

# Towards greener and more sustainable batteries for electrical energy storage

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**Ever-growing energy needs and depleting fossil-fuel resources demand the pursuit of sustainable energy alternatives, including both renewable energy sources and sustainable storage technologies. It is therefore essential to incorporate material abundance, eco-efficient synthetic processes and life-cycle analysis into the design of new electrochemical storage systems. At present, a few existing technologies address these issues, but in each case, fundamental and technological hurdles remain to be overcome. Here we provide an overview of the current state of energy storage from a sustainability perspective. We introduce the notion of sustainability through discussion of the energy and environmental costs of state-of-the-art lithium-ion batteries, considering elemental abundance, toxicity, synthetic methods and scalability. With the same themes in mind, we also highlight current and future electrochemical storage systems beyond lithium-ion batteries. The complexity and importance of recycling battery materials is also discussed.**

Today, global warming, energy production and energy storage are all popular topics of discussion in society. To cope with the energy demands of the ever-increasing global population, we must double our present rate of energy production of 14 TW by 2050 (refs 1,2). This equates to 130,000 TWh or the equivalent of  $10^{10}$  tons of oil yearly (Fig. 1). This increase, which must be achieved without increasing CO<sub>2</sub> emissions or relying solely on finite fossil-fuel resources, has made the development of advanced renewable energy technologies a global imperative.

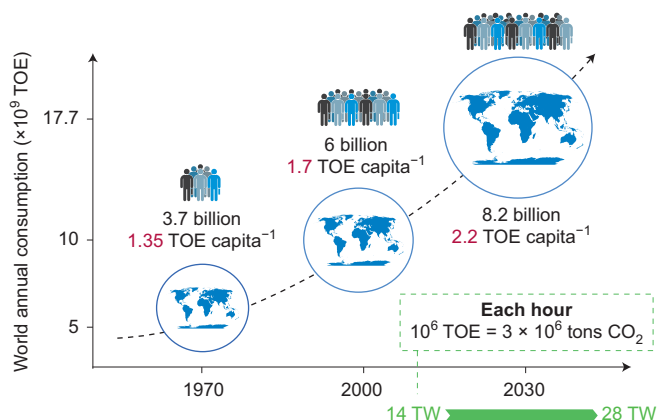
There are several renewable energy sources, for example, wind, solar, tidal, biomass and geothermal, but these are all inherently intermittent and generally dispersed<sup>3</sup> relative to the isolated large-scale facilities that currently supply the vast majority of electrical energy. To make the best use of these energy sources, we need good energy-storage systems. Unfortunately, we currently only have the capacity to store around 1% of the energy consumed worldwide, most of which (98%) is through pumped-storage hydroelectricity<sup>1,2</sup>. While such hydroelectricity is widely used, it stores just 3 Wh for every 1 m<sup>3</sup> (ton) of water lifted by 1 m. This nicely illustrates other overriding selection parameters that must be taken into consideration besides energy density, such as capital cost per cycle, scalability, safety, ease of use and cost of maintenance. It is also important to keep in mind that durability of the system can mitigate, compensate or even annihilate extra cost. Before shifting from a fossil-fuel economy to one based on renewable technologies, there is clearly a pressing need to significantly improve our ability to store energy. If we are unsuccessful in jointly capturing, managing and storing energy at a large scale and low cost, our only recourse will be to drastically reduce our total energy consumption. Considering the increasing per capita energy dependence and consumption over recent years, significant decreases in energy use over a short time period would prove difficult to achieve in our modern society.

In this Review, we introduce the concept of sustainability within the framework of electrochemical storage by discussing the state-of-the-art in Li-ion batteries and the energy cost of their electrode materials. Guided by element abundance and toxicity, we offer guidelines

for identifying sustainable electrode candidates. Having identified desirable electrode materials, available synthetic methods are discussed along with relative scalability and sustainability ratings for each method. We then present a comparison of existing, upcoming and future electrochemical storage systems beyond Li-ion batteries; new developments, challenges and promising research directions are highlighted for each technology. Finally, in the context of life-cycle analysis (LCA), we discuss the pressing need for recycling efforts to increase the overall sustainability of battery technologies.

## Costs of electrochemical storage

Perhaps, the most important consideration in our market economy is the cost associated with producing conversion and storage devices, such as photovoltaic cells and batteries, which is now



**Figure 1 | Past, present and forecast of the world's energy needs up to 2050.** With the changing lifestyles of an increasing number of inhabitants, our energy rate demand will double from 14 TW (2010) to 28 TW (2050). TOE = ton of oil equivalent. Map: © Macmillan Mexico/Haide Ortiz Ortiz, Mario Enrique Ramírez Ruiz.

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**Box 1 | Green chemistry and sustainability.**

Within this Review, the term 'green' is used to identify a process that mainly relies on environmentally friendly and benign chemicals. This idea of 'green' processes is well explained in the context of ionic liquids in the following quote by T. Welton: "For instance, we would now recognize that if the use of a more hazardous solvent gave significant improvements to the total environmental impact of the overall process than a less hazardous alternative, then the greener choice is the former"<sup>112</sup>. At this stage, let us be reminded that the goal of chemistry is to produce molecules or substances that do not naturally exist, or to produce natural ones faster or in larger quantity than nature does. 'Greener chemistry' would therefore be a more appropriate term than 'green chemistry' for current efforts in this direction — provided that life-cycle analysis calculations also support the beneficial nature of these 'green' process modifications.

The term 'sustainability' is used to refer to a technological or scientific development meeting the needs of current society without compromising the ability of the future generations to meet their own needs, in accordance with the 1987 Brundtland Report entitled *Our Common Future*, which raised awareness of the need for sustainable development in light of the environmental issues stemming from industrialization and growth<sup>113</sup>.

driven by both the abundance of the materials and the fabrication processes. It is essential to consider sustainability, renewability and 'green chemistry'<sup>74</sup> when selecting materials for storage devices (for example, electrodes, catalysts), especially when used in applications with large markets and volume (vehicles, grid). Inevitably, the proliferation of renewable energy sources will rely heavily on innovations at the materials level (Box 1).

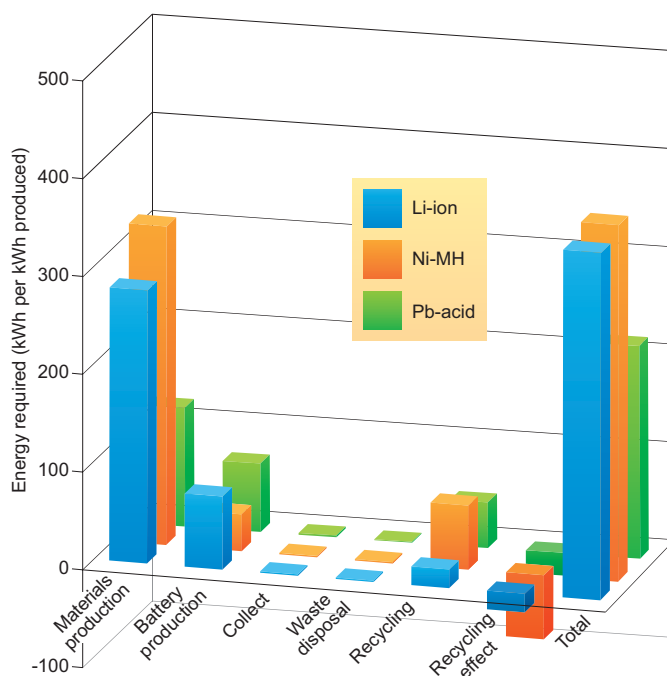
Using fuel cells to convert energy from chemical fuels obtained from renewable sources is an attractive route to more sustainable energy use. They are not limited by Carnot's principle, and hence, in theory, could have significantly higher efficiencies compared with thermal systems. Furthermore, the nature of oxidants and level of CO<sub>2</sub> emissions in fuel cells depend on the fuel used. For example, in a hydrogen fuel cell, there are no direct CO<sub>2</sub> emissions and H<sub>2</sub>O is produced. Vapour H<sub>2</sub>O is a powerful greenhouse gas, but when a maximum threshold limit in atmospheric pressure is reached, it rains; the production of water by fuel cells thus has no additional effect on climate change. In contrast, CO<sub>2</sub> accumulates in the atmosphere due the significant difference in rates of CO<sub>2</sub> release and CO<sub>2</sub> capture (by photosynthesis or sequestration and dissolution in oceans). While hydrogen fuel cells are CO<sub>2</sub> free, there is no sustainable (non-fossil fuel) and economical way to obtain the pure H<sub>2</sub> fuel required for use in these systems. Although fuel cells are an attractive technology, there are several materials challenges (Pt-based catalyst, lifetime, H<sub>2</sub> production and storage) and high costs that need to be overcome before they can be commercially used<sup>5</sup>.

The attractiveness of Li-ion battery technology resides in its versatility; it covers a wide range of applications requiring dozens of watt-hours (portable electronics), dozens of kilowatt hours (electric vehicles) and tens of megawatt-hours (grid applications), with design capabilities to meet autonomy and power requirements. However, developing Li-ion batteries for transport and that are able to deal with society's fluctuating energy needs is a formidable challenge, especially from a materials perspective. In addition to the classical figures of merit (specific energy and power, lifetime, cost and safety), other issues are not yet fully recognized, such as the low relative abundance of materials (lithium is already viewed by some alarmists as the gold of this next century) and the large energy cost of battery manufacture and recycling.

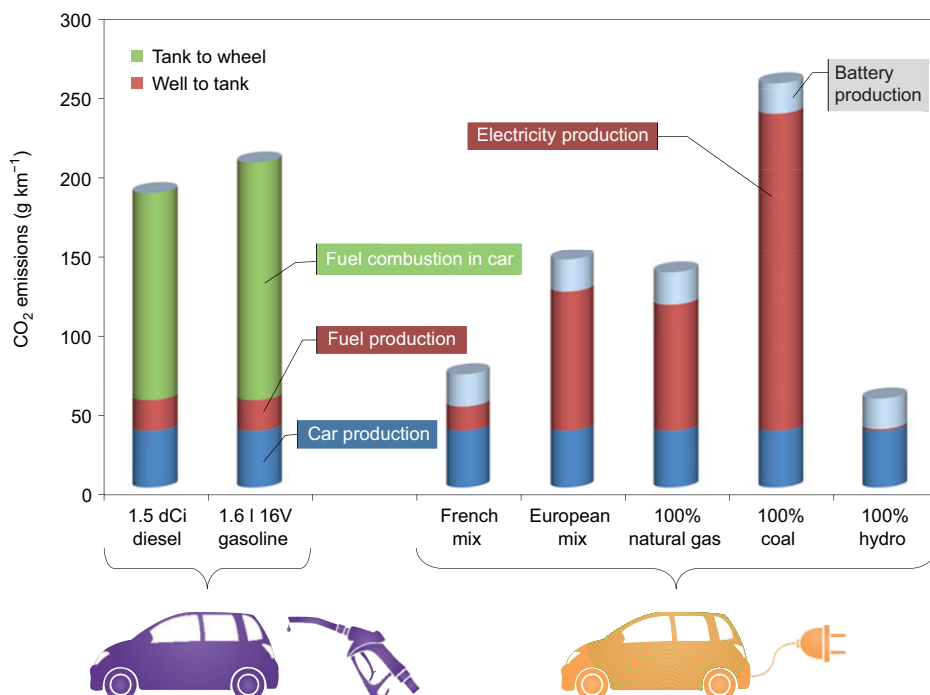
In addition to the foreseen shortage of some of the chemical elements currently used in electrodes (for example, cobalt), early integrated LCA estimations<sup>6–9</sup> (for example, taking into account the production of batteries, materials and recycling) have revealed that more than 400 kWh are needed to make a 1 kWh Li-ion battery (Fig. 2), resulting in the emission of about 75 kg of CO<sub>2</sub> — as much as burning 35 l of gasoline. In comparison, the production of 1 kWh of electricity from coal produces around 1 kg of CO<sub>2</sub> (refs 10,11). Despite the fact that the CO<sub>2</sub> generated by battery manufacturing is undesirable, the associated energy cost is still a bigger concern.

In other words, under these conditions, batteries will only begin to have an environmental benefit beyond hundreds of cycles. This also questions the benefits of developing batteries for electric vehicles to decrease greenhouse-gas emissions when we heavily rely on coal-fired plants to produce primary electricity. Figure 3 shows that a standard internal combustion engine car can release less CO<sub>2</sub> per kilometre than an electric vehicle recharged with electricity coming from coal combustion. This comparison takes into account recently reported efficiencies for the combustion of various fossil fuels and burning conditions<sup>10,11</sup> as well as the degradation of energy during electricity transport (~10%) and could therefore evolve depending on the progress made in these energy processes. However, coal has the highest carbon content (and the lowest hydrogen content) among fossil fuels, and is thus the worst choice in terms of CO<sub>2</sub> emissions per kWh produced (twice that of methane).

At this point it is worth noting that early LCA reports dealing with energy-storage systems have mainly focused on energy demands and their global warming potential (that is, how much CO<sub>2</sub> they emit). They have generally discarded other concerns such as NO<sub>x</sub>, SO<sub>x</sub>, dusts and particulates (PM<sub>10</sub>), heavy metals, and soil and water contamination, which can have deleterious health impacts or global and local environmental effects. Gladly, more recent analyses now take into account more diverse aspects and their various impacts<sup>12–15</sup>. Despite the very large discrepancy in reported LCA data regarding the relative impacts of materials production versus cell assembly, it



**Figure 2 | Energy required for the production of a 1kWh electrochemical storage system.** Data are from refs 6–9 and compare the energy cost for Li-ion, Ni-MH (nickel-metal hydride) and Pb-acid technologies. Materials production is clearly the main contributor to the energy cost of producing an electrochemical storage system.



**Figure 3 | The CO<sub>2</sub> benefit of electric vehicles relies heavily on the origin of the electricity.** Total CO<sub>2</sub> emissions (g km<sup>-1</sup>) for internal combustion engine cars (left) and for full electric vehicles (right) with various electricity origins (data from refs 6–11). We assumed that electric vehicles are used at a rate of 10,000 km yr<sup>-1</sup>, powered by Li-ion batteries (20 kWh pack, 8-yr lifespan) and consume 20 kWh per 100 km. The main contributors of the European electricity mix are: fossil fuels and waste combustion (53%), nuclear (25%) and renewable energies (hydro, wind, 21%). The main contributors of the French electricity mix are: nuclear (80%), renewable energies (hydro, wind, 11%), and fossil fuels and waste combustion (9%)<sup>11</sup>. The ‘well to tank’ step refers to the fuel production and delivery while the ‘tank to wheel’ step deals with the car operation (that is, fuel combustion). Cars: © iStock/Thinkstock.

is clear (Fig. 2) that ‘materials production’ is a main contributor to CO<sub>2</sub> emissions and energy costs. It includes various aspects such as mining, ore transport, ore treatment, and so on. In fact, a large part of this energy is utilized in mining scarce elements and achieving the high temperatures required to make the electrode materials<sup>16,17</sup>.

Consequently, the only viable path towards a ‘greener and more sustainable’ battery is rooted in our ability to design electroactive materials that have comparable performances to today’s electrodes, but cost less energy and release less CO<sub>2</sub> during production.

There are various approaches that have been explored towards this goal: (1) the development of novel eco-efficient processes<sup>18</sup> such as hydro-, solvo- and ionothermal<sup>19,20</sup> and bio-inspired<sup>21–23</sup> approaches for the synthesis of inorganic compounds; (2) the promotion of a new concept of renewable electrodes based on the use of organic compounds synthesized using ‘green chemistry’<sup>24,25</sup> from natural resources; and (3) the development of new technologies beyond Li-ion batteries such as Li–S and Li–air (Li–O<sub>2</sub>; ref. 26), Al–air<sup>27</sup>, Na-ion<sup>28</sup>, Mg, Ca<sup>29</sup> and redox-flow systems<sup>30</sup>, in combination with an increasing interest in recycling processes. Li–O<sub>2</sub> cells are often synonymously called Li–air cells even though they currently use pure O<sub>2</sub> rather than ‘air’.

Prior to discussing the details of these approaches, we show that they can be justified by cost, chemistry and availability of elements.

### Battery technologies versus element abundance

The abundance of an element is just one of the many criteria controlling its cost and availability. Hence, selecting elements from which to design eco-efficient electrodes or new technologies necessitates attention to various other chemical and industrial aspects.

For instance, as the cathode in Li–S systems, sulfur is a very cheap (US\$150–200 per ton) and abundant (~500 ppm in lithosphere) material, which is produced in its elemental state at a present rate of about 60 million tons per year (2012). Most of its production (75%) now

occurs via the Claus process<sup>31</sup>, which uses sulfur-containing side products from the oil and gas refining industry (for example, H<sub>2</sub>S) as precursors. Thus, a shortage of sulfur in the near future adversely affecting the field of energy is highly unlikely. The same applies for sulfate salts, which provide potential cathode materials for Li-ion batteries, as will be addressed later. They can be produced from sulfur and sulfides or easily extracted from wealthy deposits (for example, gypsum).

O<sub>2</sub>, as the oxidant in Li–air cells, has to be extracted from air to eliminate N<sub>2</sub>, CO<sub>2</sub> and water, all of which react with lithium or cell components. So, although oxygen is an abundant and freely available element, its use will require either on-site or on-board purification, which is energetically expensive.

In contrast to oxygen, sulfur, carbon and nitrogen, the biogeochemical natural cycle of phosphorus is referred to as ‘open’. This means that it naturally accumulates, providing a readily available source in the form of huge geological deposits of phosphates.

Thus, availability wise, designing Li-based technologies using oxygen, sulfur, PO<sub>4</sub><sup>3-</sup> or SO<sub>4</sub><sup>2-</sup> does not present any long-term risk of materials shortage. This justifies the huge research investment in Li–air and Li–S technologies. Also, ternary compounds based on SO<sub>4</sub><sup>2-</sup> or PO<sub>4</sub><sup>3-</sup> are appealing, provided the right choice of transition-metal cation is made. This is exemplified by LiFePO<sub>4</sub>, which is currently the most attractive electrode material, sustainability wise, for large-scale applications.

Forecasting the availability of materials is not an easy task, as commodity databases are not especially accurate and significant fluctuations may occur over time depending on trends in energy-related and other sectors. As an example, cobalt-based materials may soon enter the permanent-magnetism market owing to the foreseen shortage in rare-earth metals partly attributed to geopolitical issues. Such a situation is a true burden, and concerns about the long-term availability of a few elements have naturally driven researchers towards titanium, iron and manganese as 3d metals for electrode materials.

H																		He
Li	Be											B	C	N	O	F	Ne	
Na	Mg											Al	Si	P	S	Cl	Ar	
K	Ca		Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr		Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	*	Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	**	Lr	Rf	Db	Sg	Bh	Hs	Mt	Uun	Uuu	Uub		Uuq				

**Figure 4 | Mismatch between the elements constituting biomass and the main constituents of our present Li-ion batteries.** Biomass (that is, naturally recycled) elements are shown in green (main in dark green, minor in light green, traces in pale green)<sup>32</sup>. Present Li-ion batteries main elements are circled in red. Aside from the benefit of organic batteries, this chart shows for each block (s, p, d) the elements we should select to prepare environmentally friendly compounds. The arrows indicate the evolution the battery-constituting elements should follow to better match sustainability issues. Note that only light elements are present in both biomass and battery technologies.

Regarding the choice of the guest electropositive element among alkali and alkaline earth metals, the situation is much simpler because their cost is mainly governed by their abundance. As they have strong similarities in reactivity, it may be possible to adapt industrial processes to accommodate congener elements. This strongly validates the move towards post-Li chemistries based on sodium and potassium, and encourages increased efforts on magnesium and calcium chemistries.

It is certainly not a coincidence that all the elements highlighted above with respect to sustainability (H, O, S, P, Na, K, Ca, Mg, Mn, Fe, Ti) are among the main constituents of biomass<sup>32</sup>, and are thus naturally (re)cycled within the biosphere through exchanges with other compartments, in contrast to the main constituents of our current Li-ion batteries (for example, Li, Co, Ni, Cu, F) (Fig. 4).

### Electrode challenges for Li-ion chemistry

The selection of green, cheap and available elements as discussed above does not automatically guarantee that the final material or the final storage system will be green and cheap. Raw materials have to be extracted and processed, the electrode and electrolyte materials need to be synthesized, devices must be fabricated, and then all of the components should be recyclable. These various steps (equation (1)), can potentially have deleterious effects on the eco-efficiency of the final system; ergo, the need to consider these individuals steps as described next for various battery technologies, starting with Li-based batteries.

Total impact of the cell =

$$\sum_{\text{Components}} \text{Chemical composition} + \sum_{\text{Components}} \text{Synthesis process} + \sum_{\text{Components}} \text{Implementation in the system} + \sum_{\text{Components}} \text{Recycling} \quad (1)$$

**Chemical composition.** The quest for storage systems with higher energy densities has led over the years to the move from aqueous (such as in Pb-acid and nickel-based batteries) to non-aqueous based electrolytes (such as in Li-ion batteries), enabling much higher operating voltages. A great advantage of the Li-ion battery, as opposed to Pb-acid, Ni-Cd and Ni-metal hydride batteries, is its versatility with respect to the wide range of positive and negative electrodes that can be used, which offers possibilities in terms of designing new high-performance electrodes based on low-cost

elements. Research on positive electrodes is presently divided between improving layered oxides and designing new polyanionic compounds. Layered LiCoO<sub>2</sub> has long been the benchmark, but it is progressively being replaced by NMC (LiNi<sub>1/3</sub>Mn<sub>1/3</sub>Co<sub>1/3</sub>O<sub>2</sub>)<sup>33</sup>, where partial substitution of cobalt for manganese and nickel preserves the voltage but provides a higher capacity (200mAh g<sup>-1</sup>). Furthermore, driven by cost and abundance issues, the manganese content can be increased by enlisting Li-rich layered composite [(1 - z) LiLi<sub>1/3</sub>Mn<sub>2/3</sub>O<sub>2</sub> + (z) LiMn<sub>0.5-y</sub>Ni<sub>0.5-y</sub>Co<sub>2y</sub>O<sub>2</sub>] phases<sup>34</sup>, which exhibit capacities as high as 300 mAh g<sup>-1</sup>. This is an example where a single chemical approach can simultaneously combat performance and sustainability issues.

Polyanionic compounds, where O<sup>2-</sup> is replaced by PO<sub>4</sub><sup>3-</sup>, SO<sub>4</sub><sup>2-</sup>, SiO<sub>4</sub><sup>4-</sup> or BO<sub>3</sub><sup>3-</sup> anions, also offer promise for sustainable electrode materials as they readily form compounds with iron. Although the Fe<sup>3+</sup>/Fe<sup>2+</sup> redox couple operates at relatively low potentials compared with Co<sup>4+</sup>/Co<sup>3+</sup> or Ni<sup>4+</sup>/Ni<sup>3+</sup>, researchers have successfully implemented the inductive effect proposed by J. B. Goodenough<sup>35</sup> to design polyanionic compounds, such as LiFeSO<sub>4</sub>F, which displays 3.9 V Fe<sup>3+</sup>/Fe<sup>2+</sup> versus Li<sup>+</sup>/Li<sup>0</sup> — the highest voltage so far reported in inorganic compounds<sup>36</sup>. Turning to the negative electrodes, where the use of sustainable materials is equally important, the trend has been towards the use of (1) carbonaceous, (2) titanium-based (TiO<sub>2</sub>, Li<sub>4</sub>Ti<sub>5</sub>O<sub>12</sub>) or (3) silicon electrodes. Even though the production of silicon requires energy-consuming reduction of silica, it is a readily available biomass element.

The composition of the electrolyte must also be considered. While Li-ion systems use electrode materials that no longer contain toxic lead and cadmium, their electrolytes do pose toxicity issues. The use of lithium-based electrolytes containing costly fluorine-based anions such as PF<sub>6</sub><sup>-</sup>, bis(trifluoromethane)sulfonimide (TFSI<sup>-</sup>) and bis(fluorosulfonyl)imide (FSI<sup>-</sup>) present significant hazards, out of which PF<sub>6</sub><sup>-</sup> is the most hazardous. Under specific conditions, toxic or reactive products such as fluoroethanol ether (FCH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>O (ref. 37), HF (ref. 38) and fluoroethylene CH<sub>2</sub>F (ref. 39) can be generated, motivating intensive research efforts towards the use of fluorine-free salts.

A growing tendency in both energy storage and conversion systems is to imitate nature and its vast biodiversity. Such biomimetic and bio-assisted chemistry approaches have the advantage of using enzyme-catalysed room-temperature methods to design new materials with multifunctionalities, sophisticated micro- and nano-structures, and enhanced chemical and physical properties. This is



beautifully exemplified by the colossal efforts being dedicated to artificial photosynthesis with the hope of achieving order(s) of magnitude greater efficiencies than occur in natural photosynthesis. The field of batteries is now moving in a similar direction, taking inspiration from life chemistry, which is powered by chemical reactions that are dependent on ion flux and membrane potential. Among them are redox reactions that enlist, for instance, ketone groups for fast proton–electron coupled transfer reactions. Bearing in mind such a comparison, a survey was initiated of redox-active molecules capable of reacting with lithium via a (de)insertion mechanism, and that could be synthesized using ‘greener chemistry’ principles from renewable resources<sup>40,41</sup>. An oxocarbon molecule with the formula  $\text{Li}_2\text{C}_6\text{O}_6$  that could be synthesized from a renewable resource (myo-inositol, which contains four carbonyl–quinone double bonds) was identified. This molecule reversibly (de)inserts four  $\text{Li}^+$  ions<sup>24</sup> (providing a reversible capacity of  $580 \text{ mAh g}^{-1}$ ) at around 2.8 V, leading to an energy density of  $1,500 \text{ Wh kg}^{-1}$  of electrode versus  $\text{Li}^0$ , nearly twice what can be achieved with today’s NMC electrodes ( $880 \text{ Wh kg}^{-1}$ ). This approach was rapidly validated by the discovery of conjugated carboxylates such as  $\text{Li}_2\text{C}_8\text{H}_4\text{O}_4$ , which (de)inserts  $\text{Li}^+$  ions at low potential (0.7 V; ref. 41) and has a reversible capacity of  $320 \text{ mAh g}^{-1}$ . Breaking the old taboos, numerous reports have now demonstrated that Li batteries based on organic molecules can have both high rate capability and long cycle life, although the latter is still less than what has been achieved with inorganic electrodes.

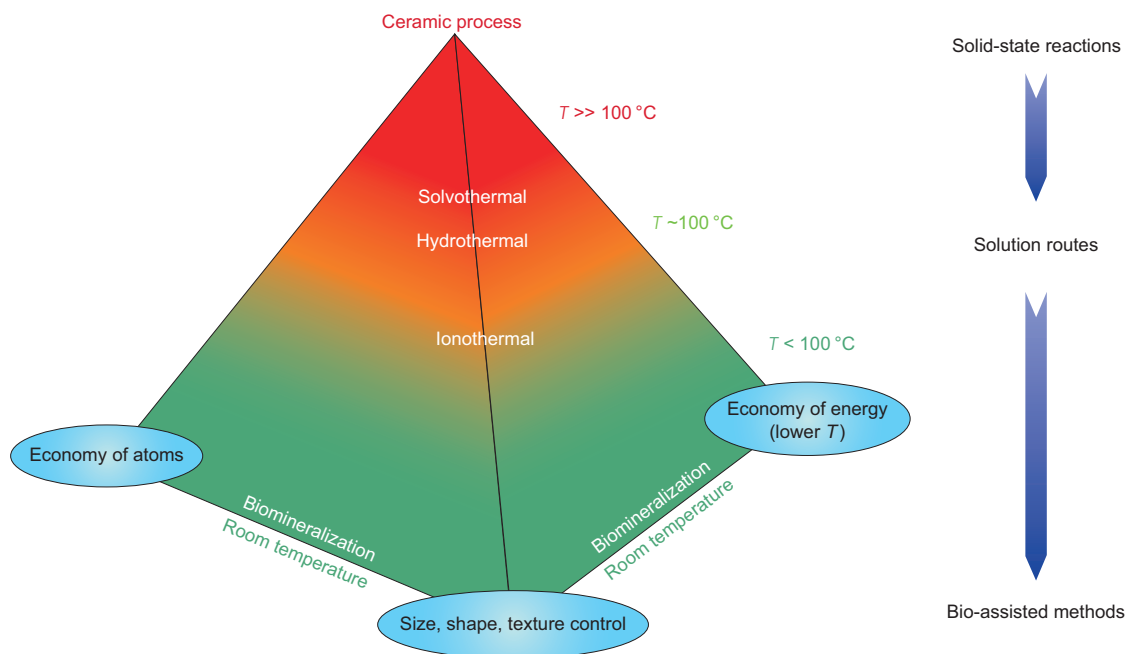
Pursuing this trend, other groups are exploring the design of novel tunable hybrid systems for energy through inspiration from bioenergetics. Kang’s group has recently reported the electrochemical activity of riboflavin<sup>42</sup>. This molecule, also known as vitamin B2, has been shown to reversibly react with two  $\text{Li}^+$  ions per unit formula, hence leading to capacities of  $174 \text{ mAh g}^{-1}$ , a value comparable to that of  $\text{LiFePO}_4$  — the material of choice for the next generation of Li-ion batteries for electric vehicle application. However, owing to the low volumetric density of organic molecules compared with inorganic compounds, such approaches will not be competitive

in terms of volumetric energy density. Biological systems rely heavily on sodium-, potassium- and magnesium-containing organic complexes, so it will not be a surprise if such complexes are soon used in either Na or Mg batteries. In fact, Na-based organic carboxylate electrodes have already been reported<sup>43–45</sup>.

Organic batteries with minimal  $\text{CO}_2$  footprints, assuming all other challenges (materials solubility, finding a highly oxidizing Li-based positive electrode, and so on) are overcome, should enable the use of Li-ion batteries for large-scale applications. With its vast examples, bioenergetics can continue to provide the necessary inspiration to researchers for new synthetic opportunities.

**Synthesis processes.** It is clear that high-temperature processes, unless using solar thermal, will eventually be replaced by low-temperature synthesis methods such as ‘chimie douce’<sup>46,47</sup>. Such a synthetic approach, which is nested in the structural preservation of the precursor skeleton, provides energy savings, as it does not enlist bond breaking to trigger the formation of new compounds. This return to low-temperature chemistry was initiated back in the 1970s in reaction to the first oil crisis, and Fig. 5 highlights the positive attributes (e.g. economy of atoms, texture–size–shape controls, and so on) of the various low-temperature methods: hydro(solvo) thermal synthesis, sonochemistry, microwave-assisted processes, ionothermal synthesis<sup>18–20</sup> and biomineralization.

Previously disregarded by inorganic chemists, ionothermal synthesis has recently been shown to be quite efficient in preparing known electrode materials with tailored morphologies. For example, phosphates and silicates can be synthesized at temperatures close to  $200^\circ\text{C}$ , which is about  $500^\circ\text{C}$  lower than that used in high-temperature ceramic processes. Additionally, a novel family of water soluble fluorosulphates ( $\text{AMSO}_4\text{F}$ , A = alkali metal and M = 3d metal) has been synthesized by taking advantage of the wide range of chemical and physical properties of ionic liquids and the richness of their chemistry<sup>48</sup>. Of this family, the triplite polymorph of  $\text{LiFeSO}_4\text{F}$  (3.9 V versus  $\text{Li}^+/\text{Li}$ ) can be made at  $280^\circ\text{C}$  (ref. 36).



**Figure 5 | The three-fold benefits in lowering the synthesis temperature of electrode materials.** Shifting from high-temperature synthesis routes (several hundred  $^\circ\text{C}$ ) towards hydro-, solvo- and ionothermal approaches ( $\sim 100^\circ\text{C}$ ) and even biomineralization methods (ambient temperature) obviously saves energy, but also concomitantly allows an increasing control of the shape, size and texture of the solids. While ceramic routes are well established, the ionothermal alternative is quite new in the field of inorganic materials. Finally, bio-assisted strategies are based on very efficient enzymatic and active biological processes intrinsically not wasting atoms. As the synthesis temperature is decreased, this three-fold pyramidal evolution therefore progressively provides more tools and freedom for the chemist.

Although ionic liquids may at first appear to be an uneconomical reaction media, they can be easily recovered and recycled, which improves their 'green' qualities, provided they are made from non-toxic and abundant precursors. Hence, there is a need to move away from using expensive anions ( $\text{TFSI}^-$ ,  $\text{FSI}^-$ ) that are complicated to synthesize as a constituent of the electrolyte, towards the use of chloride-based ionic liquids using imidazolium (imidazole is cheap and chlorine is a biomass element<sup>32</sup>). These chloride-based ionic liquids could be good alternatives; however, work still needs to be done to optimize their synthesis, stability and solvation properties. The LCA of ionothermal synthesis has yet to be performed for estimating the positive balance, if any, between the assets and costs, recycling and toxicity of ionic liquids. Ionothermal synthesis has resulted in the discovery of novel Li-based electrode materials, and once the reaction scheme is identified, they can be synthesized using alternative methods. For example  $\text{LiFeSO}_4\text{F}$ , which was initially prepared through ionothermal synthesis, can now be made at 300 °C from a dry solid-state process in less than 20 minutes<sup>49</sup>.

Hydro-, solvo- and ionothermal reactions still require heating to 100–280 °C, which is less sustainable compared with methods enlisting room-temperature synthesis. Scientists often turn to biological systems for inspiration when looking for near-room-temperature synthetic approaches. In fact, biomineralization using *Bacillus pasteurii* bacterium for precipitating calcite has long been known and used<sup>50–53</sup>. Recently, Ghodssi's group reported the fabrication of nanostructured  $\text{V}_2\text{O}_5$  cathodes and nanotubes of silicon for anodes using the tobacco mosaic virus (TMV) as a biotemplate<sup>54,55</sup>. Another elegant example of bioinspired materials used in batteries comes from Belcher's work<sup>22,23</sup>, where genetically engineered viruses are used as templates to make high-rate  $\text{FePO}_4$  single-walled carbon nanotube electrodes at room temperature. This pioneering work however has some practical limitations: the approach is so far unable to allow the formation of Li-based electrodes, mandatory for a practical Li-ion cell, and cannot be used on a large scale. Hence, there is a need for the development of other bioinspired strategies that can be easily scaled up to make Li-based electrode materials<sup>56</sup>. Among them, the use of mineralizing bacteria carries great hope<sup>57</sup>, especially considering the well-established scalability of such assisted processes in industry.

These challenges call for a collaborative effort between inorganic, organic and biochemists to develop innovative and diversified synthetic approaches to optimize existing materials and design new materials for batteries. To realize the scaling feasibility of the various approaches, LCA remains to be done. However, considering the variety of new low-temperature synthesis methods being developed and the large set of unexplored combinations of elements in our redefined 'sustainable periodic table', there are many possibilities yet to be explored.

**Material implementation.** The integration of materials in batteries enlists the formulation of composite electrodes consisting of an active material, conducting additives, thickeners, binders, and so on. Such a formulation is a science in itself that calls for (1) selection of chemically compatible ingredients, (2) adjustment of the parameters of mixing fluids and additives for the control of the rheology of the suspension and (3) adjustment of the ratio of the various components to reach an optimum porosity to ensure an ideal compromise between electrode ionic conduction and electronic percolation once the solvent is evaporated. For sustainability reasons, there is currently a strong drive to depart from the use of polyvinylidene fluoride-based binders (copolymers of vinylidene difluoride) that require environmentally hazardous N-methyl-2-pyrrolidone as the solvent, and to develop water-based processes. Water-soluble carbohydrate-based salt binders (for example, carboxy-methyl cellulose) are becoming commonly used for making water-based inks

and silicon-based electrodes<sup>58–61</sup>, but such approaches cannot be universalized as some of today's electrodes are moisture sensitive or water soluble.

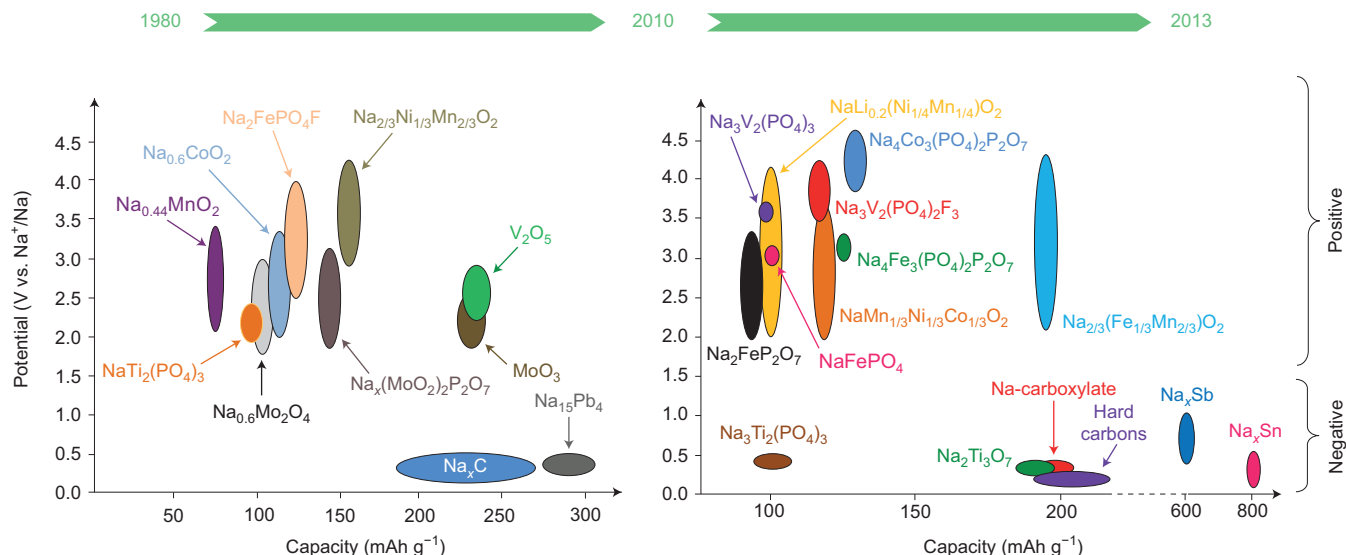
### Other Li-based chemistries

**Li–O<sub>2</sub> and Li–S.** Another option towards more sustainable battery systems is moving to metal–air systems (Li–, Na– and Mg–air batteries) using O<sub>2</sub> as the positive electrode, which is similar to the concept of fuel cells<sup>62–64</sup>. The positive electrodes in such systems are generally composed of black carbon and manganese oxide catalysts; therefore, they are much cheaper than the cobalt-based positive electrodes currently used in Li-ion batteries. Li–O<sub>2</sub> batteries have theoretical energy densities about 10 times higher than most batteries in the market today, but only two-fold improvements have been practically reached to date. The automotive transportation community thus views Li–O<sub>2</sub> systems, on which early work dates back to the 1970s<sup>65</sup>, as our 'saviours', generating a colossal amount of research worldwide. Considerable progress has been made over the past three to four years. For instance, a few tens of cycles was achieved by Sun's group using an appropriate carbon–electrolyte combination<sup>66,67</sup>. This was further confirmed by Bruce's group, which recently succeeded in achieving more than 100 cycles with excellent capacity retention, high coulombic efficiency and low polarization using a porous gold electrode<sup>68</sup> and more recently a TiC electrode<sup>69</sup>. Such findings, although not viable for practical applications, have at least the merit to demonstrate that such a system can work and be made practical provided further breakthroughs are made in terms of electrolytes and catalysts. However, in spite of its early excitement, the battery community realizes that commercial Li–O<sub>2</sub> cells still have a long way to go, both at the basic research and applied levels.

Moving away from metal–O<sub>2</sub> systems, scientists are now revisiting Li–S technology, early reports of which date back to the 1960s<sup>70,71</sup>. This technology displays an output voltage of 2.1 V, with a theoretical energy of 2,500 Wh kg<sup>−1</sup>, but has inherited classical drawbacks from earlier Li-based systems. These include the formation of Li dendrites that could cause short circuits and eventually explosion hazards, and the formation of soluble polysulfides<sup>72,73</sup> that can react with lithium to precipitate electronically and ionically insulating inactive  $\text{Li}_2\text{S}$ , which leads to poor capacity retention and limited kinetics. Numerous methods for their improvement have been tested, including: (1) confinement approaches to trap polysulfides<sup>74</sup>; (2) additives such as  $\text{LiNO}_3$  to passivate the Li electrode<sup>75</sup>; and (3) elaborating metallic Ti-based oxide–sulfur composites loaded with 70% sulfur. The third method looks promising, as a fade rate of 0.06% cycle<sup>−1</sup> after 500 cycles was achieved<sup>76</sup>, indicating that this technology is already more mature than the Li–O<sub>2</sub> system and even better results are expected to follow.

Whether it is the Li–O<sub>2</sub> or Li–S battery technology in question, safety could be enhanced by (1) replacing the metallic lithium negative electrode by a lithium alloy (for example,  $\text{Li}_x\text{Si}$ ), which is already being explored<sup>77</sup> and (2) pursuing the development of polymeric electrolytes, which hinder dendritic growth of lithium. Additionally, the risks of H<sub>2</sub>S release under uncontrolled operation result in safety concerns, with little hope for improvement. Nevertheless, this drawback should not be considered a deterrent. Indeed, the Na–S technology, which has similar risks, is the most widely used battery technology for large-scale applications.

Both non-aqueous Li–O<sub>2</sub> and Li–S batteries suffer from a lack of suitable electrolytes — the nucleophilic attack of the electrolyte by O<sub>2</sub><sup>•−</sup> (superoxide) is a problem in the former and the control of polysulfide solubility is an issue in the latter. An attractive solution could rely on the use of ceramic membranes, which give great versatility in selecting the electrolytes (solvents), to realize two-compartment cells. This would mean that less demand would be put on their electrochemical voltage stability window. Reversible aqueous Li–air systems using inexpensive and abundant OH<sup>−</sup> anions rather than costly



**Figure 6 | Recent acceleration in the number of reported electrode materials for sodium-based batteries.** Voltage versus capacity for reported positive and negative electrode materials with plausible application in Na-ion cells within the past three decades (1980–2010; left) and as studied during the past three years (2010–2013; right).

and hazardous fluorine-based anions ( $\text{PF}_6^-$ ,  $\text{TFSI}^-$ ,  $\text{FSI}^-$ ) are feasible, as demonstrated by S. Visco *et al.* (ref. 78 and presentation at the 42nd Power Sources Conference). Using a  $\text{Li}^+$ -conducting but electronically insulating membrane, such as  $\text{Li}_{(1+x+y)}\text{Al}_x\text{Ti}_{2-x}\text{Si}_y\text{P}_{(3-y)}\text{O}_{12}$ , and an accompanying interfacial ( $\text{Cu}_3\text{N}$ – $\text{Li}_3\text{N}$ ) joint for controlling the interface, the authors successfully assembled primary aqueous Li–air batteries and demonstrated the feasibility of recharging them, although the number of recharges without large decays is so far quite limited. With a similar concept design and taking advantage of  $\text{Li}_2\text{S}$  solubility in water, the same group has recently demonstrated a 3 V aqueous rechargeable Li–S battery showing good capacity retention, hence overcoming issues related to the precipitation of electronically insulating  $\text{Li}_2\text{S}$  at both positive and negative electrodes in non-aqueous cells<sup>79</sup>.

High sustainability could definitely be achieved by considering the corresponding sodium-based technologies, assuming that new sodium-based ceramic membranes are designed. Making such membranes insoluble in water will be the ultimate dream for sustainability, as they will open the development of aqueous Na–air batteries that could use sea water as the electrolyte. Such demands are presently rejuvenating the field of inorganic solid-state electrolyte research, with great advances such as the discovery of new room-temperature superionic lithium-based  $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$  (ref. 80; conductivity ( $\sigma$ ) =  $1.2 \times 10^{-2} \text{ S cm}^{-1}$ ),  $\text{Li}_6\text{PS}_5\text{X}$  (X = Cl, Br, I) argyrodites (ref. 81;  $\sigma$  =  $1.3 \times 10^{-3} \text{ S cm}^{-1}$ ) and sodium-based  $\text{Na}_3\text{PS}_4$  (ref. 82;  $\sigma$  =  $10^{-4} \text{ S cm}^{-1}$ ) solid electrolytes for room-temperature rechargeable lithium and sodium batteries. Nevertheless, such findings together with early work on NASICON (Na super ionic conductor) structures<sup>83,84</sup> should pave research directions towards new and attractive superionic solid oxide electrolytes made of abundant chemical elements via low-temperature processes, once the interface issues are positively addressed. Similarly, conducting membranes could also play a key role in developing semi redox-flow systems.

**Redox-flow technologies.** Life itself relies on bioenergy processes that require the flow of ions and matter, and most of the energy–ion exchanges are performed through intermembrane proteins and pumps. For instance, adenosine triphosphate, our final energy carrier, is produced via a proton flow through such ‘dam pumps’.

From a technology point of view, redox-flow systems stand midway between batteries and fuel cells<sup>85,86</sup>. They operate with redox

species stored in tanks that flow across an electrochemical cell<sup>87,88</sup>. The size of the tanks determines the amount of stored energy, while the surface of the cell and the flow rates determine its power. Depending on the nature of the redox species, redox-flow systems can have a high degree of sustainability as they operate (1) in a water electrolyte, with low cost and abundant salts, and (2) in closed loops to ensure maximal use of the reactants. Therefore, their main disadvantages reside in the heavy accompanying hardware and the limited concentration of the soluble redox species, both of which limit the energy density of the whole system. Current redox-flow batteries use solubilities ranging from less than  $2 \text{ mol l}^{-1}$  for the most frequently used redox batteries based on vanadium redox chemistry ( $25\text{--}40 \text{ Wh kg}^{-1}$ ) to  $8 \text{ mol l}^{-1}$  for the second most-used Zn–bromine systems ( $80 \text{ Wh kg}^{-1}$ ).

Recently, the semi-solid lithium redox-flow device relying on conducting inks rather than liquids as catholytes, and all organic non-aqueous Li-ion redox-flow batteries proposed by Chiang’s<sup>89,90</sup> and Jansen’s groups<sup>91</sup>, respectively, have resulted in a departure from aqueous systems, favouring (in most cases) performance at the expense of sustainability and cost. This contrasts with J. B. Goodenough’s recent concept of the alkaline redox-flow battery, which preserves an aqueous cathode in a flow mode, separated from metallic lithium by a Li-ion conducting solid electrolyte ceramic, thus enabling high voltages<sup>92</sup>. But, in anticipation of lithium supply shortages, the development of similar aqueous sodium-based hybrid redox-flow batteries<sup>93</sup> should be pursued, along with the design of new sodium-based ceramics. Irrespective of a lithium- or sodium-based redox-flow system, the economic viability of such systems solely lies in the use of aqueous media coupled with the use of either inorganic or organic conducting inks. Overall, redox-flow systems could be an attractive solution to the sustainable use of electrical energy, provided the research on such systems gains momentum and explores means to develop free membrane systems or assign additional function(s) to the catholyte and anolyte reservoirs, besides storage.

### Post-Li chemistries

The foreseen demand for lithium, dictated by the expanding electric vehicle and grid applications, brings fear of lithium shortage. It also raises geopolitical issues related to uneven global distribution of lithium around the world. This uncertain situation has led researchers



to look for both new chemistries dealing either with monovalent ( $K^+$ ,  $Na^+$ ) or divalent ( $Mg^{2+}$  and  $Ca^{2+}$ ) cations as well as means to efficiently recycle lithium. Sustainability wise, the most appealing alternative to lithium metal is sodium. The abundance of sodium in Earth's crust is about 23,000 ppm (17,000 ppm for potassium) compared with a mere 20 ppm for lithium, which is concentrated in a few mineral sources. Sodium is also present in sea water at high concentration, rendering its overall resources practically unlimited. Owing to such abundance, the price of  $Na_2CO_3$  is 50 times lower than that of  $Li_2CO_3$  and this gap is continuously widening. Performance wise, sodium is three times heavier than lithium ( $23\text{ g mol}^{-1}$  compared with  $6.9\text{ g mol}^{-1}$ ) and is 0.3 V less electropositive, so relatively high gravimetric and volumetric capacity penalties ( $\sim 15\%$ ) will have to be paid in moving from lithium to sodium batteries. Nevertheless, if Na-ion technology could be achieved, early estimates predict a 30% cost decrease with respect to Li-ion technology while ensuring sustainability. Such a cost reduction takes into account, besides the cost of the raw materials, the possibility of using aluminum rather than copper as the current collector at the negative electrode and developing cheaper sodium-based electrolytes.

Worldwide research on Na-ion technology is now flourishing and over the past three years the numbers of positive and negative electrode materials with attractive properties has nearly doubled (Fig. 6). Na-ion cells have already been assembled<sup>94</sup> and a 3 V secondary Na-ion battery that is environmentally and cost friendly while displaying sustained capacity retention has been demonstrated<sup>95</sup>. Similar to Li-ion challenges, mastering the solid electrolyte interphase through the use of new solvents, sodium-salts, additives, binders, and so on will be essential to realizing higher performances with Na-ion batteries. A systematic extrapolation of our Li-ion knowledge will therefore not be sufficient, as it has already been shown that the best electrolyte additive for Li-ion cells (vinylidene carbonate) drastically increases the lifetime of Li-ion cells, but has no effect on Na-ion ones. The opposite was observed when fluoroethylene carbonate rather than vinylidene carbonate was used as an additive<sup>96</sup>.

Interestingly, sodium salts ( $PF_6^-$ ,  $TFSI^-$ ,  $FSI^-$ ) are much less toxic than their lithium counterparts. They are also easier to obtain in their anhydrous state and easier to purify.  $Na^+$  is a milder Lewis acid than  $Li^+$  and hence could lead to solid electrolyte interphases of different compositions. Moreover,  $Na_2O_2$  is less stable than  $Li_2O_2$ , and this could be an advantage for Na- $O_2$  cells, with the feasibility to easily reach  $Na_2O$  (a four-electron process)<sup>97</sup>. Safety wise there is no indication or scientific grounds to tell whether Na-ion batteries will be safer or not than Li-ion batteries, but preliminary accelerating rate calorimetry tests suggest that they will be at least as safe as Li-ion batteries<sup>98,99</sup>. The advancement of Na-ion technology is a certainty, but the type of market it will conquer remains unclear. There is great hope for grid applications, for which weight is not a concern, and where Na-ion technology could stand as an attractive alternative to the presently commercialized high-temperature Na-S cells<sup>100</sup> and Na-NiCl<sub>2</sub> ZEBRA-type<sup>101</sup> systems, both of which take advantage of the high conductivity of solid beta-alumina ceramics at temperatures of about 300 °C. While Na-S has so far been regarded as 'stellar' battery technology for grid applications, its future may be compromised owing to a few recent safety incidents. For example, Na-S batteries were responsible for a fire that occurred at a power plant in Joso City (Ibaraki Prefecture, Japan) on 21 September 2011. Although the cause of the fire is still under investigation, this event underscores the fact that safety issues for Na-S batteries have not been completely resolved<sup>102</sup>.

Apart from Na-ion chemistry, cells based on divalent, abundant and cheap metals (magnesium and calcium) are also gaining some interest, despite lower specific capacities. Great advances have been made at the electrolyte level, rendering magnesium electrochemistry feasible; therefore, the main issue still resides in finding suitable

magnesium insertion compounds in addition to the currently used open framework  $Mo_6X_8$  ( $X = S, Se$  and  $Te$ ) structures, termed as Chevrel phases, which show sustainable capacity retention versus magnesium with a limited 1.4 V output voltage<sup>103</sup>.

Despite the fact that the  $Ca^{2+}/Ca$  standard redox potential ( $-2.87\text{ V}$  versus  $H^+/H_2$ ) stands midway between that of  $Li^+/Li$  and  $Na^+/Na$ , calcium-based cells have been less studied and this is mainly because of the lack of materials and more so electrolytes<sup>104</sup>. While such cells could prove to be sustainable, a huge amount of work is needed before we can predict whether these systems will be competitive.

## Recycling

Batteries have traditionally been considered to be potentially hazardous to the environment due to the wide use of toxic lead, cadmium, mercury, and so on. Li-ion batteries do not contain any of these materials but 3d metals such as nickel or cobalt are used in most of them. Their use together with lithium is problematic due to limited supply, their continuously increasing cost and the environmentally questionable extraction methods. Furthermore, a widespread use of Li-ion batteries for automotive or grid applications will, over time, require that either the 3d metals (cobalt, nickel) are partially substituted and/or that an efficient recovery or recycling method for batteries is adopted. In today's Li-ion batteries for portable electronics, cobalt and lithium contents range between about 5 and 15 wt% and 2 and 7 wt%, respectively<sup>105</sup>. Several hundred thousand tons of batteries are sold annually; this constitutes an 'urban mine' for the recovery of thousands of tons of metal with cost advantages over direct mining. For instance, to obtain 1 ton of lithium we need to handle 250 tons of mineral or 750 tons of brine compared with 28 tons of used batteries. The recycling process of e-waste should reduce scrap volume, separate battery components and enrich valuable metals, and eliminate or reduce the danger of waste release to the environment. At present, various classes of recovery processes are being pursued, the main ones being pyrometallurgy and hydrometallurgy processes, which proceed at high (pyrolysis) and low (solution chemistry) temperatures, respectively. The low-temperature process presents advantages, as it has high yields and is realized at temperatures ranging between 80 and 100 °C (thus saving energy). It is also not affected by the eventual loss of some elements by evaporation. The high-temperature process, in contrast, presents advantages in terms of materials purity, duration of treatments and not needing 7 m<sup>3</sup> of water per ton of treated batteries as estimated for the hydro process. Irrespective of the process, lithium-bearing metallurgical slag coming from battery recycling is now used as additives in the concrete industry<sup>106</sup>.

Keeping in mind the chemical complexity of a battery, it is expected that recycling processes will require multi-step treatments. For instance, hydrometallurgy involves acid-base leaching, solvent extraction, chemical precipitation and electrochemical processes. The economy and greenness of the recycling process must be considered, and its overall benefit must be computed (using LCA). Aware of such an issue, bio-hydrometallurgy processes using microorganisms are at present being investigated for their efficiency, low cost and manufacturing ease. For instance, *Acidithiobacillus ferrooxidans* (found in pyrite deposits) can metabolize sulfur and iron, and produce sulphuric acid to leach metals (bio-leaching)<sup>107</sup>. Other biological means (hyper-accumulators) can also be envisaged to concentrate scarce metals from contaminated soils and waters, as many plants are reported to efficiently and naturally accumulate nickel and cobalt<sup>108</sup>.

There is no doubt that the chemistry of recycling will become more important than ever in the coming decades, not only for batteries but also for the storage and conversion field in general. Although 'recycling chemistry' is not particularly fashionable today, it encompasses a discipline that is very important for the future of our planet. Efficient and practical recycling technologies devoted



to both the recovery of valuable metals and the disposal of materials harmful to the environment need to be developed. Within the field of batteries, a few companies have already acknowledged the importance of recycling, and have acted accordingly on scientific and political levels to try to set up networks for battery collection and recycling, as well as enforcing European norms regarding the minimum threshold of materials to be recovered. Recycling batteries is also starting to become accepted at the citizen level. Cost wise, for specific chemical elements (iron, manganese) the benefit of recycling can be negative. Hence, the cost of recycling will have to be borne by the customer as part of the tax on the purchase price of the battery. Aside from the set up of efficient disposal and collection networks, end-of-life batteries from electric vehicles can also be of potential interest for stationary grid storage<sup>109</sup>.

Recycling is definitively the missing link to close the loop in most of today's energy-related processes. Life is a complex combination of completely closed cycles, that is to say perfect recycling processes, without any loss or by-products, and should constitute the zero-impacts LCA reference. The reason why we are facing the present global energy and environment crisis is due to the fact that we have violated the time-matter cycle by rapidly consuming for the past 200 years fossil fuels that took millions of years to be formed and accumulated. It's time to manage our energy-resource consumption to return to eco-balance.

## Conclusions

Concerns regarding the 'green' quality of batteries have long existed and over the years these concerns have been addressed in a variety of ways. We must now address the issue of chemical toxicity in batteries head-on by identifying non-toxic element and additive alternatives with similar performance to their toxic counterparts. Along the same lines, we need to be aware that fluorine-based compounds present in Li-ion batteries are not ideal. This is the reason why researchers are eagerly looking for alternatives. Until an alternative can be found, it will be necessary to design batteries with sufficient confinement in the event of cell malfunction; looking at the  $\text{NaN}_3$ -based airbag technology can be of great help in handling such confinement and toxicity issues.

In contrast, sustainable development, which has been ignored for many years, is becoming a worldwide objective. This has resulted in the enabling of new plans and programmes that tackle shortcomings and take into consideration energy and development. Most of these programmes stress the importance of education, research and public funding to achieve sustainable production and consumption patterns. Within the field of transportation, the European Union is active by supporting initiatives, such as the European Green Cars Initiative, to promote cars, trucks and 'green' buses, to meet tomorrow's demands of cleaner and more sustainable transportation. Scientists are actively integrating the principles of sustainability in their day-to-day research. Present work on Na-ion systems, Li-ion systems based on renewable electrodes, aqueous metal-air systems and aqueous redox-flow systems are some of the many examples. Similar initiatives are being taken in other energy-related fields such as photovoltaics, where research on organic photovoltaics and dye-sensitized solar cells is gaining momentum.

There is little doubt that within the foreseeable future, the billions of batteries produced annually will have to be recycled. Companies have started to address this issue of reclaiming lithium into a valuable secondary raw material. Recycling relies not only on the power of chemistry but also on the culture (that is, people's initiative to recycle). Here again, we have witnessed great progress over the past decade, with most families routinely separating recycling from garbage, reusing shopping bags, and so on. The change in attitudes towards sustainability in society is promising.

Concomitantly, LCA is also coming into play with numerous ongoing studies. Caution has to be exercised, however, as they are

quite complex owing to numerous uncertainties and assumptions. Staunch believers in LCA should recall the Heisenberg uncertainty principle: the more accurate the calculations are, the tinier the target is, the larger the number of parameters is, the more non-assessed time and energy spent to reach the results will be. This should motivate the LCA community to promote the assets of approximation and error bars, and to convince other communities of LCA's accuracy and utility. It would not be a surprise if some philosophers soon address LCA and show the basic contradiction inherent to tracking the footprint of everything. While LCA is an absolute necessity, it should remain an estimate if it is to be the fast and efficient decision lever we all need. At present, impact calculations resulting from LCA are geographically, politically and highly time-dependent. Once comparative LCA of different processes is achieved, one should also question the weight their comparative costs will have in the final industrial choice. Depending on this weight, LCA could appear as a decision tool for the price to be paid to pollute. Here, a new tripartite decision equilibrium will have to hatch between politicians, scientists and economists<sup>110</sup>.

In summary, our society is more aware than ever before of sustainability issues and less willing to compromise on upcoming technologies. Scientists are on the same wavelength and are already engaged in a paradigm shift judging by the way that research directions have evolved — sustainability is now being considered as an extra dimension in addition to structure, composition and morphology when designing new materials. Moreover, the chemistry of recycling will become more important than ever and let's hope that chemists can successively give a second life to wastes. Undoubtedly, sustainable batteries can be made and bring major advances in the protection of our environment, provided we realize such an effort is only worthwhile if we use  $\text{CO}_2$ -free electricity. This is just a matter of time. The real question is: can such electrochemical storage at large scale and low cost be made in the allocated time to assist in the use of an extra 130,000 TWh  $\text{yr}^{-1}$  (14 TW) of  $\text{CO}_2$ -free energy by the year 2050?

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## Competing financial interests

The authors declare no competing financial interests.