, as these interactions constitute the core business of the respective industries. Thus, we consider researcher perspectives among the industrial stakeholder group aligned to their subject (at material, cell, and product level) and EV manufacturers and wearable producers as proxies for consumer interests.

The 3I approach calls for extensive expert consultations (usually in a confidential setting) along the entire innovation chain to refine the status and prospects of each relevant stakeholder group and explore current perception of the associated innovation interfaces. On this basis, selected experts representing all stakeholder groups can be invited to an interactive roadmap workshop [35] (under the Chatham House

Rule [36]). With regard to GRM prospects for battery applications, we conducted two 3I focus investigations in 2017 and 2022, respectively. The earlier one culminated in an in-person workshop that assembled 21 experts from industry and research who represented all relevant perspectives from materials production, over battery cell manufacturing to automotive and wearables industry. Of course, the 2022 focus investigation built on the earlier results and reached beyond a mere repetition. Utilizing a fair mix of previous consultation partners with new actor perspectives, we systematically reviewed the progress achieved in the meantime. Adhering to 3I principles, we selected 20 representatives to contribute to an interactive online roadmap workshop to complete the roadmap revision and extension.

3. The initial graphene battery roadmap (2017)

First, we highlight the key results of the initial focus investigation carried out in 2017 [37]. Among the plethora of potential GRM applications, the battery topic was chosen due to a notable upsurge in interest surrounding battery technology and battery-electric application markets across Europe at this time. Moreover, energy storage devices were one of the focus topics in the Graphene Flagship at this time. The primary objective of the inaugural roadmap was therefore to illustrate the potential integration of a novel material GRM into a nascent technology LIB for promising large application markets.

Energy storage for electric mobility as well as electronics like wearable devices still represent a potential high-volume as well as high-quality market for GRM, especially in LIB anodes, incorporating the properties of graphite and facilitating silicon integration. Both markets are relevant sectors for next-generation batteries and one of the most promising graphene applications [35], and were therefore selected for the end use and market sphere as displayed in figure 1. This chapter examines the requirements and commercialization prospects for GRMs in these application examples.

In order to make EVs (and other applications) more competitive, battery cell prices below 100 € kg^{-1} [38] and long driving ranges are required, often given by the energy density of batteries. The energy density on cell level continuously increased since the 1990s and reached the range of 150–250 Wh kg^{-1} (200–500–230–700 Wh l^{-1}) in 2017 [38]. This affected the driving ranges of few to several hundred kilometers in a positive way. Besides cost and energy density, the performance of a cell in terms of fast charging capability was a third main driver that has to be addressed by LIB technology development [39].

In contrast to EVs, batteries for (wearable) electronic devices also need to fit into functionalized designs geared towards physiological circumstances like the wearing convenience of headphones, electronic bracelets or watches. Of special interest for small in-ear headphones was the rechargeable LIB coin cell, which are intended to replace the primary batteries commonly used at the time. Long operating hours and high energy density for flexible use are requiring a lightweight and small battery inside the application. Due to the difficulty of replacing batteries in devices, the life expectancy of the installed LIB is of great importance with respect to a sustainable product lifecycle.

The strong volume changes which advanced anode materials exhibit during cycling, must not be translated to cell level. The overall volume change inside a cell must not exceed 5%, as this is seen as the safety range in LIB manufacturing [33]. Hence, there was a discussed limit to the use of the Si amount, with a threshold of <12 wt% for maximizing the volumetric energy density [33]. Several approaches were followed to mitigate or overcome the problems of Si swelling and poor cycling stability. Si nano-particles for example behave differently than bulk material and show less particle fracturing during cycling [40]. The manufacturing process of respective nano materials is however costly, which is why there were efforts to use Si micro-particles as anode material [41, 42].

The utilization of silicon/carbon composites can reduce the volume expansion of the electrode as whole, if a porous carbon structure is used, which can accommodate the volume change of the Si-particles during alloying [43]. In order to overcome the issues of Si/electrolyte instabilities, associated to the repeated breaking of the SEI during cycling, more specific requirements to the carbon structure are called for. Following several models proposed in the literature [43], a carbon wrapping around Si-particles was discussed to be a means to form a protective intermediate layer between electrolyte and Si-active material, thereby ensuring a stable SEI on the carbon surface. These requirements either call for a porous carbon structure incorporating Si-particles or for carbon particles with lateral dimensions comparable to those of the Si-particles (typically few 100 nm for nano-particles or around 1 μm for micro-particles) which can be wrapped around the Si-particles [26]. In addition, our expert consultations revealed that Si-particles must however not be completely shielded in order to function as anode material. Good electronic and ionic linkage to the current collector and electrolyte respectively are necessary to provide for adequate reaction kinetics during the alloying reaction. Graphitic particles with many carbon-layers would however not facilitate Li-diffusion to the surface of Si-particles, since Li-transport perpendicular to the layers is inhibited.

The cycling stability of silicon–carbon anode materials can greatly improve with a combination of carbon materials with outstanding mechanical stability and high electronic conductivity [44]. GRMs

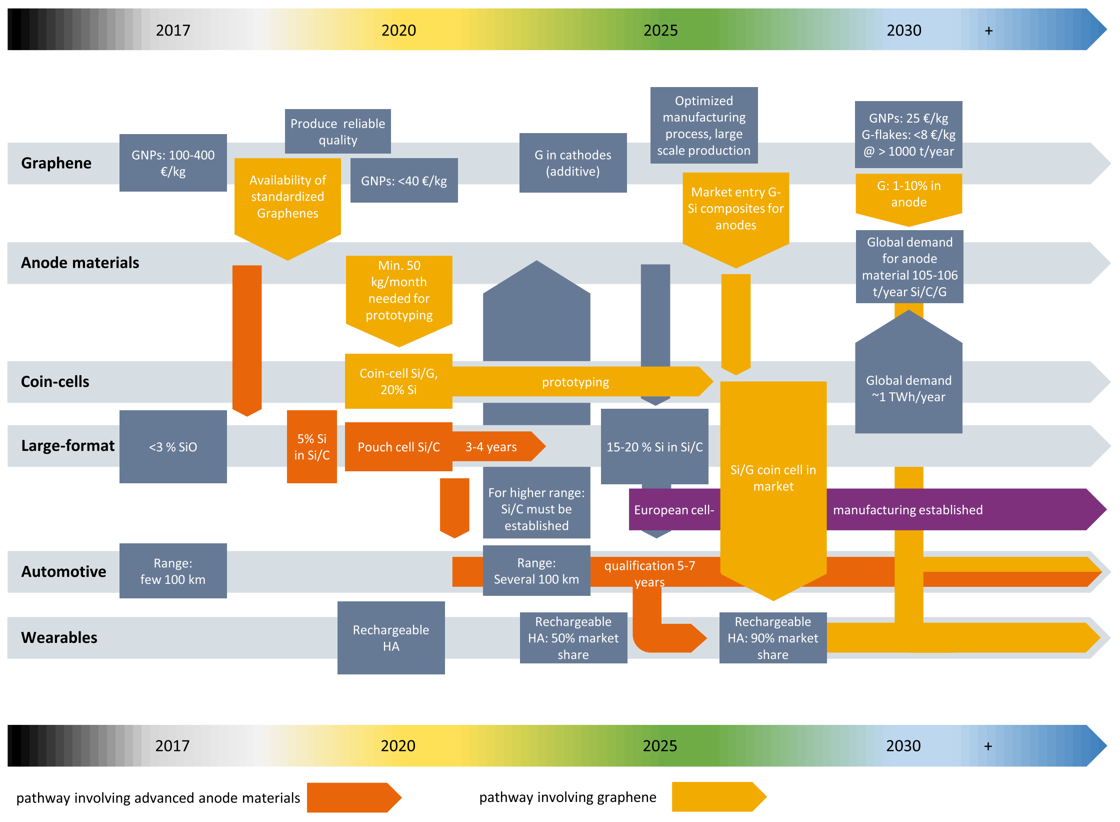


Figure 2. Graphene Li-ion battery roadmap 2017, adapted from [37]. CC BY 4.0.

were therefore discussed as a promising solution to overcome those challenges. Few-layer GNPs could meet the requirements for both, lateral size and electronic and ionic conductivity [45]. Due to the high aspect ratio and mechanical stability, GRM is of high promise to accommodate structural changes inside the electrode (volume change compensation) and at the same time serve as an effective and protective intermediate layer (chemical 'shielding') between Si-particles and the electrolyte [31].

Besides serving as enabler for Si-based anodes, G, rGO or GO materials can also be utilized as host materials for Li-ions themselves. Depending on defects and their chemical composition or surface groups, the experimentally observed capacity can even exceed the intercalation capacity of graphite [44]. Due to its low initial coulombic efficiency [46], no industrial activities were known where pure graphene would be tested as active material.

Figure 2 shows the initial 2017 roadmap for the introduction of graphene-enhanced silicon anodes in LIBs. The roadmap maps the commercialization perspective for a possible implementation starting at the material level (GRM, anode materials) with impacts on components on cell level (coin-cells, large-format) and finally takes on the application perspective (automotive, wearables). A possible pathway involving GRM in batteries is colored in yellow boxes and streams. Standardized graphene, produced in reliable quality, is needed for prototyping in coin cells and later in large-format cells (direct interface). Experts expected this to take 5+ years until after 2025. If the cells prove themselves economically and technically, they could end up in use after a further qualification and development period of 5–7 yrs (indirect interface). The graphene industry would have to master all scaling challenges by then. For Europe, the establishment of cell manufacturing was discussed as crucial to link the materials and components innovation to an application end use market. Within the 3I approach, we identified four key aspects for a successful implementation of the depicted pathway in figure 2:

- GRM maturity for batteries

For any prototyping and industrialization, a consistent supply (kg-scale) of graphene with reliable quality is mandatory. Besides particle morphology and properties, the delivery form of graphene is important. The establishment of quality standards and standardized GRM and silicon–graphene (Si/G)-compounds are considered to be of utmost importance for the further development of any graphene markets. In 2017, it was expected that standardized graphene and consistent supply for cell prototyping (in addition to further research) can be established by 2022 at the latest.

- Components and benefits

With respect to anode materials, a continuous increase of Si-content is aimed on. The prototyping phase for respective anode material compositions is typically of a duration of 3–4 yrs. In parallel to other advanced anode materials, Si/G coin cells with roughly 20% Si content were discussed as a key for further prototyping and testing. A respective cell would feature an energy density of about 350 Wh kg^{-1} and 850 Wh l^{-1}. Whereas large-format Si/C pouch cell prototyping with a low Si amount of about 5% might need only few years to reach application qualification in 2022, Si/G coin cells with a higher Si share might need a longer prototyping time until it can reach further qualification in the coin cell market, for instance in wearable devices, until 2026.

- Application markets

The automotive industry will be a main driver for Si/C anodes to address its required performance parameters, for instance range increase and cost decrease. However, it was expected that Si/G anodes will be first introduced in small consumer devices, discussed on wearables applications in the 2017 roadmap, as a strong shift from non-rechargeable batteries to rechargeable LIB were expected to start in the 2020s. A market share of 50% of rechargeable LIB in headphones (e.g. hearing aides, in-ear pods) after 2025 was expected as a first relevant market and therefore a market pull for graphene enhanced batteries.

- GRM price and scale

Optimization of manufacturing processes and large scale production for GRM are essential. In 2017, this were discussed as the basis to enter a larger market in a mid- to long-term perspective, especially as an amount of 1%–10% of graphene in the anode might be possible. Instanced, prices for GNP were expected to decrease from >100 € kg^{-1} in 2017 to <40 € kg^{-1} until 2022, with large scale production might even be able to tighten prices for GNP at 25 € kg^{-1} at >1000 t per year and increase the overall cost-competitiveness. It was unclear whether this price would be cheap enough to compete with common carbon additives. It was also unclear to the time whether common carbon additives like CB would really be the benchmark for graphene, since graphene can provide different functions. In 2022, it was prominently discussed that there is still a gap between experimental production for research and large-scale industrial production [47].

Discussed in 2017, required performance improvements for relevant applications as well as GRM standardization and general cost reduction might foster an industrialization pathway for graphene in LIBs, with a possible 1%–10% share of graphene as anode material in 2030. The pathway involving graphene is, however, influenced from the direct as well as indirect interface developments in the timeframe displayed in figure 2. The pathway of advanced anode materials in large-format cells may parallel that of graphene from 2020 onwards. Given that graphene is a novel material for LIB electrodes, it was anticipated that the latter will undergo a more protracted phase of prototyping and qualification. Successful prototyping and large scale material production is a necessary step for a market entry of Si/G composites and their application in Si/G coin cells, pinpointing to applications such as rechargeable wireless headphones.

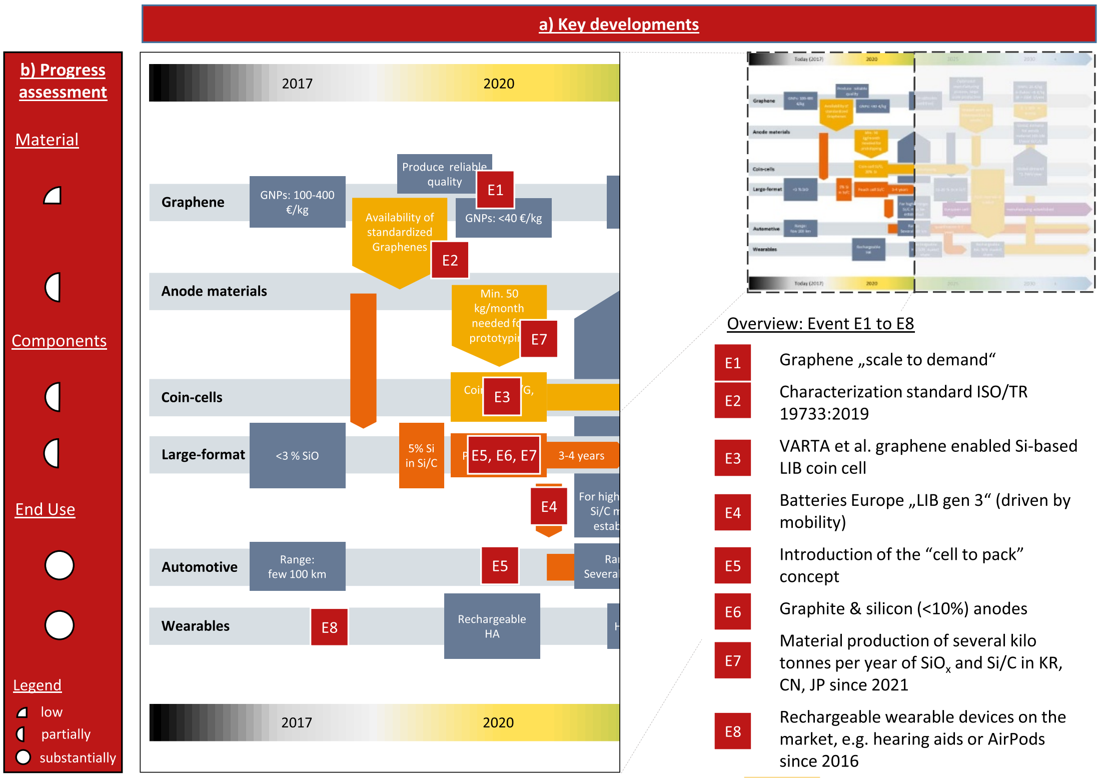
4. Progress report: 2017–2022

Several developments and intensive research efforts mark the five-year period between the two roadmaps, from material industrialization [3] to the battery technology itself [10] as well as the application markets [7]. Figure 3 emphasizes from the top downward (a) key developments between 2018 and 2022, and on the left side (b) a progress assessment of the innovation spheres from the initial value chain approach in figure 1. Each red box on the roadmap excerpt in figure 3 represents an notable event, displayed as E1 to E8, and will be discussed in the following subchapters, whereas the progress assessment at the end of chapter 4 will link to the 2022 roadmap.

4.1. LIB technology

In the 2017 roadmapping, the focus was mainly on technical performance indicators such as *energy density* for driving range or *fast-charging capability*. In 2022 battery costs and sustainability issues (e.g. battery lifetime, carbon footprint) in battery production have become much more important in general discussions [10, 18, 48]. Falling costs to well below 100 € kWh^{-1} were assumed for the future in 2017. Such a value may also have been reached for selected automotive cells in 2020. Since then, however, the costs of battery raw materials in particular have fluctuated significantly. Due to large production capacities in China along with an EV market growing less strongly since 2023 than in previous years, todays LIB cells are traded at a record low of below 100 € kWh^{-1} [49].

From 2017 to 2021, the energy density on cell level increased from approx. 150–250–200–250 Wh kg^{-1} (250–700–300–700 Wh l^{-1}) in the main cell formats [50], especially with a strong increase in the pouch cell format with nearly a factor of two (mean value 250 Wh kg^{-1} and 600 Wh l^{-1} in 2021). On a technological perspective, the target value of 350 Wh kg^{-1}



E1	Graphene „scale to demand“
E2	Characterization standard ISO/TR 19733:2019
E3	VARTA et al. graphene enabled Si-based LIB coin cell
E4	Batteries Europe „LIB gen 3“ (driven by mobility)
E5	Introduction of the “cell to pack” concept
E6	Graphite & silicon (<10%) anodes
E7	Material production of several kilo tonnes per year of SiO_x and Si/C in KR, CN, JP since 2021
E8	Rechargeable wearable devices on the market, e.g. hearing aids or AirPods since 2016

Figure 3. Progress report 2017–2022. (a) Top side: mapping of key developments to the roadmap 2017–2022 in form of event E1 to E8, (b) left side: progress assessment of each innovation sphere from the initial value chain sketch.

for gravimetric energy density in 2025 stated in 2017 appears very high as the next development step for automotive industry. Currently, work is being done on LIB cells with around 300 Wh kg^{-1} [51]. Commercially, these had not yet been broadly available in 2022. At least, some current announcements address the targeted value for gravimetric energy density for today or the near future mentioning semi-solid LIB cell technology [52, 53]. Especially the so called C2P concept (figure 3, E5), with large LIB cell formats integrated directly into the battery pack and therefore neglecting the intermediate module integration [10], affect the energy density and driving range in a positive way without incorporating silicon or GRM. This innovation, now already commercialized by large Chinese OEMs [54, 55], reflects the optimization of battery technology from a components and end use engineering point of view.

Battery cells with a certain SiO_x content were already available in 2018. Since then, this material innovation has improved and diffused to market further [13] (figure 3, E4 and E6). SiO_x seems to be continually used in a small share in blends and accordingly offers specific anode capacities that slightly exceed the graphite benchmark of 330–350 mAh g^{-1}, e.g. 400 mAh g^{-1} [13, 56]. Si/G coin cells with small amounts of graphene show very promising results and have been positively demonstrated in research [57] (figure 3, E3). In addition to the possibility of continuously increasing the Si content, concepts have emerged in recent years that provide for a direct transition to Si dominated anodes. This developed quite well in recent years and first Si-dominated and production-ready cells are announced [58].

For silicon dominant anodes, it is expected that high energy density cells with fast charging capability and high cycle stability (>1000 cycles) are close to a market readiness, for instance see the announcement of Israeli company StoreDot [59].

Since 2017, massive progress has been made in building European cell manufacturing facilities. Several European and non-European players are building giga-scale factories, complementing the landscape of existing specialty battery cell suppliers. Thus, from an industrial landscape perspective, commercialization of new technologies in Europe appears possible at all scales. However, it should be emphasized that some factories are still being built and certainly some time will be needed after the start of production to optimize established technologies before newer players can introduce new technologies. Especially the increasing activity of Si and SiO_x producers [21, 23] might support further activities in a mass production environment (figure 3, E7) for commercialization.

4.2. Markets: EVs and consumer electronics

The market diffusion of EVs evolved much faster than expected in 2017. Until 2022, many international

OEMs have announced their intention to discontinue the development of ICE technology in the medium term, at least for passenger cars, and to convert their entire product range to electric drives. Especially the European Union's commitment to phase out fossil fuel driven vehicles by 2035 [60] may boost BEV market diffusion further. The offered number of models of EVs is growing very quickly and driving ranges of $>$300 km are no longer uncommon. In the premium segment, vehicles with a driving range of over 600 km are occasionally offered [61].

In the field of smaller battery applications, many body-worn electronic devices have since become established in the trend of wearable electronics (figure 3, E8). True wireless headsets (TWS) have displaced classic wired headsets in many areas. New smartphone generations, for example, no longer have a headphone jack and provide for headphone use via Bluetooth. In general, the requirements for TWS and other consumer electronics are high. The devices must be lightweight and must offer a long service life. Especially the permanent use of Bluetooth and the requirements for high availability and thus fast charging capability result in high requirements on the battery. An exciting perspective also arises for the consumer electronics market, where research is being conducted on flexible and stretchable materials [62] that could, for example, advance printable batteries or new product designs with fully integrated batteries. For electronic devices, harsher conditions (e.g. bending or twisting in consumer electronics) facilitate electrodes with flexible substrates, e.g. CNT, rGO film or graphene monoliths [63].

4.3. Graphene: price and function

In 2017, the perspective on GRM prices was strongly influenced by the reference to the benchmark graphite. Natural graphite is available at a price of around 10 € kg^{-1} [64]. From the perspective of graphene as enabler for a new technology, e.g. Si-anodes, the price for graphene can be higher and based on the function and benefit in the overall system. The prices for GNP predicted in 2017 of below 100 € kg^{-1} have not been reached. These are still in the range of several 100 to several 1000 € kg^{-1} [64]. Suitable graphene platelets can currently cost several 1000 € kg^{-1}. The specification of target costs did not seem possible in 2022, especially as there are still uncertainties about the necessary proportion or weight share (wt%) of graphene in Si anodes [34], e.g. whether $<$1% [57] or up to a double-digit share (see figure 2) is needed, and about the quantification of the additional benefit of graphene. Hence, there is a discussed limit to the use of the Si amount in the anode with a threshold of 5–10 wt% [18] optimized for high charge rates and cell aging, which is lower than the initial discussed threshold of $<$12 wt% [33], see section 3.

Besides the discussion in research on the theoretical best Si amount, several industry activities and announcements indicate that higher amounts of Si are feasible and already in testing operations, with an eventual jump from low ($<$5 wt%) to mid ($<$20 wt%) to high ($>$50 wt%) Si content [23]. This is also reflected in a 'scale to demand' supply of graphene which addresses not only the quantity but mainly the quality as a functionalized GRM product [3] (figure 3, E1), which in turn should align with cost-effective production of GRMs on industrial scale [65].

Likewise downscaling of the particle size, see section 3, partial use of the maximum available capacity of silicon is another way to limit particle volume change and hence fracturing [66]. This method decreases the specific capacity of silicon, which however is still significantly higher as compared to graphite, even if only half the theoretical capacity of silicon is used. For both approaches, Si nano- and microparticles, the volume change on particle level remains high. Material pulverization, morphology change and continuous SEI growth remains as fundamental challenges for Si anodes in LIBs [43, 67, 68]. The graphene to silicon ratio indicates different performance improvements [34], which could be different for individual cell manufacturers or application OEMs targets. This indicates that the use of tailored materials may be more relevant than a moreless commodity product, as the interface between graphene and electrode materials critically influences ion diffusion and electron transfer, which can be optimized through composite designs or coatings. Morphology, layer number, and defect density directly affect graphenes performance [69]. Considering the structure-performance relationship, tailoring these structural parameters is there for key to developing graphene-based materials for high-energy and high-power LIBs. Although standardization is already being addressed by ISO/TR 19733:2019 in the ISO229:2019 [70], it primarily concerns the methodological approach for characterizing and analyzing nanomaterials such as GRM, but not a standardized product itself (figure 3, E2). The various developed techniques in the synthesis of graphene [71] result in variations in material composition and, consequently, correlates to the structure-performance characteristics for graphene-enhanced LIB [72]. For instance, CVD can be utilized to produce high-purity, large-area graphene with excellent electrical conductivity, whereas various exfoliation approaches, on the other hand, facilitate a controlled layer thickness but with low yield [73]. The manufacturing processes thus exert a profound influence on process scalability and critical material characteristics [74], including defect density, layer number, and purity, which in turn affect the electrodes properties such as conductivity, ion diffusion, and mechanical stability and the electrochemical performance of LIB anodes.

Interest seems also to be on graphene as an additive for electrodes in order to utilize primarily the improved thermal and electrical conductivity [47, 75]. Additionally, graphene as a coating material for LFP cathodes, which are being progressively used as *x*EV batteries at the latest [64], show improved performance and enhanced conductive network [76]. This is beneficial for automotive application requirements in terms of battery charging and application acceleration (discharge) performance improvements.

In the consumer electronics sector, some battery requirements for applications tend to be less rigorous than in the automotive industry. The trend in consumer electronics is to use embedded, customized pouch-type LIB-cell designs as they can be easily customized in size and shape. New materials like graphene or GRM might have the potential to increase the energy density on a minimalistic pack scale, extend the lifetime of the battery and address safety issues and environmental adaptability [63]. Especially for next-gen wearable devices, the battery is facing mechanical stress as they are worn directly on the human body. This adresses materials with good mechanical properties in particular and stricter safety and high economic efficiency [63].

4.4. Progress assessment

Graphene, as one innovation sphere investigated, must be considered in a differentiated way for its use in batteries. On the one hand, there is the generic view of graphene as a carbon material or product that can be available on a large scale. On the other hand, however, the use of high-quality graphene to fulfil specific properties, particularly in the anode of a battery, must be taken into account. The progress assessment in figure 3, left part b), shows the substantially, partially and low progressed indicators in each innovation sphere. The material systems in particular show that the expected price and volume development has not (yet) materialized and that ‘standardized’ graphene reflects more to nanomaterial characterization than consistant availability in supply [3]. Standardization especially pinpoints to application specific materials standards in terms of characterization [70] and the *in situ* quality control to increase the reliability of supply from the ‘Graphene zoo’ of very diverse actors and graphene products [3].

Also, the availability of sufficient quantities of advanced anode material for prototyping could not be adequately answered, especially as Si-based anode material production is mainly based in Asia which might be more difficult to source for prototyping in Europe. Both coin and pouch cells with increasing Si content were central topics in research and industry along our expert consultancies. The use of rechargeable LIBs in wearables like hearing aids, wireless headphones, smartwatches etc. is already increasing before 2020 and the range of EVs has raised continuously, particularly due to improvements in LIBs. The increase in range (and the associated increase in the energy density of the cell and pack) was not only due to an increasing share of Si in the anode or Ni in the cathode and the improvement of all other components, but is also attributed to engineering efforts, for instance the cell to pack concept in the form of large-format blade batteries from BYD or Qilin batteries from CATL—both global market leaders in battery technology.

Promising approaches for Si-dominant anodes are reducing the surface area of silicon particles, surface carbon coating to improve electronic contact and pre-lithiation to compensate for lithium losses, which seems to be in particular promising for increasing the cyclic lifetime of the full cell [66]. The incorporation of graphene into the anode is still seen as graphenes most promising application in the LIB technology [77]. Thus, the specific properties of graphene lead to optimistic expectations that GNP could be one of the new battery material innovations in the near future.

Graphene could meet the requirements for both, lateral size and electronic and ionic conductivity of a shielding material, as graphene coated silicon is able to better handle the mechanical stress induced during cycling compared to graphite-coated silicon [78], still the potential of graphene to enhance the structural integrity of electrodes is challenging, i.e. for flexible batteries [79]. Nevertheless, the fundamental lithium storage mechanism and configuration optimization with respect to safety, performance and sustainability were subjects of ongoing research in 2022 [34, 77], accentuating the prototyping activities for Si/G coin cells in figure 2.

5. The revised roadmap 2022

In light of the significant advancements since 2017, we undertook a comprehensive revision and expansion of the graphene battery roadmap. While the general structure of the projected future value chain remained largely consistent in 2022, we made notable adjustments to align it with the latest developments. The same spheres, interfaces and markets as in the 2017 roadmap were addressed in the 2022 roadmap, as displayed in figure 2. The general requirements of the application areas considered have been discussed in sections 3 and 4. The roadmap 2022 goes into more detail on specific developments of interest, especially application-relevant battery KPIs. In particular, LIB charging capability, related to fast charging, and the energy density are the KPIs that are in focus. The application-related considered energy density to this time are displayed in grey boxes on the roadmap.

Figure 4 shows the revised roadmap 2022. A possible pathway involving graphene in batteries is colored in yellow arrows and boxes. On the material side, a differentiation has been made for the graphene pathway, between the large scale application in batteries, which is still unclear, and as a high-quality material that is suitably incorporated into the battery because of a distinct need.

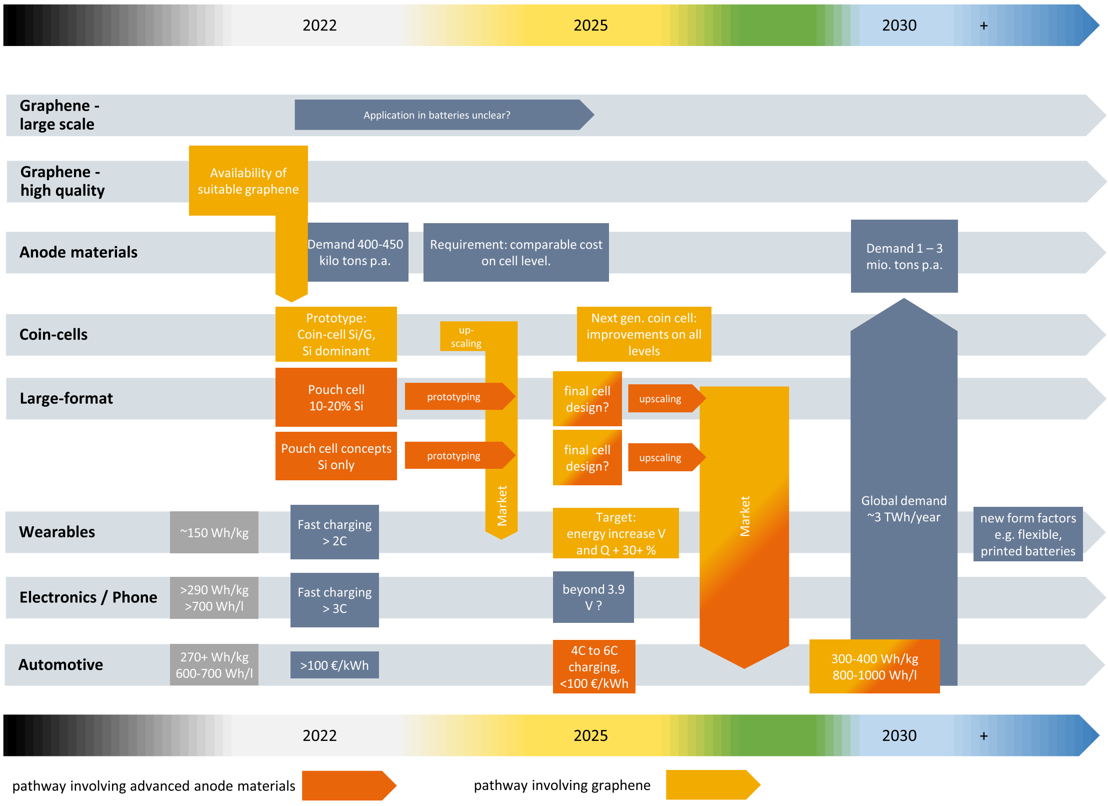


Figure 4. Graphene Li-ion battery roadmap 2022 (revision).

The term 'standardized graphene' was removed from the roadmap 2017, as this received only limited consideration in the 2022 expert consultation. In lieu of this, the term 'suitable graphene' was emphasized, as it more accurately reflects the necessity of matching the graphene material properties with the application-specific requirements of the LIB, with its Si-based anode respectively. Nevertheless, to enforce standards of graphene, we do not want to neglect standardization, as this pinpoints to *in situ* quality control as well as application-specific material standards in the upcoming GRM industrialization [3, 70], which was an important point in the overall discussions.

Generally in 2022, battery-grade quality graphene was perceived more as a useful high-quality material for fulfilling specific functions than as a stand-alone anode material. Mass-produced graphene does not seem to be the material of choice back then, as this is in competition with other advanced carbons or nanomaterials that are already more cost-competitive and produced in large scales. Whether graphene or other electrode additives, which can also enable Si-anodes, will be used commercially, is not clear yet. The yellow-orange arrows on the roadmap 2022 therefore symbolize that there may or may not be an involvement of graphene in battery technology development and industrialization.

Therefore, the 2022 roadmap in figure 4 portrays a somewhat different diffusion pathway than in 2017. Contrary to 2017, graphene rather serves to enable a new technology like Si anodes, therefore the price for graphene must be based on the function and benefit in the overall system and not only on its competitor graphite. In addition to the concept with only a small share of Si, we add the Si only concept to the roadmap with respect to current industry discussions [17, 58]. Prototype testing on large-format cells in parallel to the scale up of coin cells from research to market are of greatest importance in the next 2–4 yrs, analogous continuing the timeframe of 5–7 yrs considered in 2017. If the cells prove themselves economically and technically, application might happen after a further qualification and development period of several years.

Experts assume a duration of 3–4 yrs between a finalized battery design and its use e.g., in cars, which results from vehicle development and battery qualification. The time required for prototyping has therefore not changed significantly compared to 2017's perspective. The market launch assumed in 2017 between 2025 and 2030 for next generation Si-based LIBs appears possible from 2022's perspective, and market launch with prototype coin cells in initial applications i.e. small electronic devices or consumer electronics, was even assumed until 2025.

Our desk research and expert consultation have led us to the following key aspects regarding the commercialization perspectives for graphene in LIBs in the revised 3I roadmap 2022.

- GRM maturity and scale

Investigations on GRM commercialization particularly pinpointed towards the insufficient maturity of the GRM industry representing a major threat and critical bottleneck for its commercialization [3, 13]. The supply inconsistency was identified as the single most critical factor limiting the overall progress of GRM industrialization. The industrialization progress between 2018 and 2020 can be described best as a 'consolidation phase' [3], with little quantitative supply increase but significant qualitative improvement. Experts both expect and hope for a lead market resolution, with an initial volume market to drive the overall industrialization of GRM into a comprehensive materials innovation system by 2030. In other cases, GRM industrialization might fade into a general nanomaterials regime e.g. similar to CNTs, with different market strategies, i.e. co-developments, tailored supply stream or strategic dumping [3]. As performance and price are important battery market requirements, GRM could be competitive as a future battery material [47], still barriers exist, for instance, a reliable supply concerning quality and quantity [13].

- Technological benefits

Results from academic and industry research to date indicate that graphene can increase the performance of Si-based anodes. In the overall system between Si material and additives, increases in energy density of about 30% compared to the state of the art are possible and demonstrated. Whether this is sufficient to meet the high target values on OEM product and technology roadmaps is unclear. In terms of KPI, improvements to 300–400 Wh kg^{-1}, 800–1000° Wh°l^{-1} by 2030 seem very ambitious, but not unlikely. Simultaneously increasing fast charging capability to 4 °C or 6 °C while maintaining the highest energy density appears almost impossible but could be a chance for graphene as a multifunctional material. The cycle life of Si-based anode in LIBs is in the range of several 100 cycles, which is acceptable for many electronic applications, but may not meet the requirements in the automotive sector yet.

- Application markets

On the difference between an incremental increase in Si content to a 20 wt% maximum and a direct transition to Si-dominated, the introduction of this anode technology in the automotive sector may be more likely to be driven by an incremental increase in Si content. Si-carbon composite anodes could be more interesting for automotive applications in the next few years than Si-dominant anodes. The primary goal of employing advanced anode materials is to further enhance fast-charging capabilities, increase energy density, and concurrently reduce costs as demanded from the end use markets.

- Competing technologies

In parallel, increasing activity from industry and research are observed in general for high performance or advanced anode materials. Open questions about how a final cell design will look like should be answered by 2025. It seems most likely that GRM will be in competition with other advanced materials in the coming years, for instance graphene-like carbon-based anode materials [46]. The quantitative availability of high-quality graphene and the associated high costs must be critically questioned. In addition, alternative additives or material compositions are also being discussed especially for silicon anodes, which seem to be promising and therefore have to be seen in direct competition with graphene-enhanced materials. Promising results have been achieved for SiO$_x$ or Si/C composites even without the use of graphene. For the practical use of silicon anodes, testing in practice, in particular regarding stability (lifetime) and electrolyte compatibility, continues to be an important aspect in the commercialization of Si anodes in general [80].

With respect to other battery technologies, the time window for the introduction of graphene in Si/C composites might also be limited. Particularly for automotive application, a gradual transition towards Li-anodes and all solid-state batteries is predicted once the technology has reached the required maturity (2030+) [81]. This would again reduce the demand for Si/graphene anode materials. Noteworthy, the use of graphene-based materials for SSBs is already being discussed [82] and could be an opportunity for the industry besides the Si-anodes.

- GRM price

The prices of GRM significantly depend on its quality. A relatively low material content of the anode allows for higher prices for graphene to be accepted, particularly in comparison to low-cost graphite, provided that the desired utilisation of the material can be achieved. Despite the typical association of CVD graphene being way too expensive for high volume and rather low cost applications such as batteries, some graphene providers work on appropriate production schemes. This may be the case with high-purity graphene, which is integrated in 1–3 wt% into the Si anode as an additive, provided that significant performance improvements can be achieved than it is the case with low-cost and less-pure graphene with a double-digit wt% in the anode. Cost competitiveness of battery cells still remains an issue for the application of graphene. For a commercialization of course, graphene with specified properties and quality will need a price tag, today and in the future. Improving a batteries performance might only allow for a limited increase in material costs, especially for price sensitive mass market applications.

- Scale and production

In 2017, it had already become apparent that the automotive industry requires high volumes of battery-related materials. Figures from the roadmap 2022 highlight this: The demand for anode material was in the range of 400–450 kt in 2022 [23]. In 2017, total global demand for LIB was still estimated at 1 TWh in 2030. More recently, it is more likely to be 3 TWh in 2030 or even higher [10]. The 1000–3000 kt per year of anode material demand resulting from this volume implies a demand from automotive industry of several 1000 t graphene per year, even at less than 1 wt% graphene in the anode. This market size can be a threat for graphene producers. Providing the high quantities needed in LIBs can be challenging for small and medium sized companies. A market introduction will only happen, if cell manufacturers are guaranteed reliability of supply. Still, standardized characterization of the material purity, product quality and consistent supply on the other hand are issues that graphene producers have to struggle with already today, although production volumes are rather low. The ISO/TR 19733:2019 can enable trust to be built up and joint development to be driven forward.

From an industrial landscape perspective, commercialization of new technologies in Europe appears possible at all scales. Cell and material production in Europe is at the scale-up phase and although several players have announced to build up capacities, many of them will not be producing for several years. Particularly the limited amount of anode manufacturers in the EU [10, 21] might be a threat for local graphene production and value creation. In turn, from the perspective of materials supplier in general, the establishment of a cell manufacturing industry would yield the highest potential for their growth. If a commercialization of GRM enhanced materials, likely for Si-based anodes in LIB, is possible, it will be a strong driver for the graphene market in general, still graphene industrialization is just at the beginning, especially in Europe [3, 4].

6. Conclusion and outlook

Our graphene Li-ion battery roadmaps aggregate essential stakeholder perspectives along the emerging value chain for GRM-enhanced battery technology. As a form of futures-oriented reasoning [83] it explores an optimistic yet realistic timeline toward successful commercialization balancing the contemporary insights and expectations of key stakeholder groups. The comparison of the initial 2017 roadmap with its 2022 revision (a) highlights the progress achieved along the value chain, (b) reveals novel developments beyond the initial expectation horizon, and, thus, (c) also provides us with deeper insights into the capabilities and limitations of roadmapping.

6.1. Technological progress

Within the span of 5 years, tremendous progress has been achieved all along the potential future value chain under exploration. Among these, we recognize two overarching technological achievements that shape the trajectory of the revised 2022 roadmap.

On the one hand, suitability of graphene for improving Si anodes has been clearly confirmed in the meantime. Even small amounts of graphene can have a positive effect on the stability of silicon anodes, even if the cyclic stability does not yet come close to state of the art graphite anodes. Ample evidence demonstrates the potential of graphene to improve key performance indicators of batteries such as energy density or fast charging capability. So the 2017 vision of a GRM pathway into the LIB value chain as a part of an advanced anode material (as a highly functional additive within a Si/C/G composite) gained substantial credibility.

On the other hand, the battery industry also achieved tremendous progress in many performance characteristics. Here, incremental innovation drives LIB technology towards continuous improvements, in particular with regard to optimization of the existing material systems and application oriented cell format engineering. Hence, the bar for wide-spread diffusion of advanced material incorporation also raised in the meantime and no immediate market pull for a GRM-enhanced solution has emerged yet.

6.2. Commercialization prospects

Beyond technical progress, broader global developments impact overall battery market and GRM commercialization perspectives in this context. In particular, the emergence of a domestic LIB manufacturing industry in the European Union turned from a mere vision on the 2017 roadmap into a concrete trend setting the context for its 2022 revision. Varios insights of industrial and political stakeholders continue to fuel that trend, including (a) a perception change in the automotive industry (now considering batteries as an essential component instead of a mere external supply item), (b) the realization of the fragility of global supply chains as experienced during the Covid-19 pandemic, and (c) more systematic consideration of technological sovereignty and critical technologies by European actors in the context of geopolitical developments.

The ramp-up of the European LIB cell production constitutes a key enabler for graphene incorporation in the battery value chain based on domestic R&D progress. Of course, that does not immediately create a significant market for graphene right away but offers improving perspectives for European actors to enter the emerging value chain. In particular, silicon incorporation into advanced anode materials might constitute an attractive opportunity for overall graphene commercialization. Here, technology diffusion driven by crucial performance characteristics might coincide with market diffusion forces such as (a) overall market expansion (due to widespread BEV adoption), (b) increasing competition (creating pressure on prize and performance levels), and (c) societal level challenges (decarburization, geopolitics). Once adoption of enhanced Si/C/G LIB anode materials starts, large purchase volumes of the automotive industry would certainly pose substantial scaling challenges to graphene producers—and thus trigger an economics-of-scale trajectory eventually enabling a wider overall diffusion of GRM based on competitive cost and ample availability of high-quality materials.

Beyond establishing LIB cell manufacturing in Europe, this optimistic vision requires the development of a domestic battery production value chain upstream to materials suppliers, in particular for Si/SiO_x and GRM. At present, cell and material production in Europe is still in a scale-up phase. Several players already announced to build up capacities, but most of those are not in production for several years still [10, 84]. The limited number of domestic anode manufacturers may constitute a bottleneck for European graphene commercialization in the battery context.

In essence, the 2022 roadmap paints a more detailed picture of the commercialization perspectives of graphene-enhanced batteries compared to 2017. As the battery industry largely reaches its target KPIs also without advanced material incorporation so far, only limited market pull forces emerged and quick pathway to widespread GRM diffusion did not materialize. The principle application case for graphene in LIB anodes actually gained substance though. Its proven benefits open up credible niche diffusion pathways. The coin call market, for instance, values specific performance characteristics higher than pure price sensitivity. Industry announcements still indicate the continued use of Si/C composites in the coming years, but research results demonstrate the application case for Si/G in the coin cell format. Once these transition to industry, this may form a promising commercialization base for graphene in advanced LIB anode designs.

Large volume application cases of graphene in the battery context (e.g. as replacement or conductive carbons) have not emerged in recent years. However, specialized use for specific functions and technologies seems reasonable and likely. This is also reflected in substantial transnational patenting activities. The development of graphene in batteries shows significant regional variations, with East Asia leading due to its industrial dominance in battery technology and a strong focus on graphene applications, as reflected in patent activity and market reports [4]. While Europe lacks global leaders in battery IP, it excels in the second tier with notable players and specialists, though Asian and U.S. companies stand out with a higher integration of graphene in their battery patents. This reinforces the expectation of a possible market entry for graphene in the coming years.

6.3. Roadmap approach

As an explorative foresight measure, any 3I focus roadmap balances innovation prospects throughout an emerging value chain at a given point in time. Key stakeholder perspectives and their interactive alignment constitute its primary source. And these perspectives change over time, when unforeseen developments affect the field—or crucial interaction across innovation interfaces does not take place in the required extent. Here, the graphene battery roadmap has been a highly valuable case study in multiple regards.

On the methodology perspective, we have established a framework for the revision of a 3I roadmap that reaches far beyond mere repetition. Building on the subject expertise established in the initial 3I campaign, we systematically track and evaluate key developments that occurred in the field in the meantime. This adds further substance to the consultation process. Here, we deliberately target a healthy mix between experts considered in the earlier campaign and freshly approached actors, with particular regard to the occurrence of novel developments.

With regard to the predictive capacity of the exploratory 3I roadmap approach, the highly dynamic battery topic provides us with particularly interesting case study where multiple influences and uncertainties intersect. On the one hand, the ramp-up of a European LIB production merely constituted an optimistic vision in 2017. Yet one seen beneficial for domestic commercialization of GRM-enhanced battery technology. Hence, its inclusion on the roadmap (for the mid 2020s) defined the shortest conceivable pathway. Both expected (diffusion of electric mobility; restructuring of the automotive sector) and unexpected developments (supply chains strained by the Covid pandemic and geopolitical tensions) basically drove the adoption of that path on both political and economic dimensions. Recent announcements indicate some delays to be expected, but overall the emergence of European LIB manufacturing is much clearer today than in 2017.

However, the broad diffusion of graphene in the battery sector is yet to come still. In this context, the desirable properties of graphene actually substantiated, but commercial adoption lags behind. Both, the rapid diffusion of electric mobility and the advancement of conventional LIB technology exceeded earlier expectations. The success in the regime of the incumbent technology thus diminishes the immediate prospects of graphene commercialization in the battery sector which still remains at niche status as elucidated by the 2022 roadmap revision.

Overall, the results clearly demonstrate both the capability and limitations of technology roadmapping according to 3I principles. The balanced aggregation of expert expectation along the emerging value chain enables the exploration of a realistic pathway towards desirable technical progress way beyond individual stakeholder expectations or commercial market report predictions. The predictive capacity of any 3I roadmap result, however, critically depends on both the materialization of the required stakeholder interaction and the impact of unexpected events on the prospects. Thus, regular 3I roadmap revisions promise to track changes—while also driving desirable stakeholder interaction along emerging value chains.

6.4. Recent developments

The European Commission evaluated the anticipated application of silicon–graphene composites for batteries positively in assessing the last 10 years of the Graphene Flagship [85]. However, other application areas are considered to be more relevant for the general industrialization of graphene now, while a long-term expansion of R&I activities and close cooperation with the private sector and other EU initiatives is considered to be essential for the industrialization of graphene [85].

Recent industry activities, with more than 165 companies engaged in the development of graphene batteries [86], highlight the uncertainties surrounding the commercialization ramp-up and the potential role for graphene in battery technology. For instance, the American Chemical Society still sees the difficulties in a reliable industrial-scale synthesis as a key barrier for graphene electrodes [87]. In contrast, an AI-based global patent data analysis from 2023 forecasts graphene to disrupt EV battery market in the 2030s—it determines the actual TRL to 5 of 9 [88]. In addition, it is also apparent that companies are eager to integrate graphene into other metal-ion battery technology, such as aluminum-ion batteries, with the objective of accelerating commercialization during the latter half of the decade [89]. This is also reflected in private-sector investments for GRM for high-power electrochemical energy storage applications [90]. In the context of the establishment of a European advanced materials supply chain for batteries, the opening of the first production facility in Europe for single-walled CNTs in 2024 appears also noteworthy [91].

The attention for graphene in batteries in scientific research is still on a high level (with thousands of peer-review papers in that area published each year), while other 2D materials receive growing attention, too. Research trends include advanced engineering techniques such as surface modification, heteroatom doping and chemical functionalization [92–94] as well as graphene preparation in vertical orientation of graphene sheets for electrochemical performance improvements [94, 95] or heterostructures layers constituted of graphene and Gallium nitride for electron and ion conductivity improvements [96] are analyzed as promising for LIB electrode application. Especially heterostructure formation seems to be promising when it comes to graphene composites within advanced anode materials, also reported before for Zirconium disulfide [97] or Hafnium disulfide [98]. Recent reviews on advanced materials [99, 100] conclude that the further development of LIBs still depends on solving major challenges in a number of interrelated parameters like material stability, electrolyte optimization, scalability of production and sustainability, whereby advanced materials and nanoscale engineering with CNTs and graphene have already made a decisive contribution to increasing performance and safety.

6.5. Outlook

The widespread diffusion of electric mobility continues to shape the overall development of the battery field. Markets will likely experience shake-ups while underlying battery technologies will further progress driven by the long-term market expansion. The incumbent LIB technology still continues to advance. Efforts concentrated around (a) cathode material composition (for major cost contributions), (b) cell format optimization (according to application context), and (c) active anode materials (for performance). In particular, the latter aspect opens up an opportunity for advanced additive materials such as graphene to serve as enabler for enhanced silicon incorporation desired by the industry for battery capacity expansion and fast-charging capability. A future roadmap revision may engage in a deliberate comparison of proposed additive systems (graphene as well as competing materials) designed for silicon incorporation.

The emerging graphene supply industry may engage in deliberate co-development initiatives with potential value chain partners for bottom–up preparation of market entry. Competitive cost represents a requisite, but only superior quality and performance will drive the diffusion of graphene in the battery market. First and foremost, potential clients will usually require proof of reliable supply streams though.

Beyond LIB, emerging battery technologies promise better cost, performance and/or sustainability profiles in the future. Among these, lithium sulfur (LiS) batteries stand out for energy density and environmental friendliness—and may represent an alternative entry point for graphene. At present, LiS technology still suffers from low sulfur utilization, capacity fade and low cycle stability [101, 102]. Latest research results indicate that graphene may be the solution as a functional sulfur host or intermediate layers to overcome these problems [28, 103]. A dedicated 3I focus investigation may explore these aspects in further detail.

Data availability statement

The data cannot be made publicly available upon publication because they contain sensitive personal information. The data that support the findings of this study are available upon reasonable request from the authors.

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