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Nanotechnology for environmentally sustainable electromobility

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Abstract
Electric vehicles (EVs) powered by lithium ion batteries (LIBs) or proton exchange membrane hydrogen fuel cells (PEMFCs) offer important potential climate change mitigation effects when combined with clean energy sources. The development of novel nanomaterials may bring about the next wave of technical improvements for LIBs and PEMFCs. If the next generation of EVs is to lead to not only reduced emissions during use but also environmentally sustainable production chains, the research on nanomaterials for LIBs and PEMFCs should be guided by a lifecycle perspective. In this Review, we describe an environmental lifecycle screening framework tailored to assess nanomaterials for electromobility. By applying this framework, we offer an early evaluation of the most promising nanomaterials for LIBs and PEMFCs and their potential contributions to the environmental sustainability of EV lifecycles. Potential environmental trade-offs and gaps in nanomaterials research are identified to provide guidance for future nanomaterial developments for electromobility.
1. Introduction

Anthropogenic greenhouse gas emission rates increased by more than 80% from 1970 to 2010, and emissions from the transport sector increased at a faster rate than any other energy end-use sector. In 2010, transportation was responsible for 23% of total energy-related CO₂-emissions, with total energy consumption reaching 27% of the total end-use energy, of which about half was consumed by light-duty vehicles. There is currently an estimated 1 billion light-duty vehicles worldwide, and as a result of increasing standards of living and economic activity, this number is expected to double by 2035, with obvious repercussions for energy security, climate change and urban air quality.

Vehicles with electric powertrains are seen as attractive alternatives to conventional internal combustion engine vehicles, and many governments have introduced policies promoting market uptake of electric vehicles (EVs). With the increasing market for EVs, major automobile manufacturers now have one or more EVs in their production line. The remarkable drop in the cost of LIBs over the last decade will accelerate the adoption of EVs.

When combined with clean energy sources, EVs can offer a range of advantages over conventional vehicles, such as reduced greenhouse gas emissions and local air pollution and improved energy efficiency. However, a shift in drivetrain technology to LIBs and PEMFCs leads to changes in supply chains, introducing more environmentally intensive materials and production processes in exchange for potentially lower operating emissions. Thus, a systems perspective, such as that provided by life cycle assessment (LCA), is required to understand the environmental implications arising from transport electrification. LCA offers a way to quantify environmental impacts associated with the production, use, and waste handling of goods and services (see Box 1).

Due to their unique electrical and mechanical properties only attainable at the nanoscale, active nanostructured materials developed for LIBs and PEMFCs may significantly improve their performance. Nanomaterials can notably offer advantages over bulk-structured materials through reduced diffusion lengths of ions and electrons, and in some cases, through changes in the phase diagram resulting in changes in reaction mechanism. However, the synthesis of nanomaterials may be more energy demanding than that of their bulk counterparts, which in turn can have significant bearings on the lifecycle environmental impact of EVs, particularly with respect to greenhouse gas emissions. For EVs to offer environmental benefits, the potential technical improvements introduced by nanomaterials must be greater than environmental impact of EV production.

In this review, we investigate how nanomaterials can contribute to more environmentally sustainable electromobility and compare different candidates for development in this direction. For the purpose of this study, the term EVs includes vehicles with a fully electric drivetrain using lithium ion batteries (LIBs) or proton exchange membrane hydrogen fuel cells (PEMFCs). In section 2, we briefly review the LCA literature of EVs to identify potential trade-offs and sources of environmental impacts of the current state of the EV technology. This serves to identify areas in which the development of novel materials may bring about
the greatest improvements from a systems thinking perspective. In Section 3, the challenges identified in Section 2 are grouped into three lifecycle attributes through which nanotechnology may contribute to the development of more environmentally sustainable batteries and fuel cells for electric transport. We then evaluate and compare different nanotechnological developments and challenges with respect to the three lifecycle attributes for batteries (Section 4) and for fuel cells (Section 5). Section 6 distils the overarching evaluations from the previous sections and provides insights into the contribution of nanotechnologies for more environmentally sustainable mobility.

2. Life cycle assessment of electric vehicles

Several academic studies have assessed the environmental impact of EVs\textsuperscript{7,10,14–22}. Studies assessing EVs and relevant components have assumed LIBs for battery electric vehicles\textsuperscript{22–25} and PEMFCs for fuel cell vehicles\textsuperscript{10,15,20,21}. Compared to conventional vehicles, a larger share of EVs’ lifecycle impacts occur in the material processing and vehicle production phase, notably because of their reliance on relatively scarce materials and on production processes with high energy requirements\textsuperscript{10,14,15,19}. Consequently, studies have found up to 40-90\% higher greenhouse gas production-phase emissions for EVs compared to conventional vehicles. Whether or not EVs can compensate for their higher up-front environmental impact depends on the emission intensity of electricity sources and hydrogen for charging LIBs and fuelling PEMFCs, respectively. A lifecycle perspective is therefore required when evaluating their environmental performance\textsuperscript{7,10,14,19}.

Studies assessing impact categories beyond climate change find that EVs can offer substantial positive improvement during its use phase, such as reductions in photochemical smog and fossil resource depletion\textsuperscript{8,19}. However, EVs can also have a negative impact in other categories (e.g., human toxicity, freshwater ecotoxicity, metal depletion), mostly arising from material extraction in the production chain\textsuperscript{14,19,20,26}. Because of the relatively high environmental impacts associated with the production of LIBs and PEMFCs, the lifetime expectancy and the recyclability of these energy devices are key parameters in determining their lifecycle environmental performance. Several studies have pointed to challenges with PEMFC durability due to degradation in the membrane and catalyst layer during long-term operation\textsuperscript{27–29}. Battery EVs, on the other hand, generally suffer from limited driving ranges, and whilst larger batteries allow for longer driving ranges, they also cause more production-phase impacts and add weight to the vehicle, thereby increasing electricity consumption during EV operation\textsuperscript{30}.

As many excellent reviews already cover the contribution of nanomaterials to overcoming technological and commercialization challenges of LIBs and PEMFCs\textsuperscript{31–36}, this review rather screens the environmental effects arising from the use of nanomaterials in these devices. For example, while the battery literature indicates that increasing volumetric energy density is an important factor for LIB adoption in battery EVs due to the limited space available\textsuperscript{37–40},
the LCA literature rather focuses on the need for higher gravimetric energy density to avoid the additional material production and use-phase energy consumption associated with the transport of heavier batteries.\textsuperscript{7,18,19,23,25}

3. Lifecycle approach for early environmental screening

LCAs strive to guide product development by quantifying all environmental impacts associated with each product, but such a comprehensive assessment is typically limited by data quality and quantity. Multiple simplified, or streamlined, LCA methods have been proposed as a first iteration toward complete LCAs\textsuperscript{41–43} in order to provide lifecycle guidance as early as possible in product design, that is, before the design is decided and improvement options restricted. In contrast to full LCAs, there is no standard method to guide the performance of these scoping approaches. In this article, we develop a framework that draws elements from streamlined LCA methods, the qualitative Environmentally Responsible Product Matrix scoping approach\textsuperscript{41–43}, and key principles of green chemistry\textsuperscript{44,45}. These elements are adapted, combined, and updated to address the parameters that both can be influenced by nanotechnological research and determine environmental impacts of EVs. The development of the framework is made all the more pertinent by the fast pace of nanotechnology research, the great diversity of competing nanomaterials, and their differing technological readiness levels, which ranges from laboratory-scale proof of concept to commercialization.

The framework used here appraises nanomaterial candidates with respect to three lifecycle attributes: \textit{environmental intensity of materials, material and weight efficiency}, and \textit{energy efficiency}, which are described in detail below and illustrated in Figure 1. Together, these lifecycle attributes cover all lifecycle phases of the material: production, use and end-of-life. To guide action, we distinguish between intrinsic parameters that are attributed to the material itself, and value chain parameters that are characteristic not of the material but of the activities involved in its production. The evaluation of materials is adapted to the special nature of electromobility. Section 3 in the Supplementary information describes the criteria and basis of comparison and provides further details in Tables S1–S5 and Figures S4–S9.

\textbf{Environmental intensity of materials}

The environmental intensity of a material describes the extent to which producing and using a given mass of a given material causes damages to the lifecycle areas of protection: human health, ecosystems, and resource availability (Box 1). For example, energy intensive extraction or production processes can result in high greenhouse gas emissions, which in turn can lead to damages to human health and ecosystems. This lifecycle attribute is highly relevant since, on the one hand, LCA studies on EVs find that materials used in LIBs and PEMFCs have environment commercially intensive extraction and refining processes\textsuperscript{10,14,15,19}, and since, on the other hand, nanotechnological developments are likely to alter the materials used in LIB and PEMFC productions. Some materials can themselves cause damages through...
exposure risks and hazards. The use of non-renewable materials can increase resource scarcity, while material extraction and processing activities throughout the production chain result in embodied damage to human health and damage to ecosystems. Reducing the particle size from bulk material to a nanoscale can change both the material properties (e.g., increased reactivity) and lead to differing environmental intensity (e.g., damage to human health).

Material and weight efficiency
The material efficiency characteristic is a metric of the functionality that a material can achieve per unit of mass. As the environmental aspects of materials as described in the previous section scale directly with the amount of material used, we should strive to attain the same functionality with less material. Given the relatively high environmental impacts associated with material processing in the production of LIBs\textsuperscript{22,24,46} or PEMFCs\textsuperscript{10,15,21} for EVs, optimizing the utilization of the materials in these devices is important. Increasing gravimetric energy density in LIBs or power density increases the material efficiency as less material can be used for the same energetic output. Improvements in material lifetime and stability allow for devices that last longer and in turn can reduce the need for replacement, thereby avoiding the use of additional materials. Energy density, power density, and lifetime and stability of nanomaterials were compared to the performance of commercial ‘baseline’ material. Reducing material losses during synthesis and increasing the recyclability both improve material efficiency by minimizing waste. The use of nanomaterials in LIBs and PEMFCs may affect the material efficiency (e.g., change in energy or power density) due to large surface areas, but it may also result in unwanted side-reactions (e.g., influence lifetime and stability). Material efficiency considerations such as energy and power density allow for lighter batteries and PEMFCs; these lightweighting effects also provide side benefits in the form of gains in energy efficiency.

Energy efficiency
Energy efficiency is a measure of how much functionality a given energy input can provide; here we consider energy losses during operation and energy use in the synthesis of nanomaterials. Depending on the energy sources used for producing electricity or hydrogen, the energy losses in LIBs and PEMFCs during operation can contribute to a substantial share of the device’s lifecycle greenhouse gas emissions and other environmental impacts\textsuperscript{9,19,24,25}. Here, we consider the device efficiency to measure how well nanomaterials enable the device to transform and deliver energy. LCA studies find that energy consumption in the value chains of LIBs can also contribute significantly to their greenhouse gas emissions and production impact\textsuperscript{24,25,46}. Energy of nanosynthesis measures how energy efficient the manufacturing processes of nano-enabled LIB and PEMFC materials are. While using nanomaterials instead of bulk materials may improve the device efficiency due to increased reactivity, the differing methods to synthesize these nanomaterials require varying amounts of energy. As energy is often produced from carbon intensive sources, energy use often translates to greenhouse gas emissions.
In the following sections, qualitative and semi-quantitative comparisons will be performed in terms of the three lifecycle attributes for various nanomaterials. Figures 2-5 use colour coding to illustrate the perceived relative strengths of different nanostructure materials with respect to the above lifecycle attributes. Green denotes relative strength, red relative weakness, yellow intermediate characteristics, and white a lack of data. Nanostructures are given by circles, whereas the paler background indicates the characteristics of the material in bulk form. Absence of a circle indicates a lack of data relevant to nanostructures. The grey background denotes the ‘baseline’ material. Although many of these lifecycle attributes pertain to the device as a whole (e.g., energy density, power density, and lifetime), we will consider the materials in isolation for greater ease of analysis. Thus, a cathode with high specific capacity and operating voltage will be described as a “high energy density cathode” because its combination with an appropriate anode allows for a high energy density LIB.

4. Nanotechnologies in battery developments

Battery cells are composed of several key components: anode, cathode, separator, electrolyte, and current collectors. However, their energy density and environmental footprint are mainly determined by the properties of the electrode materials. We therefore focus on the environmental performance of different nanostructured anode and cathode materials.

Anode materials
The use of pure lithium anodes is precluded in rechargeable LIBs with liquid electrolytes because of the formation of lithium dendrites on charging, which short the cell, leading to thermal runaway and fires. Due to this increased reactivity and the associated safety issues, pure lithium anodes in nanoform are, so far, unsuitable for LIBs. Most current LIBs rely on the intercalation of lithium ions in anodes predominantly composed of graphite. More recently, the use of nanosized lithium titanium oxide spinel (Li₄Ti₅O₁₂, LTO) has also been adopted. In addition to these commercial anode materials, multiple alloys and conversion anode materials are currently under research. Figure 2 presents the material lifecycle attributes of reviewed anode nanomaterials, as well as graphite. Graphite is an abundant material, and its extraction or synthesis has relatively low environmental impact. Today, it also requires little energy during its production and allows for batteries with good cyclability and high energy efficiency. The main weaknesses of this chemistry from a sustainability standpoint relates to its low material efficiency; its limited energy density leads to heavier, larger batteries.

Alternative carbon nanostructures with higher theoretical energy densities are under investigation, but neither carbon nanotubes nor graphene have been found to be technically feasible because they have too many side-reactions. Carbon nanotubes and graphene also exhibit more environmentally intensive profiles and, like other carbon
nanostructures, their handling requires more precaution than graphite. The current carbon nanotube synthesis routes are energy intensive. Even when potential economies of scale are taken into account, energy requirements for the synthesis of carbon nanotubes through chemical vapour deposition, arc discharge, or laser-assisted methods all remain significant, which in turn result in high greenhouse gas emissions. Further, carbon nanotubes anodes have lower charge-discharge energy efficiencies. Increasing evidence points to toxicity effects of carbon nanotubes similar to those of asbestos fibres, which may affect production and end-of-life processing and recycling of the batteries.

LTO is obtained from relatively abundant resources and has moderate production impacts. It intercalates lithium in a safer manner than carbon because it is 1.5 volts away from lithium metal deposition, but must be nanostructured in order to reach acceptable power densities because of its low conductivity. Contrary to carbon nanotubes, LTO can be synthesized with moderate amounts of energy and low reagent losses, especially if a hydrothermal synthesis route is selected. The resulting nanostructured anodic material offers high cycling energy efficiency, extreme safety, high power density, and extended lifetimes. Although LTO is already used in small commercially available EVs, the 1.5V operating potential of LTO leads to inherently low energy densities, which reduces its material and weight efficiency and thus its environmental desirability for EVs. LTO nanoparticles also pose a high exposure risk. The positive properties of LTO, however, potentially make it an environmentally sustainable candidate for static and high power applications.

Even more abundant than carbon, silicon presents the highest theoretical capacity to store lithium of all studied anode materials, potentially allowing for high energy density anodes. Refining silicon to metallurgical grade for use in the chemical industry causes moderate damages to human health and ecosystems. Regarding electrochemical performances, bulk silicon anodes suffer from poor power density and extreme volume changes that lead to rapid structural degradation of the electrode, resulting in poor lifetime. The material must therefore be nanostructured to ensure that voids can buffer such swelling. Silicon nanoparticles in carbon-based nanocomposites and silicon nanowires have shown to improve electrochemical performance and lifetime with cycle life of 1000-2000 cycles. Nanostructured silicon anodes thus open the possibility for high material efficiency in the LIB lifecycle, particularly with respect to lifetime and energy and power density. However, handling silicon nanoparticles in carbon nanostructures and silicon nanowires requires some precaution. The most popular technique used to grow silicon nanowires is chemical vapour deposition, which has moderate to high energy requirements. As a result, the synthesis of nanostructured silicon may result in high greenhouse gas emissions. Furthermore, during the use-phase, silicon anodes also suffer from higher voltage hysteresis and thereby lower cycling energy efficiencies than graphite or LTO.
Tin and germanium can also reversibly alloy lithium. Nanostructured tin-based anodes cycle with a higher Coulombic efficiency than silicon\cite{47}, and germanium-based anodes allow for exceptional power densities\cite{34}. However, given the greater scarcity\cite{47,66} of these metals and the environmental impacts of their extraction and refining\cite{50,51}, their lifecycle environmental sustainability performance remains unremarkable\cite{57,80,81}. Tin may nonetheless prove attractive because of its superior performance when combined with other elements, such as abundant and low-impact iron (e.g., $\text{Sn}_2\text{Fe}$ nanoparticles)\cite{82–84}. Many nanostructured transition metal oxides can enter in a conversion reaction with lithium, which in principle offers more options as potential anode materials. Among these, iron oxides such as haematite ($\alpha$-$\text{Fe}_2\text{O}_3$) and magnetite ($\text{Fe}_3\text{O}_4$)\cite{34} are by far the most abundant\cite{47,66} and the least environmentally intensive\cite{50,51,85,86}, in contrast to more scarce elements\cite{47,66} such as chromium, molybdenum, ruthenium, and cobalt\cite{87–89}. Green synthesis routes for iron oxide nanoparticles should lead to relatively lean use of reagents and energy\cite{54,90}. Though high specific capacities have been demonstrated\cite{54,90}, their relatively high voltages during de-lithiation\cite{34} substantially reduces the overall cell voltage and consequently, energy and power density. High voltage hysteresis\cite{68,91} makes all these issues worse and also leads to low cycling energy efficiencies, typically less than 60%. Such low energy efficiencies constitute a major handicap for an otherwise environmentally attractive material.

**Cathode materials**

The energy density of LIBs is largely determined by the cathode as its practically achievable energy is much inferior to that of the anode\cite{92,93}. There are two broad categories of cathode materials: intercalation and conversion. Intercalation materials are the most widely investigated and are already used as bulk materials in commercial LIBs\cite{47}. Of the conversion-type cathode materials, none have reached commercialization\cite{47,94}. Figure 3 presents the material lifecycle attributes of reviewed cathode nanomaterials. LiNi$_{0.8}$Co$_{0.15}$Al$_{0.05}$O$_2$ (NCA) is considered to be the ‘baseline’ cathode material. By far the most commonly used cathodes today are the layered oxides, such as LiCoO$_2$ (LCO). Due to the use of the relatively scarce cobalt\cite{47,66}, commercially available LCO causes moderate direct exposure risks\cite{86,95} and embodied damages to human health and ecosystems\cite{50,51}. In addition, cobalt’s high cost has led a drive to replace most of it in many applications\cite{96}, resulting in the adoption of materials with lower cobalt content such as LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ (NMC) and NCA. The popular NMC and NCA pose exposure risks and hazards because they, as with many nickel-containing compounds, are suspected of being human carcinogens\cite{57,97–99}. Their high energy- and power densities have nevertheless made them attractive as bulk materials, and these materials are already used in EVs\cite{47}. As nanostructures, however, the decomposition of the electrolyte and formation of surface films result in insufficient lifetime for EV applications. Even though these layered oxides are not used in nanoform, alternative materials must have equal or superior energy density.
while demonstrating better lifetime and stability than bulk NMC and NCA in order to displace them from the EV market.

A promising layered oxide is the lithium/manganese-rich material (LMR)\textsuperscript{100}, often written as $\text{Li}_2\text{MnO}_3\cdot n\text{LiMO}_2$ (where $M = \text{Mn, Co, Ni, etc.}$). LMR contains more than one lithium atom per transition metal and has more manganese than other metals. Here, we focus on $0.5\text{Li}_2\text{MnO}_3\cdot 0.5\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$. Due to its higher content of manganese relative to NMC, LMR is slightly less environmentally intensive than NMC\textsuperscript{50,51,98,101}. Furthermore, LMR also has a high voltage and specific capacity that allows for a significant increase in energy density over current commercially available cathode materials\textsuperscript{102}. Despite these advantages, poor rate capability\textsuperscript{103} result in low power density, whereas thermal safety issues\textsuperscript{37} and voltage fade\textsuperscript{104} result in poor lifetime and stability, all of which complicate its commercial introduction for EVs.

Lithium iron phosphate (LFP) is found in nature as the mineral triphylite\textsuperscript{105} and has low exposure risks or hazards\textsuperscript{86}. Furthermore, environmental impacts associated with its production value chain are lower than most other cathode materials\textsuperscript{16,50,51}. As a bulk material, LFP has moderate electric potential\textsuperscript{47}, outstanding thermal stability\textsuperscript{52}, and excellent cycling performance\textsuperscript{106}, but its two-phase reaction mechanism, with low ion diffusion rate and very low electronic conductivity\textsuperscript{107}, makes it difficult to reach capacities close to the theoretical limit\textsuperscript{52}. However, research found that in nanoparticle form, the material could produce stable cycling much closer to its theoretical capacity because the phase diagram is changed and the reaction proceeds via a metastable single-phase mechanism\textsuperscript{37}. This development increased the material’s energy\textsuperscript{52} and power\textsuperscript{33} densities, but its energy density remained inferior to that of other commercially available cathode materials such as NMC\textsuperscript{47,48}. The lower energy density\textsuperscript{47} and the claimed lower charge-discharge energy efficiency of LFP\textsuperscript{106} can result in higher electricity use per kilometre driven compared to other cathode materials, which in turn would lead to higher indirect greenhouse gas emissions in the use phase. LFP can be produced through several nanosynthesis methods\textsuperscript{108}, which particularly influences the energy use, and consequently greenhouse gas emissions, associated with its production. The superior electrochemical and safety properties of nano-LFP has spurred interest in finding other phosphates that might have much higher energy densities. One approach is to use materials that can incorporate up to two lithium ions. One such material is VOPO$_4$, which must be nanosized and carbon coated to be operative\textsuperscript{109}, but has the advantage of being made of relatively abundant materials\textsuperscript{47,66}. This material forms Li$_2$VOPO$_4$ (LVP) on discharge and has a capacity of 305 Ah/kg compared to the 170 Ah/kg of LFP. However, the lifetime and stability are inadequate for EV use and much work is still needed to make LVP commercially viable.

Spinel LiMn$_2$O$_4$ (LMO) is made of abundant manganese\textsuperscript{47,66}, is relatively safe to handle\textsuperscript{86,110}, and has relatively low damages associated with its production\textsuperscript{50,51}. Nanosized spinel LMO has been synthesized in various morphologies. Studies have found increased power densities\textsuperscript{47}, and although increased energy densities have also been obtained\textsuperscript{107}, these are not as high as
those of bulk NMC and NCA\textsuperscript{47,106}. In the case of LMO, nanoparticles tend to increase the undesirable dissolution of manganese to the electrolyte\textsuperscript{32,107}, leading to lifetime issues. Porous nanorods, however, have been found to have remarkable lifetime\textsuperscript{111}. As one of very few viable options to the intercalation materials, the conversion material sulphur has received intense interest in the past decade due to exceptionally high theoretical energy density\textsuperscript{112–115}. Supply of sulphur is unlikely to become an issue as it is the thirteenth most abundant element in the earth’s crust\textsuperscript{47}. In batteries, the insulating nature of sulphur results in poor power density and creates large internal resistance and polarization of the battery\textsuperscript{116}, resulting in poor device efficiency. Furthermore, volume expansion (\textasciitilde 80\%) and dissolution of intermediate reaction products (polysulphides) in the electrolyte result in poor lifetime\textsuperscript{47,113}. The most promising approach to mitigate poor conductivity and lifetime is the encapsulation of sulphur within conductive additives to form sulphur-carbon and sulphur-polymer nanocomposites\textsuperscript{47,117}. Sulphur-carbon nanocomposites pose higher exposure risks and hazards\textsuperscript{56,118} than sulphur nanocomposites with polymers such as polyacrylonitrile, polyvinylpyrrolidone, polydimethylsiloxane\textsuperscript{118,119}, and polyaniline\textsuperscript{118,120}. Even if the issue of lifetime is overcome, the sulphur cathode must be paired with a lithium metal or a lightweight lithiated anode for high energy density\textsuperscript{47,114,121,122}. In contrast, lithium sulphide (Li\textsubscript{2}S), can be paired with lithium-free anodes, which avoids safety concerns and short lifetime\textsuperscript{122}. Although the Li\textsubscript{2}S cathode has a high theoretical capacity, it is both electronically and ionically insulating\textsuperscript{47}, which have led to various efforts using conductive additives, such as metals and carbon\textsuperscript{114}. Earlier studies tended to focus on Li\textsubscript{2}S-metal composites, but the inherent disadvantages of Li\textsubscript{2}S-metal composites have created extensive interest in the development and use of Li\textsubscript{2}S-carbon composites in the past five years\textsuperscript{116}. Due to a high content of lithium and carbon nanostructures, care should be taken when handling nanostructured Li\textsubscript{2}S-carbon composites\textsuperscript{56,86}. Studies have reported different nanostructures, synthesis methods, and carbon content in Li\textsubscript{2}S-carbon nanocomposites and this can lead to significant differences in material losses and energy use, which in turn influence greenhouse gas emissions and damages to human health and ecosystems. Further improvement on lifetime is required for Li\textsubscript{2}S cathode materials to replace the layered oxides from the EV market.

**Recycling of LiBs**

There are several competing industrial LIB recycling processes\textsuperscript{123}. LIB recycling is typically a combination of two or more of the following processes: mechanical separation, pyrometallurgical, and hydrometallurgical treatment. The various industrial recycling pathways offer different yields depending on the recycling route and electrode materials. As the metal value in batteries is mainly driven by prices of cobalt and nickel metals, current recycling processes still focus on the recovery of these metals\textsuperscript{97,124,125}. Other transition metals, such as copper and iron, are also typically recovered in the current industrial LIB recycling processes. In only a few recycling routes are aluminium, lithium, and manganese recovered\textsuperscript{97,123,125}. According to relevant literature\textsuperscript{97,123,126} and personal communication with
two European recycling companies, phosphate and graphite are normally not recycled in current industrial processes. Nanostructured LFP is currently recycled successfully, which may suggest that nanostructuring electrode materials do not affect recycling yields compared to bulk materials. During recycling, however, nanomaterials may become airborne, which can pose exposure risk and hazard to workers.

5. Nanotechnologies in fuel cell developments

While there are multiple fuel cell types, we focus here on PEMFCs, which demonstrate the most potential within the transport sector. High cost, durability and lifetime challenges are all barriers to the mainstream adoption of fuel cell EVs; in contrast to battery EVs, commercial sale of fuel cell EVs has only very recently become reality. In contrast to LIBs, the ‘baseline’ materials are already in nanoform; we review here rather alternative nanostructures and nanomaterials that have the potential to replace current state of the art materials. These advances in nanotechnology have shown promising opportunities to improve the technical and environmental performance of PEMFCs in EVs and thus encourage their widespread commercial adoption.

Figures 4 and 5 summarize the lifecycle attributes of some of the most promising nanostructured materials for cathode catalyst and catalyst support, respectively. Although the electrocatalyst often refers to the catalyst and support together (Pt/C), they are considered as two components independent of each other in this study. Electrolyte membranes, being a bulk material, are discussed in section 4 of the Supplementary Information while nanotechnological improvements to these bulk materials are discussed in the text.

Cathode catalysts

The oxygen reduction reaction occurring at the cathode is enabled by the cathode catalyst; a well-performing catalyst is therefore a determinant of the device’s overall power output. At present, both PEMFC anodes and cathodes rely on platinum catalysts supported on high surface area carbon (Pt/C), which are costly, scarce and have extremely high environmental implications from platinum extraction. In terms of efficient use of this high-impact, non-renewable material, the cathode is the key technological bottleneck as the oxygen reduction reaction occurs five to six orders of magnitude slower than the hydrogen oxidation reaction occurring at the anode, thus greatly limiting the cell power density. Furthermore, the pure platinum catalysts suffer from poisoning from impurities in the hydrogen fuel as well as dissolution and agglomeration, which can drastically shorten the fuel cell lifetime. A shorter lifetime demands more frequent replacement of PEMFC stacks in EVs, and may ultimately require more platinum extraction per kilometre driven. Current research therefore focuses on reducing or eliminating platinum use in the catalyst. Several solutions are being explored, including the use of ultra-low platinum loading, platinum alloys and platinum-free catalysts to reduce material costs while
maintaining or improving catalytic activity over current Pt/C catalysts. In comparison to the commercial Pt/C catalyst, most of these platinum-containing alternatives yield enhanced durability (Figure 4) and demonstrate similar or superior oxygen reduction reaction catalytic ability.

Alternative platinum nanomorphologies and nanostructured platinum alloys can maintain or even increase the catalytic activity relative to conventional Pt/C catalysts. Increasing the specific catalytic activity allows for a reduction in the amount of platinum used, thus improving material efficiency over the conventional catalyst. In addition to the various nanomorphologies, research using different assembly methods, such as electrospraying, improve catalytic activity by influencing the hierarchical structure of the electrode\textsuperscript{137,138}. Similarly, platinum alloys with nickel\textsuperscript{139–141}, cobalt\textsuperscript{140,142} and copper\textsuperscript{143,144} have also demonstrated good performance while decreasing platinum use.

While platinum reduction is a desirable goal for PEMFC development, the complete elimination of platinum use in PEMFCs would be an even greater improvement of the material environmental impacts\textsuperscript{66,145}. Non-precious metal catalysts using more abundant metals such as iron have been tested, but present severely depressed technical performance and stability in acidic operating conditions\textsuperscript{146}. Other metal catalysts based on niobium, tantalum, and zirconium have improved lifetime over Pt/C, but do not meet power density expectations, and are more scarce\textsuperscript{47,66} and environmentally intensive to produce than iron\textsuperscript{50,51}, although they still represent an improvement over platinum. Metal-free catalysts using functionalized carbon nanostructures, particularly N-doped carbon nanotubes and graphene materials, are promising candidates for platinum-free catalysts that capitalize on abundant precursor materials, though they require further research to improve the energy efficiency of their synthesis and to provide adequate catalytic ability in acidic environments\textsuperscript{147–149}. A clear trend, however, is that platinum-free catalysts continue to struggle in catalytic activity and lifetime in comparison to low-platinum and platinum-alloy catalysts\textsuperscript{147}.

In addition to the morphological and material nature of the catalyst, the hierarchical organization of the nanostructured materials in the device also affects catalyst performance. While such organization may increase material efficiency by increasing catalytic activity, it may also present consequential side issues such as water flooding, which in turn cancels out or exceeds the gains in performance, or causes unstable cell performance\textsuperscript{150}.

If the goal is to reduce the amount of platinum used in fuel cell EVs to the amount used in the catalytic converters of conventional internal combustion engine vehicles, the device lifetime must be accounted for. Since fuel cell EVs currently have a shorter lifetime than conventional vehicles, the amount of platinum required to drive an equal distance increases, i.e., several fuel cell stacks will be required. Furthermore, the growing light-duty vehicle market represents an unsustainable demand for further platinum extraction into the future. Rather, focus should be placed on robust, low- or non-platinum catalysts with long lifetime.
Cathode catalyst supports
Effective support materials enhance catalytic catalyst utilization and thus increase material efficiency by allowing for smaller quantities of catalyst while maintaining similar levels of catalytic activity. A catalyst support would ideally maximize the catalyst surface area available for reactions and maintain high electric conductivity for high energy efficiency. Supports made of carbon black currently used in commercial PEMFC catalysts are vulnerable to corrosion, which causes catalyst sintering and decreases the amount of conductive material in the electrode, thereby decreasing power density and PEMFC lifetime. Carbon black-based support materials also suffer from deep micropores that physically block reagent access to the catalyst and thus decrease catalyst efficiency.

Nanostructured materials can provide the characteristics needed for an effective catalyst support, including a high surface area with a mesoporous structure that does not inhibit catalytic activity. Catalyst support materials must also be sufficiently electrically conductive in order to reduce internal resistance, thereby enhancing charge transport within the cell and be stable at higher temperatures and in the acidic environment of a PEMFC. The two most promising catalyst support materials that are environmentally beneficial and demonstrate improved technical performance are carbon nanostructures and titanium dioxide, two materials with low environmental intensity in their bulk form (Figure 5). The synthesis methods for the nanomorphologies, however, may potentially have high energy demand, and thereby be detrimental to the overall climate change performance of the manufacturing process. The graphitized carbon-based nanomaterials have enhanced durability under fuel cell operating conditions, which improves the climate change performance of the PEMFC over the lifetime as a counterpoint for the increased synthesis energy. Doping the carbon with heteroatoms such as nitrogen, phosphorus or sulphur functionalizes the otherwise inert carbon to allow catalyst deposition. In some cases, functionalization, such as with nitrogen-doped carbon nanotubes, also allows the otherwise catalytically inert carbon supports to become catalytically active, thereby increasing power density of the PEMFC. Some carbon-polymer nanocomposites have shown improved material efficiency via power density, but, in some cases, this is in exchange for reduced lifetime.

Carbon-free, transition metal oxide-based supports such as titanium dioxide in mesoporous or nanofiber morphologies, while relatively robust, have not yet achieved the same performance level as the baseline carbon black catalyst support. Composite titanium dioxide catalyst supports may also be more sensitive to scarcity and material production impacts than carbon-based supports, as are supports of niobium- and ruthenium oxide-doped titanium dioxide.

Electrolyte membrane
The PEMFC membrane, with its high cost, poor durability and intolerance to fuel impurities, represents another obstacle to the widespread commercialization of transport PEMFCs. The current commercial baseline, Nafion®, is a perfluorinated membrane that
performs poorly in temperatures beyond 80 °C and in low-humidity environments, and is not stable with impure feed gases\textsuperscript{157,158}. An ideal membrane for transport PEMFCs must therefore have satisfactory performance and stability at these conditions. Research has been directed towards more robust membranes, which would allow for thinner membranes that represent an improvement in material efficiency (less membrane material used) and device efficiency (e.g., superior ion exchange/proton conductivity performances). While membrane polymers conduct protons at the nanoscale, the membrane material itself does not constitute a nanomaterial. A brief review of the main membrane polymer groups may be found in Section 4 in the Supplementary information. Nanotechnology offers several options for improving these bulk membranes. Such options include the use of nanofillers to enhance the membrane, or the use of nanosynthesis methods to provide a superior hierarchical structure to the membrane.

One attractive strategy of generating an optimum balance between ion conduction and physicochemical stability in electrolyte membranes is to create a “microphase-separated” morphology in polymers made of highly ordered ion-nanochannels and a hydrophobic phase. An example is the fabrication of ion-conductive polymer nanofibers, demonstrating distinctive electrochemical, physicochemical, and thermal properties owing to their high specific surface area and polymer orientation along the nanofiber direction\textsuperscript{159,160}. The use of a reinforcing, mechanically strong nanofiber morphology can minimize in-plane swelling changes during wet(on)/off(-dry) fuel cell operation and thus extend the device lifetime\textsuperscript{161}. Some success has been achieved with a dual electrospun composite of poly(phenyl sulfone) and Nafion\textsuperscript{162}, where PPSU provides mechanical stability to the PFSA membrane, thus improving lifetime while maintaining device efficiency (cell power output). Similarly, improved proton conductivity, leading to increased power density was achieved with electrospun acid-doped polybenzimidazole in a sulfonated polymer matrix in comparison to a similar composite membrane without nanofiber morphology\textsuperscript{160}.

In one type of composite membrane, a polymer membrane matrix may have embedded nanostructures of inorganic materials in order to improve membrane characteristics. Such materials may be metal oxides or synthetic clays to improve mechanical stability\textsuperscript{163}, water uptake, or nanocarbons or nanofibers to provide ionic channels and thus improve device efficiency of the PEMFC. Heteropolyacids such as phosphotungstic acid are used as fillers to improve proton conductivity (device efficiency), but decrease mechanical stability and therefore have a shorter lifetime. Phosphotungstic acid also has significant exposure risks\textsuperscript{164}. However, while hygroscopic particles are intended to increase the device efficiency by improving proton conductivity via increased water retention, these particles decrease device efficiency by diluting the concentration of the proton-conducting ionomer when made of material less conductive that the ionomer membrane\textsuperscript{165–168}. Nanofillers may also increase the mechanical strength of the polymer, as in the case of zwitterionic structured SiO\textsubscript{2} in polybenzimidazole\textsuperscript{163,169}. In addition, the heterogeneous hybrid membranes also experience phase separation due to differing water uptake and thermal expansion coefficients of the
nanofillers and the polymer matrix, causing stresses and strains in the membrane and thereby shortening the lifetime and decreasing material efficiency.\textsuperscript{170} Hierarchical ordering in these nanocomposites are also a promising strategy to improve membrane performance; in particular, the alignment of one-dimensional (nanotubes, nanofibers or nanorods) and two-dimensional nanomaterials (nanoflakes, nanosheets, or nanoplates) in the membrane have a two-fold benefit. In the direction parallel to the membrane, proton conductivity is improved, while the across the membrane, mechanical properties, chemical stability and fuel permeability characteristics are improved. Graphene oxide\textsuperscript{173} and electrospun\textsuperscript{160,162} nanofibers are particularly emphasized due to the creation of long-range ordered ionic nanochannels for proton conduction and excellent physicochemical stability.

**Recycling of PEMFCs**

In terms of both cost and environmental intensity, platinum catalyst and fluorinated membranes are of greatest interest for recycling and recovery processes. The most common platinum recovery approaches include selective chlorination or gas phase volatilization, hydrometallurgical and pyrometallurgical processes\textsuperscript{171}. Selective chlorination or gas phase volatilization, however, require carbon monoxide and chlorine gases or aggressive solvents such as aqua regia or cyanide. Many of these compounds pose considerable risks to workers.\textsuperscript{172–174} Many hydrometallurgical approaches also require high operating temperatures and pressures, making them energy intensive processes. Pyrometallurgical processes for PEMFCs containing fluorinated membranes such as Nafion would result in the emission of highly toxic hydrogen fluoride.\textsuperscript{175,176} The Pt/C catalyst can also be recovered using a chemical recovery process after carbon-based supports are incinerated\textsuperscript{175,176}. Generally, alloying and non-combustible elements consisting of 10% or less of the total recoverable materials will not detrimentally affect recoverability or reusability of precious metal catalysts\textsuperscript{128}.

Mechanical separation of membranes from the catalyst layers is difficult, as these components are generally hot-pressed together\textsuperscript{175}. Re-use of the membrane is also unlikely as performance drops in fuel cells are usually caused by membrane degradation or failure due to dehydration and pin-holing, which makes recycling a more likely end-of-life fate for membranes.\textsuperscript{175} Nafion membranes are generally recovered using chemical extraction\textsuperscript{175–177}, after which a new membrane may be re-cast, although possibly with some loss of quality.\textsuperscript{177} As with the catalyst, it is unknown whether the adoption of novel multi-element catalysts and alternative catalyst support materials in PEMFCs will affect the yield or quality of recovered precious metals given the current PEMFC recycling techniques.

**6. The road ahead**

Nanomaterials are opening a broad range of opportunities to improve the technical and lifecycle environmental performance of EVs. Identifying the alternative material candidates...
with the most promising opportunities for enhancing overall environmental performance of LIBs and PEMFCs in EVs at an early stage is therefore important. To this end, we performed an early stage lifecycle environmental screening and mapped their potential strengths and weaknesses with respect to key lifecycle attributes (Figures 2-5). We found that no single nanomaterial seems poised to outcompete its rivals in terms of all reviewed sustainability criteria for any of the reviewed LIB and PEMFC materials. Rather, the current research frontier presents multiple promising candidates for continued development, each subject to non-trivial environmental trade-offs that should be addressed.

To maximize climate change mitigation benefits offered by EVs, we must improve both the electrochemical and environmental performance of LIBs and PEMFCs. Nanomaterials show great promise in providing the necessary technical breakthrough in these devices, but their ability to be a part of the mitigation solution for transport-related greenhouse gas emissions depends on several life cycle attributes spanning from extraction, refinement, synthesis, operational performance, durability and recyclability. As such, the next generation of LIBs and PEMFCs should ideally be based on abundant resources that can be extracted and refined with low energy consumption and environmental impacts. It should be resource and material efficient, achieved through improvements in synthesis yields, lightweighting, durability and ultimately, recyclability. Finally, it should be energy efficient, both in the production and use phase. In practice though, we are likely have to make some trade-offs.

Our analysis of the current situation clearly outlines the challenge: the materials with the best potential environmental profiles during the material extraction and production phase (less environmentally intensive materials, lower nanosynthesis energy use, and facile synthesis) often present environmental disadvantages during their use-phase (lower energy efficiency, heavier battery, or shorter lifetimes), and vice versa. Meeting this challenge will require concerted efforts and a new focus within the nanotechnology community. Throughout this review, we found that publications on novel nanomaterials rarely explicitly communicate synthesis yields, solvent use, and energy consumption during production. These are all are key parameters that significantly influence the environmental performance and that can largely be improved through the choice of alternative synthesis protocols and foreseeable economies of scale. Improved, systematic and consistent reporting of these attributes would remove a very avoidable source of uncertainty. Improved flow of information would be of mutual benefit to both the LCA and nanotechnology communities; through joint efforts, both communities would be able to direct research efforts towards the materials and synthesis protocols with the best environmental sustainability potential. An extension of the above aspect is the current lack of data regarding potential toxic effects, which unfortunately remain a challenge for nearly all of the investigated nanomaterials. Similarly, we also found little literature on how the physicochemical properties of novel nanomaterials affect existing recycling and disposal processes. Addressing these issues would over time allow us to efficiently manoeuver towards the most environmentally superior options. As more detailed and consistent
information becomes available, one can move from screening studies to detailed LCAs in order to refine our understanding and ultimately make the right design tradeoffs that optimize LIB and PEMFC nanomaterials for EV usage towards mitigating climate change. This will require a cross-disciplinary collaboration between material scientists and LCA practitioners to reap – and maximize – the benefits offered by simultaneously incorporating nanotechnology, nanotoxicology, eco-design and green chemistry considerations. If we succeed, nanotechnology can be a key contributor to climate change mitigation in the transport sector.

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Additional information
Supplementary information accompanies this paper at www.nature.com/naturenanotechnology. Reprints and permission information is available online at http://npg.nature.com/reprintsandpermissions/. Correspondence and requests for materials should be addressed to LE.

Competing financial interests
The authors declare no competing financial interests.
Figures

**Figure 1 Early lifecycle environmental screening of LIB and PEMFC for electric vehicles.** Solid lines denote intrinsic aspects of the material itself. Dotted lines and italic font denote properties that are attributes of the value chain aspects, or embodied activities related to the material’s production. Red lines denote production aspects, dark grey lines use phase aspects, and blue lines end-of-life aspects. Abbreviation: EOL – end-of-life.
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Figure 2 Anode materials for lithium ion batteries. Nanoarchitectured materials are given by a circle. Background colours reflect characteristics of bulk materials. Green denotes relative strength, red relative weakness, yellow intermediate characteristics, and white no data. Absence of circle indicates no data for nanomaterial. The grey background denotes the ‘baseline’ material. Abbreviations: LTO – lithium titanium oxide, Si – silicon, Sn – tin, SnO\textsubscript{2} – tin oxide, Ge – germanium, Fe – iron, Co – cobalt, Cr – chromium, Cu – copper, Mo – molybdenum, Ru – ruthenium, Ni – nickel, and Mn – manganese. Data from graphite from references 33,47,50–52,54,57,66,178,179; data from carbon nanotubes from references 12,33,52,56,60,61,180; data from LTO from references 13,33,34,47,50–52,66,67,71; data from Si from references 23,33,47,50,51,57,60,66,67,73,75–79,91,181–184; data from Sn/SnO\textsubscript{2} from references 34,47,50,51,57,66,80,81,185,186; data from Co and Cr oxides from references 50,51,66,86–88,97,196,197; data from Cu oxides from references 47,50,51,66,86,89,199; data from Mo or Ru oxides from references 50,51,66,86,200,201; data from Ni oxides from references 50,51,66,86,200,201; data from Mn oxides from references 47,50,51,66,86,202,203. See the Supplementary citation data for reference details.
### Figure 3 Cathode materials for lithium ion batteries.

Nanoarchitectured materials are given by a circle. Background colours reflect characteristics of bulk materials. Green denotes relative strength, red relative weakness, yellow intermediate characteristics, and white no data. Absence of circle indicates no data for nanomaterial. The grey background denotes the ‘baseline’ material. Abbreviations: NCA – lithium nickel cobalt aluminium oxide, NMC – lithium nickel manganese aluminium oxide, LCO – lithium cobalt oxide, LMR – lithium/manganese rich transition metal oxide, LFP – lithium iron phosphate, LVP – lithium vanadium phosphate, and LMO – lithium manganese oxide. Data from NCA from references 47, 50, 51, 56, 60, 66, 86, 92, 97, 99, 204, 205; data from NCM from references 33, 46, 50, 51, 60, 66, 98, 99, 204, 206–209; data from LCO from references 32, 33, 39, 47, 50, 51, 60, 64, 86, 96, 106, 210–204, 208, 59; data from LMR from references 47, 50, 51, 60, 66, 97, 98, 101, 204, 211; data from LFP from references 33, 39, 50, 52, 60, 66, 74, 86, 97, 96, 105–108, 204, 212; data from LVP from references 50, 51, 60, 66, 86, 97, 109, 211, 214; data from LMO from references 32, 33, 47, 50, 51, 60, 64, 86, 106, 107, 110, 111, 204, 215; data from S from references 47, 50, 51, 56, 60, 66, 93, 112, 116, 118, 120, 216–231; data from LiS from references 47, 50, 51, 56, 60, 66, 97, 113, 114, 116, 121, 226, 233. See the Supplementary citation data for reference details.
Figure 4 Cathode catalyst materials for polymer electrolyte membrane fuel cells. Nanoarchitectured materials are given by a circle. Background colours reflect characteristics of bulk materials. Green denotes relative strength, red relative weakness, yellow intermediate characteristics, and white no data. Absence of circle indicates no data for nanomaterial. The grey background denotes the ‘baseline’ material. Abbreviations: PGM – platinum group metals, CoS - cobalt sulphur (on non-carbon black support). Data from Pt nanoparticles from references\(^{47,50,51,60,66,177,233,234}\); data from Pt nanostructures from references\(^{47,50,51,60,66,177,233,234,236,238,243}\); data from Pt alloys (with Pd, Ni, PdAu, AuCu, Cu, Co, Fe, W, P) from references\(^{47,50,51,60,66,133,139–144,177,233,236,242,244–258}\); data from Pd and Pd alloys from references\(^{47,50,51,60,177,236,252,259–261}\); data from Fe from references\(^{47,50,51,60,66,85,146,262,263}\); data from Nb, Ta, Zr from references\(^{47,50,51,60,66,139–144,177,233,236,242,244–258}\); data from CoMo, CoS from references\(^{47,50,51,60,177,233,234,271}\); data from N-doped carbon from references\(^{50,51,60,180,272–274}\); data from N-, S-, B-, P-, I-, S-, Se- multi-doped carbon from references\(^{50,51,56,147,149,275–279}\). See the Supplementary citation data for reference details.
Figure 5 Catalyst support materials for polymer electrolyte membrane fuel cells. Nanoarchitectured materials are given by a circle. Background colours reflect characteristics of bulk materials. Green denotes relative strength, red relative weakness, yellow intermediate characteristics, and white no data. The grey background denotes the ‘baseline’ material.

Box 1 Life cycle assessment

Life cycle assessment (LCA) is an analytic method for estimating the environmental impacts associated with the production and consumption of products and services. This method first strives to inventory all exchanges with the environment necessary to deliver a function, considering the material and energetic inputs required at all stages, from raw material extraction, to processing and manufacturing, to product use, recycling, and final disposal. The total emissions and resource use associated with the delivery of a functional unit (e.g., transporting one person over one kilometre) are thus compiled in a lifecycle inventory. Examples of such emissions include carbon dioxide, methane, particulate matter and volatile organic carbons. These inventoried emissions are then linked to potential environmental impacts, such as climate change, eutrophication, acidification and ecotoxicity, using characterization factors determined by modelling, experimental results or physical properties. These potential environmental impacts, also referred to as midpoint indicators, may be further characterized based on their negative effects on key areas of protection, or endpoint indicators, as valued by humanity: damage to human health, damage to ecosystems, and damage to resource availability. Unfortunately, current characterization methods do not provide characterization factors for quantifying the impact of emissions of different nanomaterials in the environment. Nevertheless, despite data limitations and important sources of uncertainty, LCA provides a useful “whole system” perspective over entire supply chains. This perspective helps identify environmental “hotspots” and the processes where efficiency measures would have greatest effect.