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A perspective on organic electrode materials and technologies for next generation batteries

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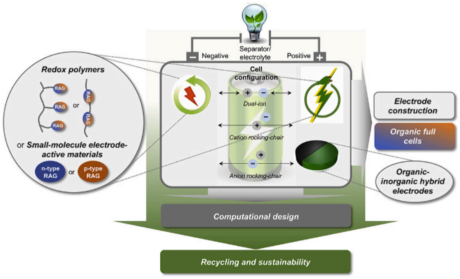
HIGHLIGHTS

- Organic solid electrode materials are promising for new generation batteries.
- A large variety of small molecule and polymeric organic electrode materials exist.
- Modelling and characterization techniques provide insight into charge and discharge.
- Several examples for all-organic battery cells have been reported to date.
- Environmental impact and sustainability of organic electrode materials are beneficial.

GRAPHICAL ABSTRACT



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ABSTRACT

In this perspective article, we review some of the most recent advances in the emerging field of organic materials as the electroactive component in solid electrodes for batteries. These comprise, but are not limited to, organometallic salts, small molecular systems, redox-active macromolecules, as well as hybrid formulations with inorganic electrode constituents. The materials are first scrutinized in terms of their general electrochemical performance and most apparent challenges, while an outlook is then made into how to best utilize them in battery electrodes and in all-organic cells. An insight into the fundamental structural-dynamic properties of these compounds, not least explored through a range of modelling and characterization techniques, is also given to complement the experimental advances. The major advantages of these materials as compared to competing

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technologies are most likely their potentially low environmental impact and general sustainability, which forms the context of this summary of the research field and corresponding technology area.

1. Introduction

Organic material-based rechargeable batteries have great potential for a new generation of greener and sustainable energy storage solutions [1,2]. They possess a lower environmental footprint and toxicity relative to conventional inorganic metal oxides, are composed of abundant elements (i.e. C, H, O, N, and S) and can be produced through more eco-friendly procedures as well as require lower energy-consuming recycling processes. Beyond the high structural designability, two electrochemical storage mechanisms can be used: “n-type” electrode reactions that involve an ionic compensation with cation release upon oxidation and “p-type” electrode reactions that involve an anion uptake [3]. In the past decade, much research has gone into the development of organic batteries. This includes in particular the synthesis and investigation of organic electroactive materials [2,4–11], but also other aspects, such as electrode construction [12,13], realization of all-organic batteries [3,14] and sustainability considerations [1]. This Focus Review aims at providing an overview of the state-of-the-art of different aspects of organic batteries, highlighting selected studies rather than compiling all examples from the literature, and giving future perspectives. The aspects of organic batteries covered herein are shown in Fig. 1.

Concerning the composition of the organic electroactive materials, both redox polymers and small-molecule electroactive compounds will be covered in Sections 2 and 3, respectively. Different strategies have been identified to obtain high electrochemical performance of organic compounds in batteries [15–18]. The most important parameters are their charge/discharge potential, specific capacity, rate capability and cycling stability [7,15]. The charge/discharge potential is determined by the nature of the redox-active groups, and the specific capacity by its molecular weight and the number of electrons participating in the redox process. The rate capability depends on the rate of the electron- and ion-transfer processes in and out of the electrode [19]. It is influenced by the electron-transfer rate constant of the redox-active group and the morphology of the composite electrode, usually consisting of a mixture of organic material, conductive carbon and polymeric binder. The cycling stability is often limited by either dissolution of the active material into the (usually liquid) battery electrolyte or by decomposition processes. Different strategies have been developed to overcome this issue in non-aqueous electrolyte media, such as the incorporation of the redox-active units into a polymer backbone [5,20–23], salification (i.e. introducing conjugated anionic groups on the redox-active moiety), and hybridization with insoluble materials, such as different forms of carbon [15]. Most of the reported organic electrode materials have been tested in half cells (e.g., against Li or Na as negative electrode), but an increasing number of studies report on all-organic batteries, which will be discussed as part of Section 6 [3,14].

With increasing available computer power and software quality, computational modelling is gaining relevance in the field of organic batteries. This includes predicting properties, such as the redox chemistry of organic electroactive materials, but also gaining mechanistic understanding of charge/discharge processes on the molecular and microscopic level. A variety of computational methods are available, which will be reviewed and exemplified in Section 4.

As mentioned above, the fabrication of battery electrodes usually involves mixing the organic electroactive materials with other components. Of major importance is the interfacing with conductive additives, given the insulating nature of most organic materials. Since so far these are added in large amounts, ranging typically between 30 and 70 wt%, the specific capacity and energy density of the organic composite electrode are significantly reduced, which may have been the main reason hampering commercialization of organic batteries so far. Hence, electrode construction is an issue of high importance to organic batteries and will be covered in Section 5.

Apart from their use as sole electroactive material, organic redox-active compounds are also attractive candidates for organic-inorganic hybrid electrodes. These have the potential to merge the best of both worlds (technologies), such as a limited functionality and power performance of inorganics and low energy densities of organic electroactive materials, and to unlock high-power and high-energy performances. Several examples have been reported in literature, which will be discussed in Section 7.

Lastly, since one of the main motivations of developing organic electroactive materials is for greater sustainability, it is important to highlight the need to develop truly sustainable electrode materials for future electrochemical energy storage [24] and how organic batteries can play a major role. This includes a rapid overview of the current

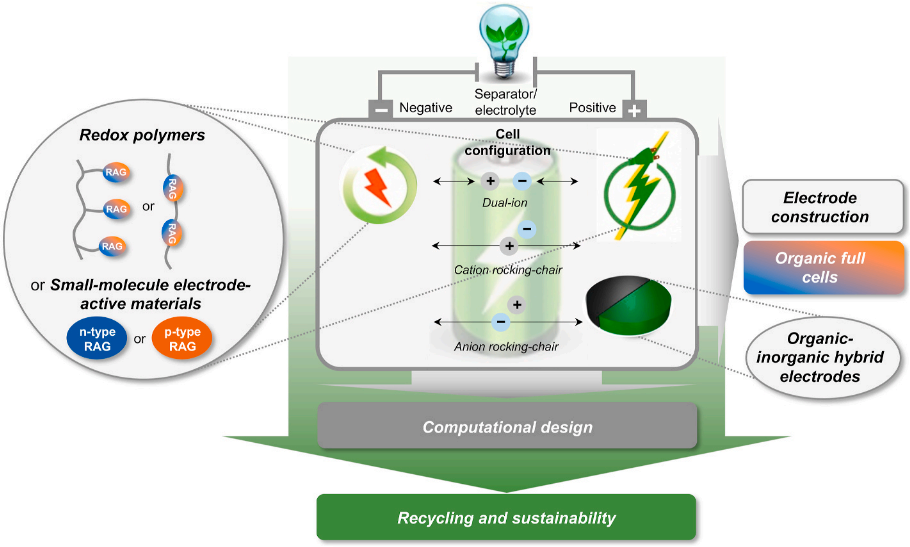

Fig. 1. Organic batteries – components, research areas and issues to be considered with respective Sections covered in this Focus Review.

situation with the materials' origin, manufacturing processes and the recycling of used cells. These recycling and sustainability factors will be discussed in Section 8.

2. Redox polymers as electroactive materials

One prominent strategy to improve the cycling stability and – in some cases – the rate capability of organic electrode materials is to incorporate the redox-active molecular units into a polymeric structure. This can lead to an insolubility of the resulting polymers in the commonly used liquid battery electrolytes, while the polymers can remain swellable to allow for good mass/ion transport. Such polymers are called redox polymers and defined as polymers containing groups that can be reversibly reduced or oxidized [25]. We herein provide an overview of design principles and the performance of selected examples of organic redox polymers as electroactive battery materials out of many reported in the literature [5,8,20,21,23,26,27].

There are two main structural types, A and B, of redox polymers (Fig. 2a) [25]: In Type A, the redox-active groups are attached as side-groups to an aliphatic or conjugated polymer backbone, and in Type B they are incorporated into the conjugated or non-conjugated polymer backbone (Fig. 2a).

Alike other organic battery materials, redox polymers can also be classified based on their preferential redox reaction: p-type polymers are more easily oxidized ($p \rightarrow p^{\bullet +}$) than reduced, n-type polymers more easily reduced ($n \rightarrow n^{\bullet -}$) than oxidized (Fig. 2b), and bipolar polymers can undergo both types of redox reactions. For the choice of redox-active group, a highly reversible redox chemistry is desirable in combination with a low molecular mass and attractive redox potential for a battery application. Herein, only a selection of redox-active groups will be discussed, namely stable (nitroxide) radicals, heteroaromatics, quinones and diimides, but other types of redox-active groups have also been reported [28]. Furthermore, the electrochemically inactive part of the polymer backbone should add the least possible amount of additional molecular weight to obtain a high specific capacity for the resulting redox polymer. The theoretical specific capacity of a redox polymer can be calculated according to the following equation (where n is the number of electrons transferred per subunit, <I>F</I> is the Faraday constant and <I>M</I>(polymer subunit) is the molar mass of a polymer subunit):

$$Q_{\text{spec., redox polymer}} = \frac{n \times F}{M(\text{polymer subunit})} = \frac{n \times 26801}{M(\text{polymer subunit})} \text{mAh g}^{-1}$$

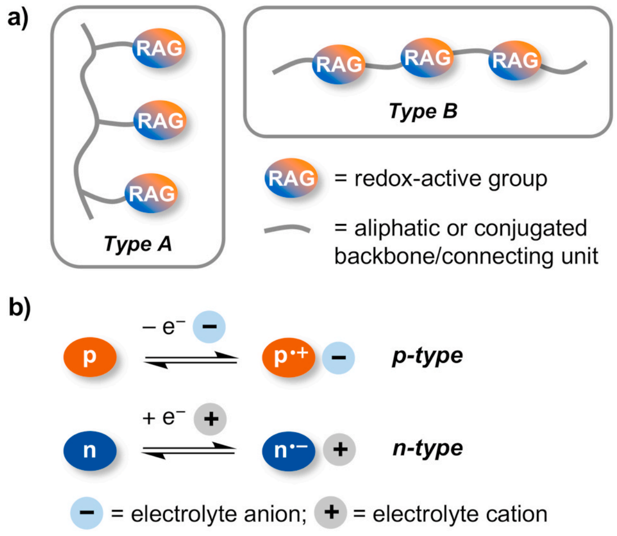

Fig. 2. (a) Design types A and B for redox-active polymers with the redox-active groups (RAGs) as pendant side units (Type A) or incorporated into the polymer chain (Type B); (b) redox reaction of p- or n-type organic compounds and charge balancing by electrolyte ions. Reprinted with permission from ref. [5].

To fabricate the final electrode for the battery cell, the redox polymer is usually mixed with a conductive carbon additive to obtain sufficient electronic conductivity [23] and with a binder for better mechanical and adhesive properties of the resulting film. Organic redox polymers are in most cases first tested as cathode-active materials in half cells against lithium as counter electrode to evaluate their electrochemical performance [10,23,29]. For a full-cell, two redox polymer-based electrodes can be combined to furnish an all-organic battery (see also Section 6) [3,14] or intercalation materials as well as other metals than lithium; for example, sodium [7,13,16,30,31] and multivalent metals (magnesium, zinc and aluminum) [32], can be used as anodes. In the following, the most prominent types of redox polymers will briefly be discussed, which are conducting polymers (mostly for historic reasons), radical polymers, carbonyl-based polymers and polymers incorporating heteroaromatics as redox-active groups.

2.1. *Conducting polymers*

Already in the 1980s, shortly after their discovery, conducting polymers such as poly(acetylene) [33], polythiophene, polypyrrole and polyaniline were used as battery electrode materials [34]. Their application as such has been reviewed several times [20,26,35]. Most conducting polymers are of p-type, including some ambipolar materials. They are a typical example of Type B redox polymers (Fig. 2a), in which, however, defined and localized redox-active groups are absent. As a consequence, sloping charge/discharge curves were often observed for electrodes based on conducting polymers, since their redox potential changes with increasing doping level of the polymer, together with low achievable doping levels [23]. Advanced methods have been used more recently to improve the performance of conducting polymers, such as nanostructuring [23,36–38]. However, non-conjugated redox polymers with separated redox centers, as will be discussed in the following subsections, have generally proven to be better suited as battery electrode materials.

2.2. *Radical polymers*

One of the best performing and most investigated redox polymer classes to date for charge storage are radical polymers. These Type A polymers (Fig. 2a), usually polymers with aliphatic backbones and side-groups functionalized with stable radicals, were first introduced in the early 2000s [39–45]. The most prominent radicals are nitroxides, such as TEMPO ((2,2,6,6-tetramethylpiperidin-1-yl)oxyl, **1** in Fig. 3a). These are typically used as p-type cathode materials and undergo a one-electron oxidation to oxoammonium cations (see Fig. 3a). Ambipolar [46] or n-type [40,46,47] nitroxide radicals have also been reported. **1** and redox polymers based on this or other p-type nitroxide radicals possess an attractive discharge potential of around 3.6 V <i>vs</i>. Li/Li^{+}. **PTMA**, a TEMPO-radical functionalized polymethacrylate (Fig. 3b), is the most investigated radical polymer to date [48]. It possesses a theoretical specific capacity of 111 mAh g^{-1}, allows for fast C-rates up to 100C [49] and enables high cycling stabilities. **PTMA**-based electrodes have been used even in thin-film batteries [40,50] and pouch cells [51]. An example of an n-type radical polymer is **PGVS** (Fig. 3b) with the galvinoxyl radical **2** (Fig. 3a) as side group [52].

2.3. *Heteroaromatic redox polymers*

Another type of redox-active group with reversible redox chemistry and a wide range of discharge potentials are heteroaromatics. Most are of p-type [2,7,10,11,23], and so are the examples highlighted herein.

Phenothiazine (**3** in Fig. 3a) has a similar redox potential of 3.6 V

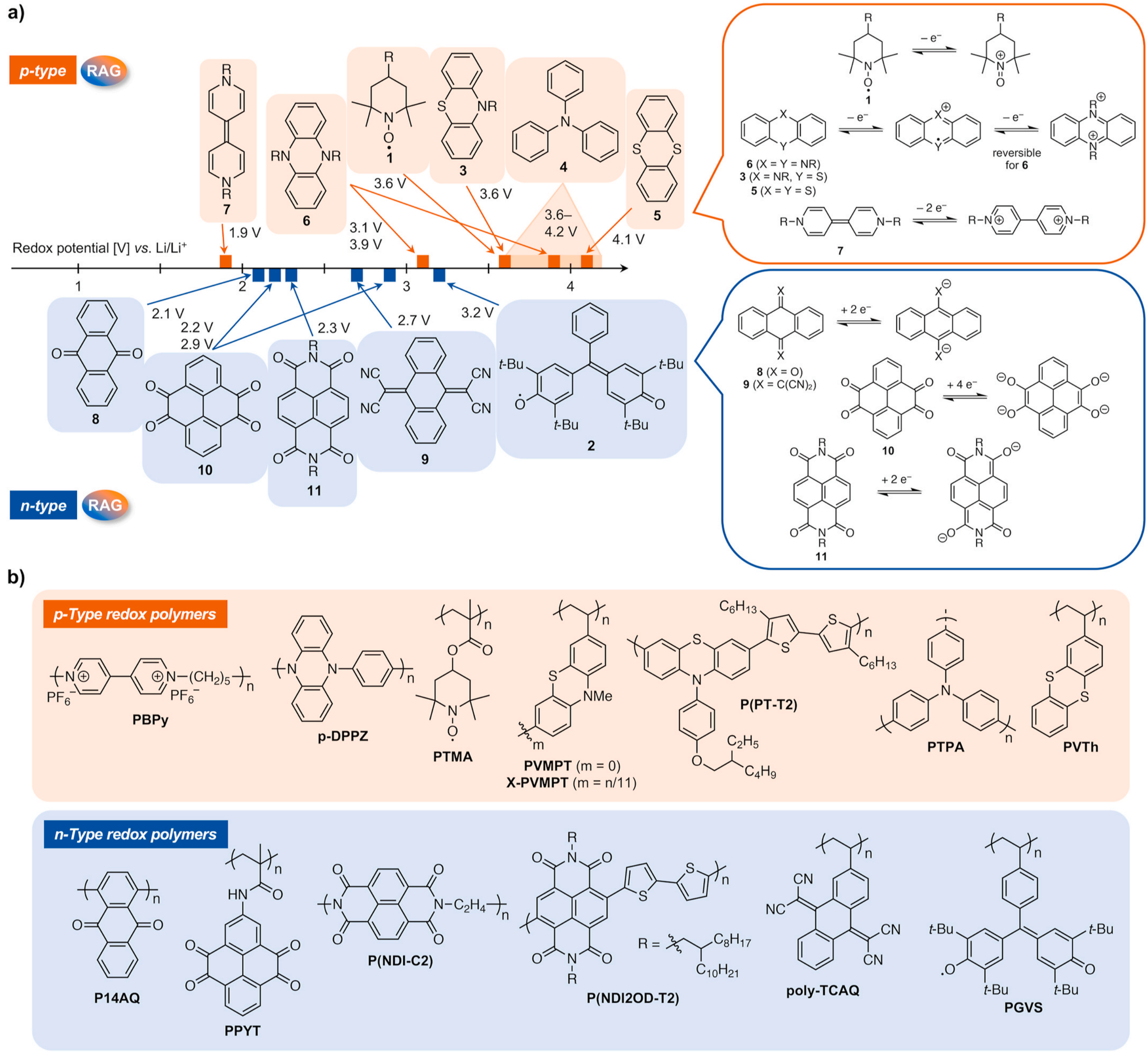

Fig. 3. a) Redox potentials of selected p- and n-type redox-active subgroups incorporated into redox polymers with the corresponding redox mechanisms of selected groups; b) selected examples of redox polymers based on the redox-active groups shown in a).

<i>vs</i>. Li/Li$^+$ as the TEMPO radical mentioned above. To date, ten studies using phenothiazine-based redox polymers as electrode materials have been published. The polymers were of both Type A [53–58] or Type B [59–62]. **PVMPT** (Fig. 3b) convinced with an outstanding cycling stability due to π-interactions between neighboring oxidized (and neutral) phenothiazine units [54,55]. This enabled fast C-rates of up to 100C, using thin electrodes, and long-term cycling at a 10C rate with only 7% capacity loss during 10,000 cycles [54]. However, the available specific capacity of **PVMPT** was limited to about 50 mAh g^{-1}, corresponding to half of the theoretical value. This was rationalized through mechanistic studies indicating strong π-interactions between the phenothiazine groups [55]. Diminishing these interactions through cross-linking in **X-PVMPT** (Fig. 3b) led to the full specific capacity of 112 mAh g^{-1} being accessible [56]. The phenoxazine-based congener **PVMPO** and its cross-linked derivative **X-PVMPO** also demonstrated high rate performances [63]. By using a conjugated backbone in **P (PT-T2)** (Fig. 3b), the rate capability and cycling stability in phenothiazine-based polymers could be increased further [61]. Cycling at 100C rate for 30,000 cycles was possible with a capacity retention of 97%. The large amount of inactive side groups and co-monomer, however, reduced the specific capacity of **P(PT-T2)** to 30 mAh g^{-1}. Nevertheless, this example conceptually shows that intrinsic conduction through the polymer backbone in the doped state enables high charge/discharge rates.

Triarylamines (**4** in Fig. 3a) have the advantage of making higher discharge potentials of 3.6–4.2 V <i>vs</i>. Li/Li$^+$ accessible. Poly (triphenylamine) (**PTPA**, Fig. 3b) is an example of such a redox polymer [64]. It is a Type B polymer with significant semiconductive nature and poor separation of redox-active centers, causing sloping charge/discharge curves. However, an impressive rate capability was demonstrated, where a specific capacity of 91 mAh g^{-1} was available at 20C rate. Hyperbranching the polymer backbone even further improved the rate capability [65].

Further increasing the discharge potential to 4.1 V <i>vs</i>. Li/Li$^+$ was demonstrated with thianthrene (**5** in Fig. 3a) as redox-active group. Two examples of Type A-polymers were reported with a poly (norbornene) [66] or poly(vinylene) (**PVTh** in Fig. 3b) [67] backbone. **PVTh** was used as cathode-active polymer in an all-organic battery with **poly-TCAQ** (Fig. 3b) on the anode side, featuring a cell potential of 1.35 V [67].

In all redox polymers discussed so far, only one oxidation process of the redox-active, p-type group could be used, limiting their specific capacities. Dihydrophenazines (**6** in Fig. 3a), on the other hand, show two reversible oxidation events. Their use in battery electrodes has been demonstrated for polymers based on **6**, for instance **<i>p</i>-DPPZ** (Fig. 3b) and related Type B-redox polymers [68–70]. Specific capacities around 200 mAh g^{-1} with good rate performance were reported [70]. The electrodes featured two flat plateau potentials at 3.1 and 3.9 V <i>vs</i>. Li/Li$^+$.

As a final example for a p-type unit incorporated into redox polymers, bispyridinylidene **7** (Fig. 3a) shall be mentioned. Its advantage is a low redox potential of only 1.9 V <i>vs</i>. Li/Li$^+$, allowing for the resulting redox polymer to be used as negative (anode) electrode material in full-organic cells against a high-voltage p-type cathode material. Such a cell can operate in anion-rocking chair mode, which brings the advantage of the cell being completely metal-free, among others, as will be discussed in section 7 [3]. As example of a **7**-based redox polymer, **PBPy** (containing **7** in its oxidized state) with a redox potential of 1.9 V <i>vs</i>. Li/Li$^+$ was reported (Fig. 3b) [71]. Its utility was demonstrated by testing this polymer in an all-organic anion-rocking chair battery with poly(<I>N</I>-vinylcarbazole) as cathode-active material [71]. A cell voltage of 1.8 V resulted, and the cell delivered a specific capacity of 100 mAh g^{-1} at 1C-rate.

2.4. *Redox polymers containing carbonyl or bisimide moieties*

Redox polymers incorporating conjugated carbonyl groups (quinone derivatives) or bisimides can show highly reversible redox processes and high specific capacities [72–75]. These redox-active groups are of n-type, as shown in Fig. 3a. Prominent examples are anthraquinone (**8**) or derivatives thereof (**9**), pyrenetetraone (**10**) and naphthalene diimides (NDI, **11**, Fig. 3a).

One of the best-performing redox polymers of Type B with anthraquinone as redox-active group is **P14AQ** (Fig. 3b) [76]. Due to the atom-efficient incorporation of the anthraquinone groups into a polymeric structure, a high specific capacity of 248 mAh g^{-1} was obtained, and **P14AQ** showed excellent cycling stability at 1C-rate with 98% capacity retention after 1000 cycles. The two-fold reversible reduction of the anthraquinone groups takes place at a potential of 2.1 V <i>vs</i>. Li/Li$^+$, with flat plateaus in the charge/discharge profiles. By transformation of the groups into dicyanomethylene groups in **poly-(TCAQ)** (Fig. 3b), the redox potential could be raised to 2.7 V <i>vs</i>. Li/Li$^+$ [77]. The increased molecular mass of the redox-active group and the added weight of the polymer backbone decreased the specific capacity (compared to **P14AQ**) to 156 mAh g^{-1}, which, however, remains a respectable value. A good cycling stability was obtained with 88% capacity retention after 500 cycles at 1C-rate, however, at a low active material loading of 10 wt% in the composite electrode. A poly(methacrylate) carrying the same redox-active group was employed as negative electrode (anode)-active material in an all-organic battery against **PVTh**, as previously mentioned [67].

Using pyrenetetraone (**10**) instead of anthraquinone (**8**) increases the specific capacity of the redox-active group. However, incorporation into a polymer has to be feasible. **PPYT** with pyrenetetraone side groups (Fig. 3b) allowed for a four-electron redox process at potentials of 2.8 and 2.2 V <i>vs</i>. Li/Li$^+$, but due to the added weight of the methacrylate backbone with 231 mAh g^{-1}, the specific capacity was lower than in **P14AQ** [78]. **PPYT** demonstrated good cycling stability and retained 83% of its specific capacity after 500 cycles at 1C-rate.

The NDI group (**11** in Fig. 3a) is widely used in organic electronics for its n-type character and reversible redox chemistry. One of the most atom-efficient ways to incorporate NDI into a polymer structure are poly (bisimides), Type B-polymers such as **P(NDI-C2)** (Fig. 3b) [79] among many other examples. Furthermore, synthesis through polycondensation is simple and transition-metal free. Each NDI unit can reversibly take up two electrons, in spite of the four-electron reduction, which is possible in theory. This furnishes a specific capacity of 180 mAh g^{-1} for **P(NDI-C2)** [79]. Both electrons are transferred at almost the same potential of 2.3 V <i>vs</i>. Li/Li$^+$, resulting in a flat charge/discharge profile. The rate capability of NDI-containing polymers can be improved by using a conjugated polymer backbone, as impressively demonstrated for **P(NDI2OD-T2)** (Fig. 3b) [80]. This polymer is well-established as an n-type semiconductor in organic electronics. The two-electron reduction takes place at a potential of 2.5 V <i>vs</i>. Li/Li$^+$, and an excellent cycling stability and rate capability were reported with 96% of the initial specific capacity of 54 mAh g^{-1} retained after 3000 cycles at 10C-rate. **P(NDI2OD-T2)** also performed well as cathode-material in a Mg-organic battery, demonstrating reversible MgCl$^+$-insertion for charge balancing [81].

3. Small molecular organic electroactive materials

Molecular or crystalline organic electroactive materials (OEMs) possess most of the desired qualities of organic materials, especially a high design flexibility both at the molecular and structural level, and a well-defined redox signature. Generally, their gravimetric and volumetric capacities are higher than for redox-active polymers, since they do not contain any electrochemically inert polymeric backbone, and their molecular structures can allow for efficient packing. But they are also usually plagued by the intrinsic disadvantages of OEMs: a notably

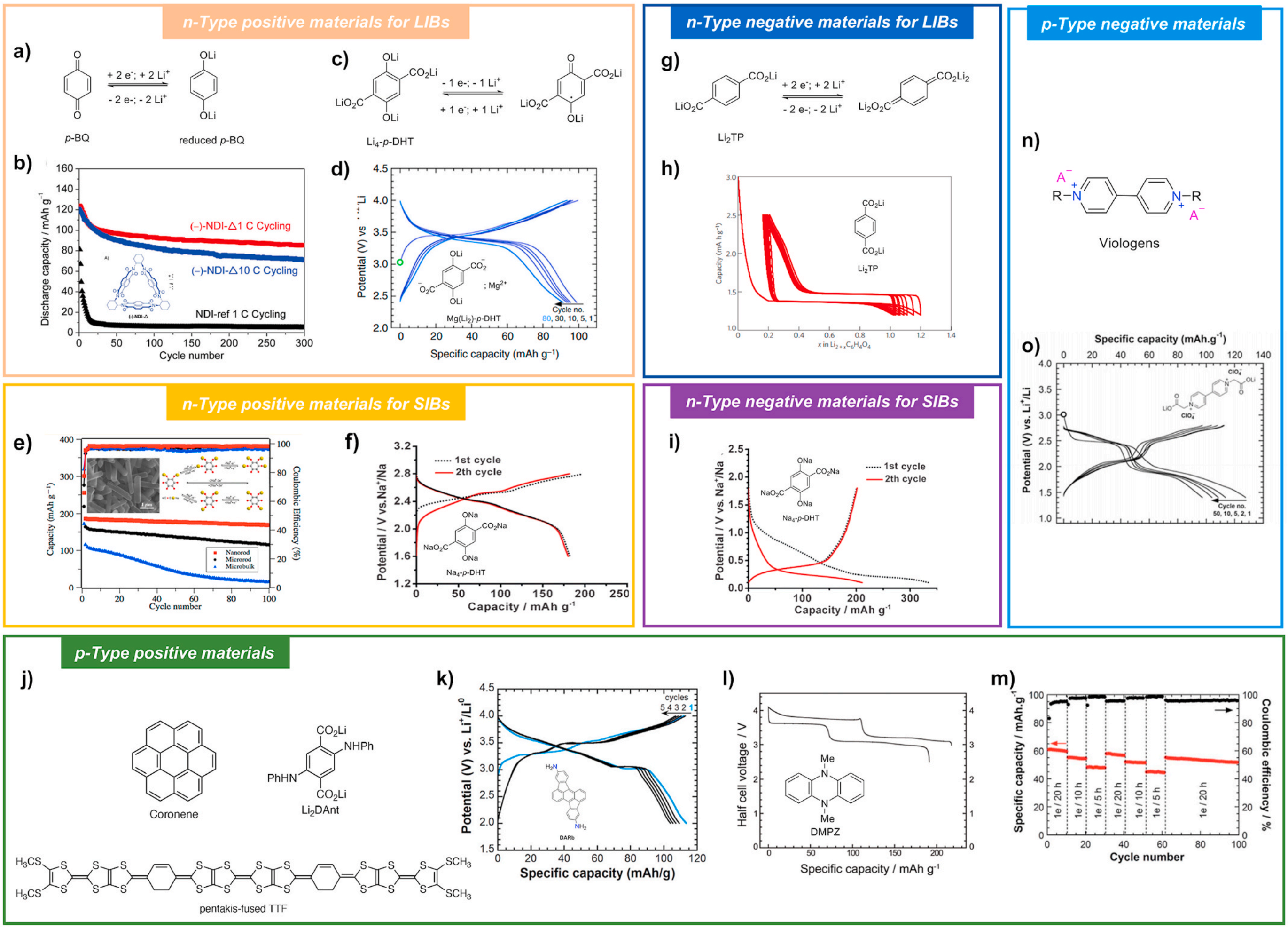

Fig. 4. n- and p-Type molecular OEMs used as electrodes together with their representative electrochemical behavior; a) Reversible electrochemical two-electron reaction of <i>p</i>-BQ, starting with its reduction; b) Chemical structure of (−)-NDI Δ and its cycling performance compared to the monomer showing better rechargeability against Li; reprinted with permission from Refs. [94], © 2015 WILEY-VCH; c) Reversible electrochemical one-electron reaction of Li$_4$-<i>p</i>-DHT, starting with its oxidation; d) Potential <i>vs</i>. specific capacity curve of a Li half-cell using Mg(Li$_2$)-<i>p</i>-DHT [99]; e) Cycling stability of Na$_2$C$_6$O$_6$ nanorods against Na; reprinted with permission from Ref. [100]; f) Initial discharge/charge profiles of Na$_4$-<i>p</i>-DHT in Na half-cells [101]; g) Reversible electrochemical two-electron reaction of Li$_2$TP, starting with its reduction; h) Potential-composition profile for Li$_2$TP, galvanostatically cycled at a rate of 1 Li$^+$/10 h in a Li-half cell [102]; i) Initial cycles of Na$_4$-<i>p</i>-DHT as electrode against Na; reprinted with permission from Refs. [101], © 2014 WILEY-VCH; j) Chemical structures of coronene, pentakis-fused TTF and Li$_2$DAnT; k) Electrochemical performance of DARb in a Li half-cell; reprinted with permission from Ref. [125]; l) DMPZ initial charge and discharge in a Li half-cell. Reproduced from Ref. [126] with permission from The Royal Society of Chemistry; m) Discharge capacity retention curves with coulombic efficiency of a Li half-cell using carbon-free Li$_2$DAnT powder, reproduced from Ref. [127] with permission from The Royal Society of Chemistry; n) General chemical structure of viologens; o) Galvanostatic cycling curves of (Li)$_2$diacetate-V(ClO$_4$)$_2$ measured in a Li half-cell; reprinted with permission from Refs. [128], © 2019 WILEY-VCH.

high solubility in most electrolytes, leading to poor cyclability.

If OEMs are to be used, high energy densities and long cycle life materials are mandatory. Fortunately, dealing with organic matter offers incomparable opportunities for molecular engineering and improvement of properties. Since 1969 and the assessment of dichloroisocyanuric acid in a Li primary battery [82], different electrochemically active structures have been disclosed with redox potentials ranging from nearly 0 to 4 V <i>vs.</i> Li^+/Li, leading to a vast range of positive or negative electrode materials, exploiting n-type or p-type electrochemical mechanisms (see Fig. 2), in metal, metal-ion (M-ion), dual-ion or even anion-ion batteries [2,6–8,15,18,24,31,32,73,74,83–85].

3.1. n-Type small organic electroactive materials

n-Type molecular OEMs have been extensively studied [6,7,14,85,86]. This interest is nested in their electrochemical reactions (see Figs. 2 and 3a for examples), which resemble those of classic inorganic insertion materials used in M-ion batteries such as LIBs. Among small OEMs, the n-type electrochemical activity is assured by different redox-active organic functions, carbonyls being the most represented (C═O in quinones, ketones, carboxylates, anhydrides, and imides). Next are nitrogen derivatives, such as –CN (conjugated nitriles) and C═N (Schiff's base and pteridine derivatives), others are the recently disclosed N═N (azo) and the older –S–S– (organodisulfides) moieties. All of these organic electroactive functions (oxidized state) are embedded into a structural environment, where the neighboring groups and/or the inherent structure of the molecule (e.g. isomerism, aromaticity) influences their final electrochemical properties. A facile cation exchange is also possible, and opportunities to propose materials for post-LIBs technologies will rise. We can already witness their spreading use with other battery chemistries (Na, K, Mg, Ca, Zn or Al batteries) as positive or negative materials [31,32].

Most of the n-type OEMs disclosed to date are being prepared and handled in their oxidized state, do not initially contain lithium (or metal), and the first electrochemical reaction needs to be a reduction, exemplified in Fig. 4a through the two-electron redox mechanism of <i>p</i>-benzoquinone (<i>p</i>-BQ). Therefore, most of them have been evaluated against lithium (half-cells), or must be used in combination with a metal-containing negative electrode material, or need to be pre-reduced/lithiated before assembly in the final electrochemical device. The latter option would be a difficult step for scale-up [24,87]. Ubiquitous in nature, quinone- and carbonyl-containing molecules have been studied extensively, and <i>p</i>-BQ is thereby a representative compound, characterized by two discharge plateaus averaging to a discharge potential of 2.7 V <i>vs.</i> Li^+/Li with a theoretical specific capacity of 429 mAh g^{-1} (considering the molar mass of the lithiated Li_2-BQ phase), making it a promising positive electrode material [88]. But <i>p</i>-BQ is highly soluble in liquid electrolytes, typically showing poor cyclability. Modifications through molecular design have been shown to lead to improvements. For example, addition of electron-withdrawing groups raises the potential, and electron-donating groups result in lower values while also affecting the solubility and thus the cyclability. For example, 2,5-perfluoroalkyl-<i>p</i>-BQs (3.0–3.1 V <i>vs.</i> Li^+/Li) are less soluble [89], like 2,5-dialkyl-<i>p</i>-BQ [90], but the potential is lowered for the latter. However, addition of such electro-inactive moieties negatively impacts the specific capacity, and these additions should consequently be kept as low as possible, or should be electrochemically active as in the case of some oligomers. For instance, dimerization of <i>p</i>-BQ leads to 2,2′-bis-<i>p</i>-benzoquinone (BBQ) with a discharge voltage of 2.9 V <i>vs</i> Li^+/Li, where 52% of its initial capacity was preserved after 20 cycles [91]. Recent electrode composite optimization (50% of CMK-3 carbon) allowed for 152 mAh g^{-1} of specific capacity remaining after 3000 cycles [92]; improvements compared to <i>p</i>-BQ, but still not fully satisfactory. Grafting four phthalimides to <i>p</i>-BQ leads to 2,3,5,6-tetraphtalimido-1,4-benzoquinone as a nice way to multiply redox centers on the same structure, which allowed for 91.4% retention of capacity (204 mAh g^{-1}) after 100 cycles and improved rate capability owing to an extension of the π-conjugation [93]. Designing the molecular skeleton can be of great value, as shown with the rigid triangle NDI (Fig. 4b), where the elaborated structure presented an improved performance in terms of rate capability, cyclability and coulombic efficiency due to a unique spatial arrangement [94]. Sometimes, however, oligomerization is insufficient. Calix[4]quinone (C4Q) as cyclic tetramer of <i>p</i>-BQ was used with a gel polymer electrolyte to increase capacity retention [95]. Following this example, the development of solid (or quasi-solid)-state electrolytes (SSEs) could present an opportunity for small OEMs, like it has been shown with the efficient two electron-cycling of tetramethoxy-<i>p</i>-benzoquinone within the commercial Lithium Metal Polymer (LMP) technology framework [93].

Other small OEMs can possess high capacities, with one of the highest values demonstrated for dilithium rhodizonate ($Li_2C_6O_6$, 580 mAh g^{-1}), obtained from non-edible renewable resources [96]. However, its cyclability was limited by crystal structure changes during lithium extraction [97]. This value was recently surpassed by the elusive C_6O_6 oxocarbon with a theoretical specific capacity of 902 mAh g^{-1}, however, the electrochemical response was in part due to a pseudo-capacitive contribution [98].

As stated earlier, usually n-type materials are in their oxidized state, but to assemble a classic "rocking chair" type organic LIB, OEMs are needed also in their lithiated reduced state. Dilithium (2,5-dilithium-oxy)-terephthalate (Li_4-<i>p</i>-DHT, Fig. 4c), a carboxylate-substituted <i>p</i>-hydroquinone salt, was prepared in such a discharged (lithiated) state [103,104]. It is well-known that the introduction of permanent ionic charges by addition of, for example, carboxylates or sulfonates decreases the solubility [1]. However, carboxylates are donor-inductive groups and thus decrease the redox potential of the material: Li_4-<i>p</i>-DHT had an average working potential of 2.55 V <i>vs</i> Li^+/Li (Fig. 4g); low for practical use. If small OEMs are to be competitive with inorganic electrode materials ($LiFePO_4$ for instance), metal-containing cathodes must show higher operating voltage. Simply replacing the Li^+ counter cation of the carboxylate groups with a high-electronegativity cation such as Mg^{2+} mitigated the electron-donating effect and increased the redox potential of (2,5-dilithium-oxy)-terephthalate ($Mg(Li_2)$-<i>p</i>-DHT, Fig. 4d) up to 3.4 V <i>vs</i> Li^+/Li, offering a lithiated organic cathode material nearly able to compete with inorganics [99].

Switching from lithium to the sodium-ion battery technology or even other metals is readily feasible with small OEMs. Most of them, being in their oxidized state when incorporated into the battery, are theoretically useable with any M-ion battery technology, and exchange of the cations is also simple. Therefore, different sodiated materials have been assessed against metallic sodium for positive electrode utilization [4,7,31]. In Na-based cells, small OEMs encounter almost the same limitations as in their Li counterparts, but it is reported that flexibility of organic materials leads to a better accommodation of larger Na^+ cations [31]. Consequently, several organic compounds tested <i>vs</i> Li were transferred to Na-based cells. For instance C4Q was also studied as positive electrode <i>vs</i> Na after encapsulation in CMK-3 to lower its solubility [105]. Disodium rhodizonate ($Na_2C_6O_6$) was evaluated, and, interestingly, it seemed to perform better than its lithiated counterpart which was plagued by structural modifications upon cycling. The sodiated material retained 90% of capacity after 100 cycles (Fig. 4e) [100,106].

As in the case of inorganic LIBs, the rise of SSEs will certainly be beneficial for small OEMs, since dissolution in the liquid electrolyte is one of the major obstacles [31]. Recently, $Na_2C_6O_6$ was used together with a sulfide electrolyte in a sodium all-solid-state battery [107]. Considering "rocking chair" type SIBs, Na_4-<i>p</i>-DHT (Fig. 4f), the sodiated version of Li_4-<i>p</i>-DHT, was used as positive electrode and characterized by a capacity higher than 180 mAh g^{-1} [108].

Furthermore, oxocarbon potassium salts have for example been reported as electrode materials for potassium ion batteries (KIBs) [109]. Noticeably, $K_2C_6O_6$ (di-potassium rhodizonate) was investigated as positive electrode material in an all-organic KIB [109]. In Mg-ion batteries, among others, 2,5-dimethoxy-<i>p</i>-benzoquinone (DMBQ) [110] and $Na_2C_6O_6$[111] were evaluated. DMBQ showed a specific discharge capacity of 260 mAh g^{-1} against a magnesium quasi-reference electrode, but with limited cyclability [110]. Nanocrystalline $Na_2C_6O_6$ achieved a high rate performance of 200 mAh g^{-1} at 5C [111]. A series of quinones were evaluated as electrode materials in aqueous zinc batteries, C4Q showing the most interesting cyclability with 1000 cycles [112]. 9,10-Phenanthraquinone was tested in aluminum cells, and its trimerized triangular version exhibited a reversible capacity of 110 mAh g^{-1} over 5000 cycles [113]. These few examples represent only the potentiality of using small OEMs as electrodes for M-ions and multivalent batteries that surely will be developed further in the future.

Low redox potential n-type small OEMs can be used as negative electrodes materials. Concerning their energy density, a direct comparison with graphite is to their disadvantage, but they could offer an alternative to LTO and bring post Li-ion anode materials, improved safety and the possibility to use aluminum current collectors instead of copper. Concerning their practical use, major issues are usually low rate capabilities and low initial coulombic efficiencies. The pioneering report on small OEMs as negative electrodes materials concerned the reversible electrochemical reaction of dilithium terephthalate against lithium (Li_2TP, Fig. 4g) [102]. This material, potentially obtained via recycling of terephthalate polymers, was able to be discharged at 0.8 V <i>vs</i> Li^+/Li (Fig. 4h). To further lower the potential, it is possible to add electron-donating groups (the observed potential of dilitihum 2,5-dimethyl terephthalate was 0.65 V <i>vs</i> Li^+/Li [114]) or modify the π-conjugated system connected to the carboxylate groups. A biphenyl in dilithium 4,4′-biphenyl dicarboxylate (Li_2BPDC) enabled a discharge potential of 0.7 V <i>vs</i> Li^+/Li together with a rate capability enhancement (86.4% of retention of capacity at 1C), attributed to a higher lithium diffusion rate and structural robustness [115]. Beyond the terephthalate family, azo compounds recently appeared as low potential material (~1.5 V <i>vs</i>. Li^+/Li) [116]. One representative member, the 4-(phenylazo) benzoic acid lithium salt (PBALs), can reversibly store two lithium cations at an average potential of 1.4 V <i>vs</i> Li^+/Li with good cycling stability and high rate capability.

One of the reported problems in the development of SIBs is the low efficiency of appropriate anode materials because of the large size and sluggish kinetics of Na ions. Small OEMs are therefore of special interest [117], and the terephthalate family has logically brought some anode materials. Switching lithium cations for sodium in Li_2TP leads to Na_2TP, capable of delivering 300 mAh g^{-1} at 1C rate and even 100 mAh g^{-1} at 10C rate [118,119]. The high rate capability was attributed to a longer diffusion path in the crystal when substituting Li for Na ions. Na_4-<i>p</i>-DHT, already used as positive material, was also used as negative electrode material since the carboxylate functionalities are reduced at around 0.4 V <i>vs</i> Na^+/Na (Fig. 4i) [108]. Azo-compounds were transferred to Na-based cells with potentials ~1.2 V <i>vs</i> Na^+/Na [120]. Specific to SIBs, the oligomeric Schiff bases act as negative electrodes in sodium-ion batteries and can reach voltages as low as 0.3 V <i>vs</i> Na^+/Na depending on the structure of the oligomer [121].

3.2. *p-Type small organic electroactive materials*

The implied electrochemical mechanism with anion uptake and release allows p-type materials to be evaluated against nearly any metal in dual-ion cells. But of utmost importance, in principle, they enable the development of fully “molecular” organic-ion batteries if the shuttling anion is metal free [71]. Electrochemically speaking, the anionic rocking-chair cell configuration has dual benefits: i) the anionic transference numbers are usually significantly higher than 0.5, ii) anions are poorly solvated in polar electrolyte media due to their low solvation Gibbs energy (in absolute value), which facilitates the ion transfer into the electroactive materials. For comparison, the working potentials are given against Li^+/Li.

Intrinsically p-type materials are characterized by higher formal potentials (>3.5 V <i>vs.</i> $Li^{+/}$Li) compared to n-type ones, making them natural positive materials [122]. High potential p-type small OEMs exploit carbon, oxygen, or nitrogen centers stabilized by aromatic structures and electron-donating functional groups, typically leading to OEMs with limited capacities due to significant dead mass (redox--inactive) and/or impartial reactions of the electrochemical groups [86]. For example, one of the highest average discharge potentials of 4.0 V <i>vs.</i> Li^+/Li is delivered by the polycyclic aromatic hydrocarbon coronene (Fig. 4j), but it unfortunately renders only 0.68 electrons per molecule upon charging, thereby limiting the capacity [123]. Therefore, it is important to develop organic structures where all the dedicated redox centers are exploited or with limited molecular weight. The well-known π-conjugated tetrathiafulvalene (TTF) has been incorporated in a pentakis-fused TTF (Fig. 4j), giving a material characterized by a potential above 3.5 V (3.56 V <i>vs.</i> Li^+/Li) and an attractive discharge capacity of 196 mAh g^{-1} owing to the use of almost two electrons per TTF unit [124]. Aromatic amines have been studied as p-type OEMs, also including small molecules like 5,12-diamino-rubicene (DARb, Fig. 4k) characterized by an average potential of 3.4 V <i>vs.</i> Li^+/Li and an initial capacity of 115 mAh g^{-1}, but with limited cyclability [125]. The same trend was observed when the two-electron reaction of 5,10-dihydro-5,10-dimethyl phenazine (DMPZ, Fig. 4l) was fully exploited. 50% of the initial capacity (191 mAh g^{-1}) were lost after ten cycles [126]. Dilithium 2,5-(dianilino)terephthalate (Li_2DAnT, Fig. 4j) can reversibly react at a relatively high operating potential (3.22 V <i>vs</i>. Li^+/Li) but with a modest specific capacity of 80 mAh g^{-1} due to a limitation to a one-electron process. Nevertheless, Li_2DAnT is worth mentioning, since it was cycled without any carbon additive for one hundred cycles (Fig. 4m) [127]. Its semi-conducting behavior was attributed to its overall structure, resembling a phenyl-capped aniline dimer.

Since p-type materials are naturally characterized by a high redox potential, finding those suitable to act in negative electrodes for the assembly of an anion-ion battery can be difficult. One specific backbone stands out: the 4,4-bipyridinium one, also known as viologen (Fig. 4n), because of its low redox potential [129]. However, only a few polymers were studied and exhibited limited performances [130]. Concerning molecular OEMs, very recently a non-polymeric double zwitterionic viologen derivative was designed: a 4,4′-bipyridinium capped with two acetate units to prevent electrolyte solubility. Li_2[diacetate-V]$(ClO_4)_2$ (Fig. 4o) was assessed against lithium, leading to nearly the theoretical specific capacity of 130 mAh g^{-1} (two-electron reaction) with pseudo-plateaus located at ~2.4 and ~1.9 V <i>vs</i>. Li^+/Li (Fig. 4o). This material gave an efficient p-type negative electrode material to be assembled in an all-organic anionic “rocking-chair” battery [128]. Nevertheless, molecular p-type materials characterized with lower reduction potential still have to be designed and prepared.

4. Computational insights and materials design

Due to the increasing strength of computer power and the improvement of relevant software, different types of modelling techniques are increasingly being applied in material science and the energy storage area. While being useful to understand and analyze chemical problems throughout several decades, they are now becoming more and more predictive tools, which can foresee and suggest materials with the desired properties. It could be argued that organic electroactive materials constitute a particularly useful category to explore computationally: the large possible structural variations and the complex interrelation with the electrochemical properties of these species together constitute a space, which is too challenging to map with other techniques. Three emerging trends can be seen. First, since the chemical problems for energy storage materials span several length and times scales – from ultrafast kinetics and atom-atom interactions to long-scale mass transport problems, macromolecular structure and electrode design – the connectivity between different methods constitutes a key for their simultaneous resolution. This is the core of <i>multi-scale modelling</i> [131]; interconnecting fine-structure computational techniques, such as ab initio methods and Density Functional Theory (DFT), with force-field methods, such as Molecular Dynamics (MD) and kinetic Monte Carlo (kMC), up to electrochemical models of the entire battery cell (often solved by Finite Element Methods; FEMs). Second, different <i>high-throughput methods</i> with the ability to analyze very large data sets and categories of compounds, for example machine learning techniques, are becoming rapidly available. Third, a stronger <i>correlation with experimental studies</i> is being employed for computational analyses, where the output data are directly compared and the models improved accordingly.

4.1. First-principles methods

DFT is perhaps the most extensively explored computational technique for organic energy storage materials, both for low-M$_w$ metal-organic salts and redox-active polymers. The methodology is straightforwardly used for predicting electrochemical voltage based on suggested molecular structures. By estimating redox potentials and accurately correlating it to the contributions from each part of the molecule, it is possible to provide selection criteria for molecular design. Such an approach was, for example, used by Tomerini et al. [132], who explored isomerism and N-substitution in pentalene dione derivatives. It was seen that these combined modifications led to a possible global tuning of the voltage to 2.2–3.6 V <i>vs</i>. Li$^+$/Li. Since the effect of isomerism alone was limited to 2.2–2.8 V, it was largely the (double) N substitution for CH motifs, which controlled the redox properties. Similarly, Zhang et al. developed theoretical methodologies to predict the performance of new organic electrode materials for Na-ion batteries [133], computationally evaluating the voltage of carbonyl-containing materials through Gibbs free energy before and after sodiation. Through the electronic

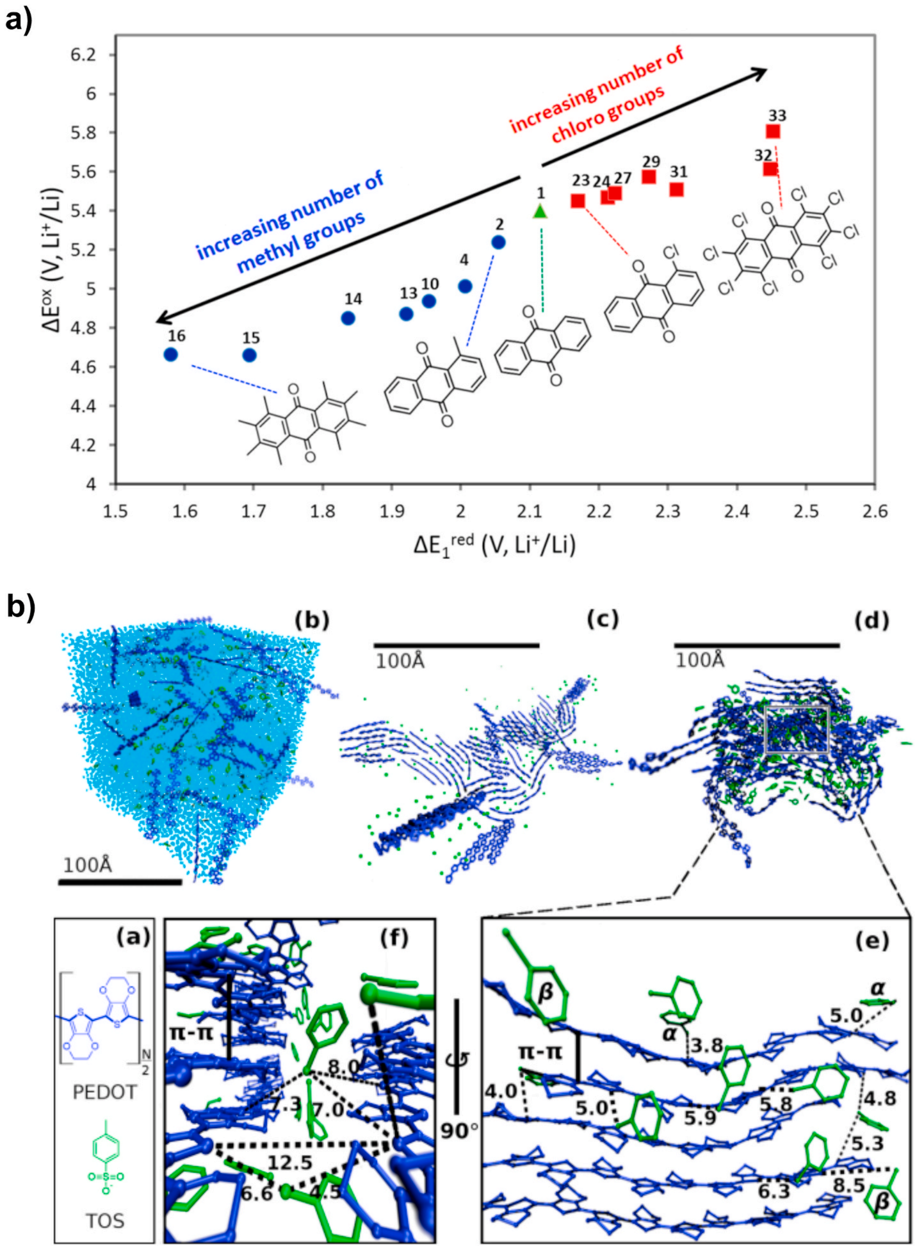

Fig. 5. a) Computed reduction (Δ<I>E</I>$_1^{\text{red}}$) and oxidation (Δ<I>E</I>$^{\text{ox}}$) potentials of selected anthraquinone derivatives. Reproduced from Refs. [139] with permission from the American Chemical Society; b) MD snapshots of the structure of PEDOT (blue, (a)) and TOS (green, (a)) either in water (b, c) or dry (d–f) on different length scales or perspectives. Reproduced from Refs. [153] with permission from the American Chemical Society. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

structure depicted by the density of states (DOS) throughout the electrochemical reactions, it was possible to see that the first unoccupied band became fully populated during the second sodiation step. The energy gap for the Na^+ insertion/extraction could thereby be evaluated.

For low-M_w crystalline organic electrode materials, DFT calculations also allow following the simultaneous changes in crystal structure with the electronic structure during the reduction process and the corresponding insertion of a counter-ion. Such studies have been important to understand the electrochemistry of this category of battery materials, which are challenging to explore experimentally [134–136]. Banerjee and co-workers [137], for example, performed a DFT study of the lithiation process of the classic dilithium terephthalate (Li_2TP) battery material and some of its derivatives, evaluating the impact of molecular modifications on the electrochemistry. It could thereby be observed that distinctly different redox mechanisms appeared during the lithiation of these compounds. For example, while Li_2TP displayed a disproportionation reaction, Et_2TP instead showed a stepwise redox reaction.

When modelling conjugated polymers for energy storage applications, a more systematic approach has to be employed in order to achieve an appropriate description of the desired electrochemical properties. An oligomeric model that is able to reasonably describe the properties of the polymeric material is necessary, which then constitutes a compromise between accuracy and computational power. As compared to non-conjugated systems, a sufficiently long oligomer has to reproduce the conjugation length, for example being six units for polythiophene [138]. This is also evident if observing the HOMO and LUMO states, which are fully delocalized for these systems. By extrapolating the redox potential versus the inverse of the number of monomers, the potential for high-M_w analogues can be estimated. Using this approach, Acker and co-workers [61] could computationally explain the superior cathode performance of alternating phenothiazine and bithiophene units in π-conjugated phenothiazine-based copolymers. DFT calculations showed that the localized phenothiazine redox center was maintained along the polymer chain for this structure, which resulted in well-defined plateau potentials on both charge and discharge.

In solution, the redox potentials can be evaluated following the Born-Haber thermodynamic cycle, including the solvation energy of the electroactive species. The assessed solvation effects and the use of the full Gibbs free energy are important for an appropriate description of the electrochemical reactions, which often involves interactions with liquid electrolyte solvents. For organic energy storage, this methodology has been applied to an extensive set of organic functionalities: quinones [139–141], ketones [142], carboxylates [143], nitroxide radicals [144,145], PEDOT [146,147], etc. For example, when Renault et al. [148] investigated the obscurely high capacity (1363 mAh g^{-1}) observed for dilithium benzenedipropiolate (Li_2BDP) when cycled under low currents, the DFT-based thermodynamic assessment could support that up to 16 Li^+ per BDP molecule could be inserted energetically favorably, thereby highlighting the possibility of such an extreme level of lithiation. Generally, studies of carbonyl compounds are particularly illustrative: Park et al. [142], Kim et al. [140] and Bachman et al. [139] have all assessed electrochemical properties, in particular redox potentials, for a large number of such derivatives. Through thermodynamic analysis, they could show interesting trends in how the number of carbonyls as active units alters the potentials in, for example, phenanenyl and anthracene, also showing good agreement with experiments. Moreover, by changing the number and character of the functional group on anthraquinone from electron-donating to electron-withdrawing, the reduction window and oxidative stability could be systematically tailored (see Fig. 5a). It was also shown how the dielectric properties of the solvent change the redox potential in this type of compound.

4.2. Molecular Dynamics simulations

Since organic electrode materials are often disordered, amorphous, or possess a tendency to not form highly symmetrical and periodic structures, they are generally problematic for straight-forward exploitation by electronic structure calculations that rely on periodicity, e.g. DFT. Using MD, on the other hand, means that molecular structures can be obtained also for complex and disorganized systems, which is perhaps especially useful for redox-active polymers. But since MD cannot intrinsically capture the redox processes that ultimately determine the functionality of any organic electrode material, the technique is often used complimentary to DFT when modelling organic energy storage materials.

One useful example is MD studies of the 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO)-based cathode material poly(2,2,6,6-tetramethylpipridinyloxy methacrylate) (**PTMA**, for structure see Fig. 3b) [149,150]. Large uncertainties exist regarding the structure and morphology of these polymers, their electronic transport pathways, how solvent and ion interactions with the polymer determine conductivity, how ion mobility in the polymer matrix occurs, etc., but which can be explored computationally. Through MD, it was possible to generate the fundamental morphologies to understand its global structure and assess the polymer-structure features associated with electron transport. Thereby, it could be seen that the TEMPO methyl-methyl interactions control the inter-nitroxyl distances, inducing a series of packing structures in the polymers. By then calculating the electronic properties using DFT, a strong coupling between rings on different chains was found. Thereby, the stacking motif allowed for charge transport over larger distances than if the electron is transported to the adjacent TEMPO rings. When adding acetonitrile solvent and BF_4^- counter-ions to the system, the main structural features remained intact. The BF_4^- counter-ions strongly bonded to the charged TEMPO motifs, irrespective of state of charge (SOC), which could explain the difficulties to fully discharge the resulting electrode, and suggested that ions with weaker coordination strength would be preferable for improved performance. Similarly, polyaniline (PANI) and its cyano-group functionalized counterpart were studied computationally by Chen et al. [151,152]. Since PANI comprises a substantial degree of disorder, several co-existing local energy minima exist, and thereby exemplifies the shortcoming of DFT methodology. Resorting to MD simulations showed to be fruitful to better map the configurational space and provided a route forward to pre-optimizing the possible structures, whereupon DFT could be employed for charge-discharge estimations.

Naturally, being one of the most extensively employed electroactive organic materials, PEDOT and its doped versions have been extensively investigated by different computational techniques. Also here, MD with electronic structure calculations has shown to be a powerful combination for scientific insights at multi-scale levels, where redox kinetics, mass transport and morphology can be studied simultaneously. Not least, Zozoulenko et al. [153–156] have been exploiting these methods to study a range of tosylate (TOS)-doped PEDOT materials (see Fig. 5b). A dry-state structure of small π-π stacked crystallites could be determined, forming an interlinked percolating network through the surrounding amorphous matrix. These swell and bend upon addition of water, but the π-π stacked distances were largely unaffected by solvent content or charge concentration. By coarse-graining the model, morphological structures approaching the micrometer length scale were possible to model for hydrated PEDOT-TOS with NaCl. The simulations could then elucidate the interplay between the components in the system and show how the ionic diffusivity decreased with doping level when the TOS counterions entered the PEDOT matrix. Percolation thresholds for the water level could also be identified.

4.3. Emerging computational concepts

Apart from materials modelling techniques, electrochemical modelling of cell devices using FEM is emerging as a versatile tool for conventional Li-ion batteries. Relevant studies also exist for organic electrodes. For example, a polypyrrole/nanocellulose [157] energy storage device was simulated based on Ohm's law and Fickian diffusion models, enabling a direct comparison with experimental counterparts. Thereby, the effect of electrode porosity on the electrochemically limiting processes could be elucidated, showing that charge transport was mainly controlled by the diffusion of counter ions in the electrolyte matrix within the electrode. With more extensive parametrization, employment of FEM-based methods are likely to be more extensively employed for organic electrode materials in the near future.

Data-driven methods, or so-called machine-learning (ML) techniques, are also likely to have a profound impact on the development of organic electrode materials. In these methods, statistical algorithms are applied to a dataset to build up correlations of underlying properties, which are then used to make predictions. Naturally, the success is highly dependent on the amount and quality of data. While data can be generated by both experimental and computational techniques, the latter is clearly useful for mapping a vast molecular space and construct a database, especially relevant for organic compounds. A vital part of constructing a useful ML approach is also to convert the molecular structures into 'descriptors', which can represent large sets of molecules and still capture their relevant properties. Approaches have been used to search for redox-active molecular materials for Li-ion batteries [158] or to predict properties of redox active polymers [159,160] based on a DFT-generated database. These examples have shown that very good predictions of redox properties can be achieved.

In a similar context, significant predictive power has been demonstrated by developments in high-throughput computational materials design (HCMD). Here, supercomputers are employed to virtually study thousands (or more) of compounds in parallel while targeting the best molecular components for a new material, exploring properties such as combinations of redox potential and capacity. HCMD work oriented towards redox-active organic molecules have indeed resulted in the proposal of a number of novel compounds, not least for flow-batteries. Er et al. [161], for example, proposed novel quinone-based electroactive molecules, while Flores et al. [162] analyzed over 10,000 functionalized thiophenoquinones and identified a large number of potentially useful materials for energy storage.

Finally, evolutionary algorithms constitute another novel computational approach, which has recently been applied to resolve crystallographic structures of organic electrode materials [163,164]. This method allows prediction of the crystal structures without any experimental input, and are therefore highly useful for analyzing the structural evolution of reactive low-M_w organometallic salts. By selecting crystalline structures in a free energy landscape based solely on the components of the system, the global energy minimum is approached through selection within a large structural space, which undergoes a number of 'mutations' after every generation. The method has been highly useful to study the complex lithiation reactions of compounds such as thiophenedicarboxylate and its nitroxide-functionalized analogues.

The potential molecular space for organic materials comprises billions of potential compounds for a rather limited amount of atoms, and whose redox behavior, capacity, stability, etc., are strikingly different. Since synthesis and experimental exploration of such an enormous amount of structures is impossible, the comparatively novel approaches of ML, HCMD and evolutionary algorithms are likely to contribute in this field to both assessing the fundamental structure-property relationships, as well as being employed as predictive tools for improved storage systems. This could act disruptively into bringing this category of energy storage materials tremendous steps forward.

5. Electrode construction

Recent advances in the field of organic batteries have mainly focused on the intrinsic properties of materials (redox potential, specific gravimetric capacity, specific power density, etc.). As a consequence, and as depicted in Sections 3 and 4, there are plenty of organic molecular or polymeric materials able to profusely store electrical energy over a wide range of potentials, thus opening the way towards full-organic batteries displaying an output voltage of more than 2 V and a correspondingly good energy density [24]. Despite this, a striking observation is the absence of a commercial prototype on the market, raising some questions about the veracity of the organic alternative and its efficiency to store electrical energy. These questions are legitimate and need to be solved to clarify if a decrease of the environmental impact of batteries, for instance, is possible or not. Having a first full prototype would be of high interest to situate this technology in the battery landscape in terms of energy density and to target specific applications, which would lead to a decrease of the demand of inorganic counterparts [86]. The lack of a full prototype could be explained by different factors, but is presumably due to the low energy density of the associated electrodes made from organic active materials. Indeed, it is well-known that organic materials require a large amount of conductive additive (from 30% to 70% in weight) to circumvent their insulating properties and display good specific performances. This fact has a dramatic consequence for practical applications, since these conductive additives constitute dead weight in terms of storage capacity. Thereby, the capacity of the entire electrode is generally divided by two or three.

5.1. *State of the art electrodes*

At the early stages of organic electrode materials revival, electrode construction was made by simply mixing active material with a conductive carbon additive, thereby mimicking well-established procedures originating from intensive research on inorganic battery materials. Surpassing insulating properties of organic materials was possible by mixing with carbon-based conductive additives with or without a polymeric binder through a time-consuming scheme, adapting the proportions of these compounds and their agglomeration to maximize the electrochemical performance of the electrode. Different types of carbon additive, such as carbon black [165], multi/single-walled carbon nanotubes [166], graphene or reduced graphene oxide [167] or carbon replica of mesoporous silica (CMK-3 for instance) [168] were successfully employed. However, up to date it is still not yet clear which of these is the best to build organic-based electrodes, since they offer specific and different advantages: for example, very high and oriented conductivity for nanotubes, or strong interaction with the active material for CMK-3 which prevents its dissolution and stabilize the capacity upon cycling. A trend that seems to emerge, however, is the adequacy of textures between the carbonaceous additive and the active material, which might favor charge transfer. This was underlined for Li2-DMT in a study by Lakraychi et al. [114] The way of mixing these components together, using different methods like milling [104], in-situ "coating" [169], wet impregnation with NMP solvent or direct crystallization/deposition of a soluble organic compound onto the surface of the carbon additive [170] have also revealed that the way of mixing is not the key issue for organic material. But despite these efforts, the storage capacities of these electrodes are still limited, since a large amount of conductive additive remains necessary (typically 30–70 wt%), and sometimes even the addition of a second conductive additive is required to obtain satisfactory performance.

At this stage, it is important to keep in mind that the electronic conductivity is not the only parameter limiting the performance. Indeed, since the electrochemical reactivity also implies cations or anions through an ambipolar mechanism, the mobility of these species within the electrode also affects the performance and, consequently, electrolyte penetration inside the electrode must be considered. While being well-known for inorganic-based battery material processing, the importance of a calendaring step and a relative porosity/tortuosity was pointed out a few years ago for dilithium benzyldiacrylate (Li_2-BDA) [171], revealing that this parameter could be tailored to allow for good electrolyte diffusion. Recently, this parameter was cleverly associated with a highly conductive additive to produce a high-performance negative electrode based on Li_4-PTC with only 0.5% of carbon, rendering real hopes that organic materials could actually be employed in high performances electrodes [172]. The difficulty resides in controlling the porosity of the entire electrode, where it is important to also consider the material's texture itself, which has an impact on the capacity and ionic conductivity/diffusion, as recently highlighted for dilithium naphthalene dicarboxylate (Li_2-NDC) used as a negative electrode [173]. In that case, the impact of material porosity was clearly demonstrated since the bulk material exhibits a rapid capacity fade while the mesostructured material present a more stable capacity upon cycling and higher conductivity attributed to a better ionic diffusion inside the material. Milling and formulation steps can affect the particle size and stability of organic compounds. To date, only layer deposition techniques (e.g. molecular layer deposition, MLD) have shown able to control the material's architecture to produce carbon-free electrodes, independent of the redox entities [174]. Although this method is interesting, it generally leads to the formation of very thin films, and therefore to limited storage capacity (0.41 µAh cm^{-2}), which prevents its use for the production of large quantities, but can be potentially interesting for miniature devices especially in medicine, such as healthcare monitoring.

Despite all of these attempts, the performance of organic-based electrode still lags behind that of inorganic-based systems. This is explained mainly by the lower number of studies dedicated to electrode optimization coupled to a clear understanding of the material reactivity and associated phenomena. Based on this fact, there is still a lot of progress margin to achieve the intrinsic theoretical performances of organic-based materials.

5.2. Perspectives on electrode development

Owing to the specific nature of organic compounds (i.e. solubility phenomena, charge transport inside the material, flexibility of the structure, chemical reactivity and stability, etc.), it is sometimes difficult to determine if the limitations belong to the electrode formulation or to the active material itself. It is clear that both aspects must be investigated together to reach the maximum electrode performances and, in the end, enable the building of a first full competitive battery on the market to benchmark this technology. Of course, in addition to the good electronic percolation brought by the conductive additive, it is essential to keep in mind that other parameters should also be considered. Electrode and material porosity and tortuosity are important to favor electrolyte penetration. Surface reactivity with electrolyte should be understood to limit and/or manage both dissolution of the active material and surface side reaction forming Solid Electrolyte Interphases (SEI) and Cathode Electrolyte Interphases (CEI). All of these parameters are important for practical properties, but cannot be carefully addressed without first solving the conductivity issues. This can be achieved by rethinking electrode architecture to consider electrolyte penetration, contact and charge transfer at the interfaces in close association with electrolyte management [175].

<i>At the material scale</i>, different strategies can be proposed to improve conductivity, especially the ionic mobility (ambipolar conduction mechanism). For existing materials of interest, the first logical approach, coming from inorganic materials, would be to reduce the diffusion pathway to speed up ion mobility inside the material. This is reminiscent of the $LiFePO_4$ story, which was set aside for a long time due to its intrinsic insulating properties, but finally emerged at the center of commercial batteries. Thus, the improvement of conductivity properties of organic material seems possible through nanosizing or meso-texturing the materials. Downsizing particle size of organic compounds was, for example, recently realized using an antisolvent method or electrochemistry, respectively, for sodium rhodizonate [100] and carboxylic acids [173]. However, such processes are of limited use since it is difficult to precisely manage the textural properties of the material and thereby determine the best size to maximize performances while limiting side reaction and dissolution. Templating methods seem interesting to meet this challenge since well-defined nano-object or mesostructured materials with a wide range of sizes can be obtained using "hard" or "soft" templating [176].

In addition to the particle texture, two other aspects should be investigated. First, it is evident that maximizing the surface area will induce an increase of the surface reactivity of the active material, which will probably affect the formulation with the conductive additive but could amplify or generate solubility/exfoliation issues, or favor surface reactions (formation of SEI/CEI). Second, the intrinsic reactivity of the organic material itself has to be considered, since it corresponds to the useful reactions of the material. At this point, it is difficult to clearly describe the reactivity of molecular organic materials on the basis of their reactivity in solution, since there is not a clear correlation between well-known molecular electrochemistry and electrochemistry of organic materials in the solid state. In solution, molecules can move to the current collector or electrode to exchange electrons and are close to the counter ion. In the solid-state, however, this is different. The spatial arrangement of molecules in the material (i.e. crystallographic structure) might hinder the charge propagation (absence of sufficiently large channels enabling, or not enabling, the diffusion of solvated ions) or influence the redox potential and the specific capacity. Furthermore, molecular "tipping" between the reduced and oxidized form (implying electronic density rearrangement and insertion of the ion) have an impact on the conduction pathway, as illustrated for conjugated lithium carboxylate [177] or anthraquinones derivatives [178], or even on irreversibility phenomena (the "yield" of an electrochemical reaction). This may explain why organic materials display asymmetric behavior between charge and discharge but, of course, this calls for further studies, for instance, using operando X-ray or neutron diffraction [177,179,180].

One interesting approach to overcome the conductivity issue (and formulation step) while understanding material reactivity would be to judiciously select the electroactive molecule in order to directly create conductive or semi-conductive supramolecular assemblies. In this sense, two examples of charge transfer complexes based on TCNQ derivatives (associated with phenazine or TTF core) were described recently [181,182]. In both cases, supramolecular and crystalline assemblies using TCNQ derivatives allow the formation of highly conductive materials enabling the formation of very low carbon content and binder free electrodes displaying high power and energy densities.

<i>Electrode construction</i> is at the crossroads of electrode performance. Based on general knowledge, it is essential to promote charge transfer between active material, conductive additive and current collector while limiting the electrode weight. Maximizing the surface contact appears to be the key point to obtain good performances and is in line with the increase of the specific surface area of the active material. Owing to the specific nature and properties of organic materials, electrode construction and architecture should be rethought, for instance, using new types of conductive additives like organic-modified carbons, metallic or hybrid particles, and conducting polymers. The latter represents the most suitable option since conducting polymers are composed of the same atoms as active organic materials (presumably easier interface management) and can add a contribution to the storage capacity while affording both binder and conductive properties. The way of associating them into the final structure is the key point. Ideally, a particle coating of a few nanometres would be the best alternative to reach high energy and power density, but this requires well-defined organic materials and careful investigation of their surface reactivity; aspects which are not yet studied. Another alternative would be to get inspired by other fields such as microelectronics. Many organic materials have been used in this field for applications including flexible screens, data storage, and light-harvesting, among others. In all these cases, the way of preparing the material affects their properties and performance [183]. One can notice that considering fabrication processes such as printing [184] and coating techniques to form, for instance, layer-by-layer electrodes (one layer of storage material and one of conductive polymer) might give an added value in the energy

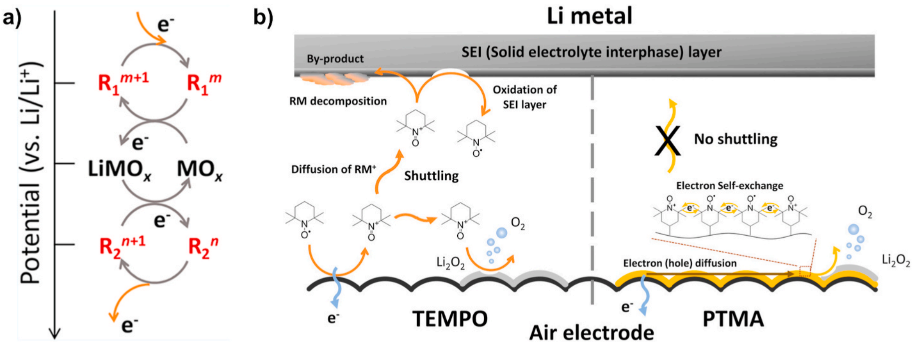

Fig. 6. (a) Schematics of the charge transfer mediation process between a target inorganic material (LMOx) and an organic redox system (R1,2). Depending on the relative position of redox potentials between the organic and inorganic phases, distinct charging (R2), discharging (R1) and combined both can be attained. Reprinted from Ref. [202] (b) Shuttling <i>vs</i>. no shuttling of redox mediators impact on the negative electrode in a Li–O$_2$ cell. Reprinted from Ref. [201].

storage field, especially in the view of the emergence of new technologies like solid-state [107] or new concepts like self-healing and sensing [185].

As introduced before, a dual strategy of investigation on both material and electrode scales in an <i>in-situ</i> mode is mandatory to cross new barriers for organic-based electrodes. <i>In-depth understanding</i> of charge propagation in the material and interaction with conductive additives need to be investigated systematically. Of course, electrochemical methods must be used extensively, but the association with spectroscopic techniques like NMR [186] and EPR would also be of great interest in order to follow ions/molecules and electrons [187] in addition to the recent studies realized using XRD techniques [177,180]. On the other hand, surface reactivity, especially with the electrolyte, should be investigated more intensely to gain a general understanding of these phenomena and to determine if this reactivity can be controlled. Addition of an additive can, for example, stabilize the performance of the electrodes and, above all, prolong their lifespan as much as possible. Different techniques will be useful for this, such as electron microscopy in association with analytical techniques to characterize solubility or erosion phenomena, or surface spectroscopy like XPS in order to unravel possible reactivity between the electrode and electrolyte and degradation upon cycling [188].

The quest for answering these questions is still at an early stage. Many answers are presumably possible owing to the different chemical natures and types of organic-materials that can be used to store electrons (n-type/p-type/both, rocking-chair or Li-metal systems, liquid or solid-state electrolyte).

6. Organic-inorganic hybrid electrodes

Organic redox materials are not limited to only their use as the main energy storage constituent in the electrode. Hybrid organic-inorganic compositions recently started to emerge as a means to mitigate the drawbacks of both technologies: limited functionality and power performances for the conventional inorganic battery constituents on one side, and low energy densities of the organic chemistries on the other side, to thus unlock high-power and high-energy performances. Furthermore, hybrid approaches to increase surface area, electrical conduction and enforce the mechanical properties, or tune the active material and electrode morphology have been reported [189], as well as devices that merge different energy storage technologies or redox mechanisms in the opposite electrodes (for example, Li-ion capacitors)

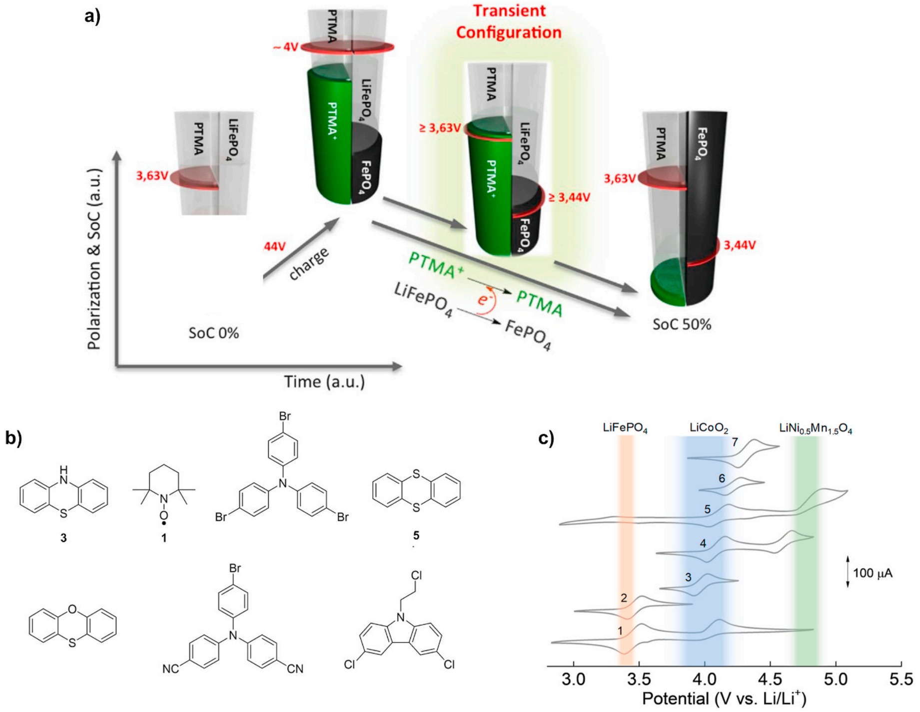

Fig. 7. a) Oxidative mediation process of a nitroxide polymer applied for fast charge of an LiFePO$_4$ electrode. An exciting feature of the hybrid LiFePO$_4$-**PTMA** electrode is that the difference in the redox potentials between the hybridized materials will always lead, at equilibrium, to the preferential charging of LiFePO$_4$ at the expense of **PTMA**. Nonetheless, **PTMA** is the faster recharge component resulting in a highly relevant practical fact: whenever the electrode needs to be recharged, the rapid response of **PTMA** will ensure fast recharge. Reprinted from Ref. [205]; b) Chemical structure of redox-active chemistries proposed by Oyaizu et al. c) Electrochemical response of compounds shown in b) overlaid with the redox potential of LiFePO$_4$, LiCoO$_2$ and LiNi$_{0.5}$Mn$_{1.5}$O$_4$ Li-ion cathodes. Adapted from Ref. [202].

[190].

When it comes to bridging the energy and power gap within a single device, the probably most intuitive approach is to combine, within the same electrode, the different types of energy storage sources that could provide this, namely supercapacitors with battery-like storage. Cericola and Kötz were amongst the first to review the advances in this direction, focusing mainly on hybridization between conventional EDLCs and LIBs [191]. However, the electrochemical response was found to be the sum of the responses of the separate components, with the contribution to the stored charge being proportional to the amount of each component, whereby the electrode configuration and composition control the power and energy delivery performances. The primary drawback is that power and energy performances are decoupled: at high current densities, the response was found to be dominated by the EDLC component, considerably diminishing the energy density of the hybrid device. Another broadly embraced approach for faster charge dynamics of the inorganic electrode constituents was through the use of conjugated conducting polymer composites, with many of these also displaying a reversible redox mechanism [192]. However, it is still not certain whether the observed improved power performances are due to the redox mediation, improved charge transfer or a combined effect of both, with most certainly the unfavorable overlap of redox potentials (Fig. 6) preventing the former effect. Recent developments also point towards slower electrode kinetics, with blocked mass transfer (or ion diffusion) due to low electrode wetting by the aprotic electrolyte for certain classes of conducting polymers [193].

Organic redox chemistries for fast charge mediation have their inspirational origin in the use of conventional (dissolved in electrolyte) 'redox mediators' in Li-ion cells (Fig. 6a) [194–196]. These have long been explored for overcharge cathode protection, given their fast redox kinetics and tunable operating voltages [197]. After reacting, these species are re-generated at the positive or negative electrode side. In addition to the conventional cell configuration, redox flow cells have also been shown to benefit from soluble redox mediators, when in particular using inorganic solid electroactive dispersions [198]. However, the conventional organic redox shuttles have little contribution to the stored charge, and their reactivity at the opposite electrode (Li metal, graphite, silicon) can inhibit their activity or pose safety, stability and cell complexity design issues [199]. Permanent fixation at the electrode surface can therefore be an efficient solution to this problem. For instance, surface-grafted molecular charge transport layers were shown to suppress the shuttle effect, while effectively unlocking the electrochemistry of otherwise inactive $LiFePO_4$ electrodes [200]. A very recent study particularly focused on the impact of the mobile and fixed redox mediator species as applied to Li–O_2 cells [201]. Whereas both approaches provided efficient Li_2O_2 oxidation mediation, SEI oxidative decomposition and TEMPO decomposition was observed for the mobile species configuration, leading to lower cycling stability (Fig. 6b).

The efficiency of the charge mediation will depend on the relative position of the redox potential of the active compounds (Fig. 6a) [202]. If the mediator has a higher redox potential than the inorganic constituent, it will most efficiently function as an oxidative mediator for the inorganic material (for example, as shown in Figs. 6b, 3.6 V for nitroxide-TEMPO/PTMA and 3.1 V for Li_2O_2 oxidation). For the reverse redox potential order, the reductive mediation will be the most efficient. In other words, fast charge or fast discharge rates can be attained depending on the relative position of the redox potential of constituents in the hybrid composite electrodes. The most studied redox chemistries for fast charge mediation are thus for redox-active polymers, with many of these bearing organic radical type redox centers similar to conventional redox mediators (Figs. 6–7). Other than the fact that these have recently been shown to have a finite electrical conductivity [19,203] (thus, also potentially contributing to enhanced charge transfer in the composite electrode), the additional advantage over conventional conducting (conjugated) polymers is that their redox response is well defined (given the charge localization <i>vs.</i> delocalization) so that the hybrid electrode design can be precisely controlled in terms of either fast discharge or charge mediation (Fig. 6a). The rate constants for electrochemical reactions of the polymers are lower than those of the corresponding monomers, due to the suppressed mobility of the redox moieties at the molecular level. For charge mediation, however, faster electrochemical reactions have been shown to be favorable, considering the heterogeneous charge transfer between the polymers and the inorganic electrode active materials as well as charge propagation throughout the electrode [204].

One example demonstrated by Gohy and co-workers was by designing and studying the charge transfer dynamics under fast charging conditions in a mixed $LiFePO_4$-nitroxide radical bearing polymer (**PTMA**) [205]. The hybrid electrodes made of $LiFePO_4$ with variable amounts of the polymer could be charged within several minutes by virtue of the electrocatalytic oxidation of the $LiFePO_4$ with the radical polymer. The study indirectly revealed that under high current density charging conditions, the polarization of the **PTMA** and $LiFePO_4$ constituents overlapped above the equilibrium values and both components were charged (Fig. 7a). The faster redox kinetics of **PTMA** resulted in its excess charging. When stopping the current supply, the potential of both components in the electrode tended to reach the equilibrium open circuit potential: 3.63 V <i>vs</i>. Li^+/Li for the **PTMA**/**PTMA**$^+$ couple, which is higher than that of $LiFePO_4$/$FePO_4$ (3.44 V <i>vs</i>. Li^+/Li). This led to a thermodynamically unstable/transient configuration, thus forcing a spontaneous oxidation of $LiFePO_4$ in the presence of **PTMA**. A combination of pulse charge and relaxation of the **PTMA**-LFP hybrid electrode was found to lead to an elevated electrode SOC in short time intervals.

The relative position of the equilibrium redox potentials of PTMA and $LiFePO_4$ enabled hybrid synergy primarily during the charge process (the rationale in Fig. 6 exemplified by experiments in Fig. 7a). During discharge, the effect was less pronounced, and it was only presumed that a similar behavior will take place in the reverse sense when hybridized with a LIB cathode material >3.65 V. This was later demonstrated by designing and analyzing the electrochemical response of a **PTMA**-$LiMn_2O_4$ (LMO) hybrid electrode [206]. Variable rate galvanostatic charge/discharge tests showed improved energy-power performance and capacity retention of the hybrid LMO/**PTMA** electrodes when compared to pure LMO electrodes. This enhancement was again correlated to a sequential power delivery mechanism brought by the faster kinetics of the PTMA constituent, subsequently recharged by the high energy and high voltage LMO constituent. Hybrid electrodes were also shown to deliver higher energy and longer cycle life when subjected to power pulse tests. The combination of the **PTMA** power buffer component with the LMO high-energy sink proved the potential of the hybrid energy-power electrodes in pulsed discharge applications as well.

Recently, Oyaizu extended this hybridization principle to other than nitroxide-based organic polymer mediators as well as to additional inorganic chemistries, including also the broadly commercialized $LiCoO_2$, NMC and high voltage $LiNi_{0.5}Mn_{1.5}O_2$ chemistries (Fig. 7b and c) [202]. To overlap the redox potential of the LCO and NMC, new 4 V class organic redox-active polymers were proposed. For example, a thianthrene (**5**)-based polymer with a potential as high as 4.1 V <i>vs.</i> Li^+/Li, successfully operated as an oxidative mediator for LCO charging, enabling a fast response with smaller potential loss. These results further support the benefit of the mediation concept to facilitate the charge/discharge reactions.

Whereas mainly the charge mediation functionality was targeted and demonstrated thus far, recent developments point towards additional functionalities that could be imparted to these charge-storing polymeric materials. Amongst the investigated and envisioned added functionalities, the following can be assumed (provided further development and optimization) possible:

- A rapid charge/discharge mediation process accompanied with overcharge protection, provided the intrinsic fast redox kinetics of organics as well as a suitable redox potential. The challenge here remains to identify stable and reversible chemistries that operate at around 4.5 V, the voltage of the current Li-ion technology. Some compositions (Fig. 7b and c) are approaching this, yet further improvements and analyses are required.

- If a suitable polymer structure (backbone) is designed and can be prepared, these could also serve as binder material for better cohesion of conductive additive and inorganic particles as well as adhesion to current collectors. Additionally, the ion reservoir properties of gel polymer electrolytes can be explored to further enhance the rate capability [207,208].
- Efficient charge transport can also be enabled in these materials (conducting polymer backbone for example) and used to improve the charge conduction in the composite/hybrid electrode. The intrinsic charge conducting properties has been demonstrated in nitroxide polymers, whereas an aliphatic backbone polymer nevertheless was shown to conduct through the pendant redox groups [19,203,209].

The last two aspects, albeit requiring further optimization, can lead to a complete elimination of non-active (dead) mass in the composite electrodes and thus to higher energy densities for Li- or Na-ion batteries. In fact, the <i>p</i>-type mechanism of many redox mediating polymers are compatible with both, as well as other metal-ion chemistries cells.

These developments clearly show that hybrid composites based on organic redox chemistries may have an important role to play in the future of electrochemical energy storage technologies. Organic chemistry is more diverse and versatile in designing and imprinting specific functionalities, and the same material could supply different properties and functionalities needed in an electrode. A conducting polymer, bearing redox centers that can undergo fast kinetics while also having the required mechanical properties and adhesion to current collectors and to active material particles, can be regarded as the ultimate additive of Li-ion batteries. A very recent example highlighted an ultrafast charge and discharge of a 99.9 wt% $LiFePO_4$ electrode, enabled by redox mediation of a redox-active fluoflavin polymer with a record low catalytic content of only 0.1 wt% [210]. The operating redox potential and the adjustment to the inorganic constituent can be tuned either by working on the redox center chemistry or by differentiating p-type and n-type processes via the ionic strength in solution and ion solvation structures on the potentials of different redox couples [211].

What is currently missing, however, are hybrid organic composites suitable for negative electrode charge mediation. The high power limitation of current Li-ion cells are due primarily to the negative graphite or graphite-silicon electrodes. High polarization during cell charge (corresponding to graphite or silicon lithiation) can lead to dendritic Li metal plating that can translate into safety issues, irreversible Li losses, and, ultimately, premature failure of the cell. Designing a redox mediator that can avoid polarization at the negative electrode could avoid these problems, yet it is also scientifically challenging as it will require reversible redox mediator systems that operate near 0 V <i>vs.</i> Li^+/Li. There exist no such reported chemistries yet, but polyaromatic compounds could be good candidates. Naphthalene, anthracene, biphenyl and other fused or conjugated polyaromatic small molecules are known to form stable anionic salts with alkali metals with a reversible redox process close to 0 V <i>vs</i>. Li^+/Li. This has recently been explored to realize an organic oxygen battery [199]. Since the redox species were free in solution, a special two-compartment cell (or side-by-side bi-cell) design was required to avoid migration of these reactive species to the cathode side. The difficulty here is that these poly-aromatic radical anions are stable only in solvated form (glymes or ethers), with the solid phase not being possible to isolate. It is now a challenge for organic chemists to design a polymeric material that would not allow this solubilization, yet swell enough to make a stable solvated radical anion polyaromatic chemistry for negative electrode redox mediation.

7. Organic full cells

After several years of intensive research to develop the reversible redox chemistry of organic electroactive materials, more and more full organic cell assemblies at the lab scale are now reported in the literature. The organic electrode material can be paired either with a conventional inorganic electrode material or with another organic compound (and sometimes with the same organic material in symmetric cell configurations). As mentioned in the introduction and in Fig. 1, the two accessible electrochemical storage mechanisms offered by organics (i.e., n- and p-types) make various cell configurations possible. We herein discuss selected examples [3], focused on both cationic and anionic "rocking-chair" cell configurations. Many dual-ion cells employing a p-type organic compound as the positive electrode material have also been reported [52,62,212–214] that can lead to good cycling stability even at very low temperatures (down to −80 °C) [215,216]. However, the drawback is that the electrolyte in this case acts as the reservoir of ions for the charge compensation within the electrode materials, so that substantial electrolyte volume is necessary [217]. Interesting achievements in full organic microbatteries to power small electronic devices are also reported in the literature, especially by Nishide, Oyaizu and co-workers [218].

Following the pioneering 1 V cell reported in 2009 based on lithiated oxocarbons [96], a second prototype of an organic LIB was reported by Chen's group based on dilithium (2,5-dilithium-oxy)-terephthalate ($Li_4C_8H_2O_6$) [103]. The cell exhibits an average operational voltage of $\sim$1.8 V and an energy density of $\sim$130 Wh kg^{-1} together with long cycle life (1000 cycles) when supported on graphene [219]. Very recently, the output voltage of this Li-ion organic system has been increased to 2.5 V by cationic substitution with Mg [99]. Similar chemistries have been applied in SIBs and KIBs. For example, Chen's group provided the first all-organic SIB through the sodiated counterpart of $Li_4C_8H_2O_6$. Such a cell works at an average operation voltage of 1.8 V with an energy density of 65 Wh kg^{-1} [101]. In addition, this group also reported the use of potassiated oxocarbons ($K_2C_6O_6$ as cathode paired with $K_4C_6O_6$ as anode) to assemble a KIB giving rise to an operation voltage of $\sim$1.1 V and an energy density of 35 Wh kg^{-1} [109].

One particular exciting option with electroactive organic compounds is the possibility of storing electricity through cell reactions devoid of metals (molecular-ion batteries); shuttling ion could be ammonium-type cations but also non-metallic anions. Thus a cationic rocking chair cell was assessed towards the reversible shuttling of a quaternary ammonium [46]. From an anionic rocking-chair point of view, however, very little has been reported probably due to the lack of efficient p-type negative electrodes. The first interesting anionic cell was studied by Yao's group [71] by assembling two redox-active polymers: poly(1,5-pentylene-4,4′-bipyridinium)dihexafluorophosphate as the negative electrode paired with poly(N-vinylcarbazole) as the positive electrode. This full organic cell achieved 1.8 V, but its performance was hampered by a noticeable capacity loss upon cycling. Good stability upon cycling was very recently achieved by pairing dilithium 2,5-(dianilino)terephthalate with an original double zwitterionic viologen derivative, but the resulting output voltage was lower than 1 V due to the selected positive electrode material [128]. In fact, there is room to promote anion-ion cells and push the concept of (metal-free) molecular-ion batteries forward.

For now, it must be recognized that the few full organic batteries tested at the lab scale are far from being competitive with their inorganic counterparts. A lot of chemical/technological innovation work remains to be done to improve energy density, power density and cycle life. As previously mentioned, the propensity of numerous electroactive organic materials to be solvated in aprotic liquid electrolytes is an issue, which requires the synthesis of truly robust organic scaffolds, unless using the other option consisting of designing all-solid-state organic batteries [220]. Besides, intrinsic limitations exist in terms of volumetric energy density values because organics possess lower densities. The irreversible capacity observed with several negative organic electrode materials during the first cycles is also problematic notably for the cell balancing. Hence it is likely that in the medium term the interest in using organic active materials will be considered only in half-cell configurations (e.g., vs. Li, Na) with n-type redox-active compounds [86], making the direct use of organic cathode materials prepared in their oxidized state possible [76].

8. Recycling and sustainability aspects

At this stage of this perspective article, it is relevant to provide certain factual data supporting that there is a real need for truly sustainable electrode materials in the future electrochemical energy storage (EES) landscape [24], and how organic batteries could be perceived as a part of the solution. This section summarizes a general inventory of the most decisive factors in this respect while highlighting the potential benefits of organic materials.

8.1. Inexorable trends and key factors

The first critical factor, which cannot be denied, is the ever-growing need for EES systems driven by the following two levers and fueled by the global economic growth [221]:

1. The threats of climate change call for radically curbing the share of fossil fuels in our energy engineering, particularly in transportation and power supply. Hence, rechargeable batteries are expected to play a beneficial environmental role in the management of intermittent renewable energy sources [222] (by ensuring the global safety of the future “smart grid”; kWh to MWh-range) as well as the deployment of decarbonized transportation systems through the massive use of electric motors (namely “e-mobility”; Wh to kWh-range) [223].
2. The worldwide boom in electronic devices, drones, service robots and digital technologies (mWh to Wh-range) [24,224], with probably low compensatory benefits from an environmental point of view.

Second, since the pioneering developments of electrochemical generators by Volta, Sinsteden, Planté, Leclanché, and Jungner in the 19th century, commercialized batteries (including primary batteries) still work thanks to the redox chemistry offered by inorganic species and especially the metallic redox centers, as well illustrated by the dominant LIB technology [225,226]. Tremendous progress has thus been achieved over the last three decades to make advanced batteries exhibiting increased gravimetric and volumetric energy densities while improving safety, power, lifetime, and cost. However, the use of inorganic chemistry to store electrons can pose some problems at large-scale perspectives because:

- Metallic elements are generally scarce natural resources [24].
- Such elements are provided through destructive mining operations with harmful effects and environmental pollution, especially when the chemical element is toxic [227].
- Their excavation can be very energy intensive and costly depending on the nature of the mineral deposit [228].
- Several refining steps and high temperature synthesis reactions are necessary to obtain the final electrode material.
- The high chemical stability of inorganic compounds complicates their recycling while generally requiring high energy consumption.

Recent and relevant data have been provided by Dehghani-Sanij et al. [226] in an instructive and comprehensive review on the environmental impact of batteries on people and the planet. As a first example, it is reported that the global consumption for making batteries already accounts for large fractions of annual produced raw materials, including metals and non-metals: lead (85%), cadmium (75%), cobalt (50%), lithium (46%), antimony (27%), lanthanum (10%), and natural graphite (10%). The sources of raw materials being limited on Earth and their supply uncertain or difficult due to excavation difficulties and geopolitical tensions, the necessity to ensure their supply have resulted in introducing the “criticality” notion [225,229,230] (together with critical metals (CMs) or critical raw materials (CRMs)) for all useful chemical elements in industry, which goes beyond the field of batteries. The classification level varies from country to country and changes over time. For example, the European Commission identified in its reports 14 CRMs in 2011, 20 in 2014, and finally 27 in 2017 [231] considered as fundamental to European Union economy and hence essential for the development and stability of each society. This situation should logically prompt the collection and recycling (for re-use) of spent batteries as well as the development of cells with higher round-trip efficiency. Nevertheless, regarding the latter aspect, some predictions reported by the International Energy Agency (IEA) have underlined that all the material demand just for electric vehicle LIBs will have to be supplied by resource extraction at least up to 2030, irrespective of the collection and recycling of used battery electrode materials, due to that the eight-to-ten-year lifetime of EV batteries will keep most of them in use by this year [222,232]. For comparison, spent LIBs used in electronic devices return to the factory approximately five years after production [232] (Table S1).

Based on this background and returning to the five points underlined above, the use of the reversible organic redox chemistry should offer some valuable assets because:

- Organic compounds are basically composed of the most abundant elements (i.e. light elements such as C, H, O, N, and S).
- Such elements are naturally present in the pedosphere, sea water, and atmosphere, including the particular biomass compartment [24] making the sustainable biosourcing possible (spurred by the emergence of biorefineries) [233].
- Organic precursors can also be provided by the well-established petrochemical industry; even today, crude oil processing is mainly aimed towards the production of fuels and only a small fraction is used for the synthesis of olefins and aromatics [234,235].
- Temperature synthesis reactions are typically below 150 °C; the challenge being to find the most suitable/efficient synthetic route [236].
- Organic compounds (polymer, small molecules and organic salts) can be either recycled at moderate temperatures (as recently demonstrated with PET via an original enzymatic route) [237] or used as fuel with energy recovery [238].

8.2. The tricky question of battery recycling of today and the future

There already exists an exhaustive literature with precise and recent data regarding recycling end-of-life (OEL) batteries, processing, and collection [232,239–248]. Following the concept of “urban mine” (or secondary resources for metal recovery) [249,250], the collection and recycling of spent batteries are obviously essential to reduce raw (inorganic) materials consumption while mitigating the global environmental impacts.

The demonstration has been made for a long time in OECD countries with lead-acid batteries (LABs) for which high rate collections are reported (>95% thanks to environmental rules due to the lead toxicity) and low-cost efficient recycling processes exist due to the simplistic battery chemistry [226,247,251–253]. The production of LABs is still noticeable as well as the need for fresh Pb [253]. The global lead production has risen from 5 Mt y^{-1} in the 1970s to 11 Mt in 2013 due to the increase in demand for LABs [254], especially in China with a capacity production having reached +200 10^6 kWh for that year [255]. Similar to LABs, well established recycling processes are applied for alkaline Ni-based batteries, with Ni, rare-earth elements, and Cd in focus [245].

However, too many of these batteries still spread out into nature and are not collected at the end of life.

Since they were commercially introduced in 1991, LIBs have been used extensively to power an increasingly diverse range of applications from small electronic devices to the emerging large-scale application markets (e-mobility, micro-to large-scale grid applications) coupled with impressive cost reductions in recent years [223]. Currently, consumer electronics make up the bulk of spent LIBs, but Richa et al. [242] predict that as many as 0.33–4 Mt of LIBs could be generated in the US from EVs between 2015 and 2040. Relevant data have also been reported by Winslow et al. [232] as shown in Table S1. Unfortunately, the recycling of Li-ion batteries is still nowadays limited to lower than 3% [226]. A first reason is the lack of international regulations to guide the recycling of LIBs [232]. For example, LIBs are managed as a general solid waste in the US, whereas the current European directive 2006/66/EC^{256} states that at least 50% by average weight of battery waste should be recycled (Tables S2–S3). Second, their recycling process is still essentially driven by economical profits following the current global economic model based notably on profit maximization and competition. In fact, the complex chemical composition of LIBs (Co, Ni, Li, Mn, Fe, etc.) complicates their recycling, explaining why lithium is not even recovered, and that pyrometallurgical processes (<I>T</I> ≈ 1400 °C) are currently favored. Moreover, the cobalt content (the highest value element) tends to decrease in new battery chemistries, which is not in favor of the purely economic equation of recycling. Although the recycling of LIBs requires a robust EOL infrastructure not existing nowadays, it is nevertheless expected that used EV batteries will emerge as a future waste management challenge.

No accurate data on the recycling efficiency of organic batteries can be provided at this time because they do not exist on the market yet. However, it can be anticipated that little attention will be paid to elaborate thoughtful recycling processes if the active materials mainly incorporate abundant chemical elements and/or are inherently inexpensive. Nevertheless, the question about the toxicity of organics could be raised that would lead to favor their destruction by combustion. Generally speaking, the possibility to discard spent batteries by simple combustion of unit cells is attractive not only in terms of energy recovery but also because the metallic elements of cells (e.g., current collectors) would be easily recovered [257]. Additionally, compared to the current technologies, if spent organic batteries were not collected, the loss of scarce and costly metallic chemicals will be notably reduced.

8.3. *The sustainability issue*

The extent of battery manufacturing and recycling processes (when collected) therefore appear significant because of the high energy consumption compared to other energy storage processes. Relevant data to evaluate the potential environmental concerns as well as the related energy cost for the production of batteries can be found in life cycle assessment (LCA) studies that should give a multicriteria vision of the different generated environmental impacts (i.e., impact categories) [243,258]. Although LCA can be considered as a standardized methodology (described in the ISO 14000 environmental management standards), it depends on the inventory database used, the system boundaries and even the Functional Unit (FU), which is used as a basis for the comparison of several solutions. Consequently, caution should be exercised because LCA results differ significantly due to these uncertainties. Obviously, the most instructive LCA data should relate to "cradle-to-grave" analyses (i.e., from raw material extraction through product use and disposal) with impact categories like Cumulative Energy Demand (CED), Cumulative Fossil Energy Demand (CFED) or Eco and Human Toxicity Assessment (EHTA). Unfortunately, existing studies deal with "cradle-to-gate" analyses (i.e., from raw material extraction to factory gate) with Global Warming Potential (GWP) as the principal impact category. Interestingly, Majeau-Bettez et al. [259] reported in 2011 impressive data regarding the life cycle environmental impacts of storing 50 MJ of electrical energy (FU) by comparing the Ni-MH technology with Li-ion batteries. More recently, Peters et al. [260] reported a very useful review of LCA studies performed on LIBs to better sort through the various data published in literature. After a thorough review of 113 available publications on the topic, a total of 36 LCA studies were identified as very reliable because they provide detailed results for LIB production and sufficient information to recalculate the reported results. The conclusion is that, on average, a cumulative energy demand of 328 kWh is needed across all chemistries to produce 1 kWh of stored electrochemical energy, producing GHG emissions of 110 kg CO_2-equivalents. Thus, while electric vehicles are claimed to be "environmentally friendly" because of their "zero emissions" during the utilization phase, the dominant LIB technology is not as benign as expected.

One important conclusion is that researchers and industry have to be focused on the entire life cycle of next generation batteries. The design of an effective battery system and its recycling process with low environmental impact require the assessment of the environmental performances early in the design process, as stated in the European directive 2006/66/EC [256]. As a reminder, the concept of sustainability [261] was developed in order to improve the present human living standards

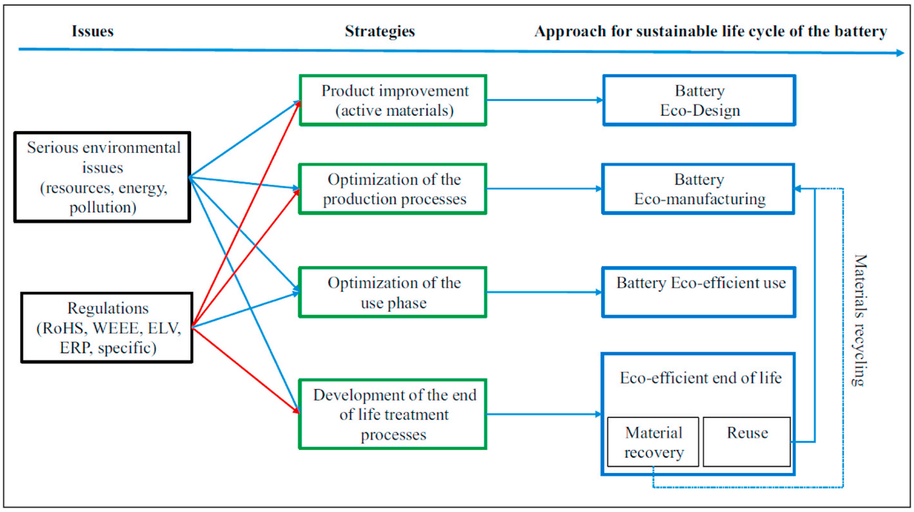

Fig. 8. Organization chart showing the eco-design in the battery life chain. Reproduced with permission from ref. [243]. Copyright 2015 Elsevier Inc.

while maintaining the availability of the natural resources for future generations. We should be sure that the generated impacts are actually counterbalanced by the improvement of the living standards on Earth (eco-design). As shown in Fig. 8, the sustainability of the battery life cycle depends on four stages going from the design of the new active material up to the EOL management. The development of an appropriate recycling chain is only the last of them without forgetting the potentialities offered by a second life of spent batteries (typically EV batteries) [243].

Without advocating the end of inorganic batteries, it has now been recognized that organic electrode materials can promote the fabrication of "greener" EES devices [1,262] by fulfilling several eco-design requirements shown in Fig. 8. Interestingly, Mercedes-Benz took advantage of the Consumer Electronics Show in January 2020 to present the VISION AVTR futuristic concept vehicle [263] using a "revolutionary" battery technology based on a graphene-based organic cell chemistry, compostable and therefore completely recyclable! Indeed, compared with their inorganic counterparts, organic electrode materials are expected to be less energy-intensive due to use of more abundant chemical elements with easier access to raw resources, coupled with low-temperature synthesis processes [24]. Simplified recycling management is also expected because organic structures can be easily consumed by combustion at medium temperatures producing heat with potential energy recovery [1]; biodegradability being also an option [264]. First described in 2008 [96], the proof-of-concept of the virtuous circle of sustainable batteries made from biomass was demonstrated in 2009 with the first all-organic Li-ion battery working thanks to oxocarbon lithiated salts derived from phytic acid [265]. More than one decade later, substantial advancements have been achieved regarding sustainable biomass-based electrodes in various EES technologies thanks to organics (not only as active electrode materials) [266]. To sum up, although much remains to be done to obtain attractive performances, organic batteries can be perceived as an alternative chemical and sustainable choice depending on the targeted application with a good margin of progress expected; the first commercial applications being expected in the field of Organic Redox-Flow Batteries [267].

9. Conclusions and outlook

As seen from the above summary and discussion, solid-state organic electrode materials continue to constitute a <i>promising</i> field of next-generation batteries. Promising, in the sense that it promises significant advantages as compared to competitor systems – not least the currently dominating LIBs – but also still quite far from being realized into large-scale implementations in the energy system. Thereby, direct comparisons with the Li-ion chemistries are often neither adequate nor straightforwardly made. LIB materials and cells have been explored by researchers for almost 50 years, and intensively tailored by engineers dedicated to commercial systems for more than 30 years. While inorganic LIBs constitute a mature technology with engineered components, the organic counterparts are still moving from infancy to youth. Performance targets adapted for the former systems and cell chemistries are therefore not easily met – and should not be easily met – for the latter.

It is not strange that this field, as is clear from the above summary, is dominated by a tremendous and rapidly growing flora of different novel materials, which are benchmarked electrochemically. This constitutes a natural starting point, just as it did for LIBs during the 1970–1980s. Without materials with a high theoretical and practical performance, there is little to develop further. We are thereby now arriving at a stage where challenges are likely to move from the basic development of electroactive materials to their incorporation into functional electrodes and cells, including tailoring of the electrolyte system and interfacial chemistry, inactive cell components, matching of electrodes, upscaling, battery pack production, etc., while avoiding the hurdles which have been plaguing the LIB field (i.e. ageing, safety, efficiency problems, etc.). There is, in this development, naturally a lot to learn from the LIB field, while the chemical nature of organic materials is inherently different, and input from other neighboring scientific areas will be greatly welcomed. While academic science can contribute in this development, it is industrial development which will likely bring this technological area to blossom, and which naturally will address the above issues. It would be of high importance for these valuable lessons learnt by researchers to find their way into potential products, where materials and systems can be further designed. Presently, the development of flow-batteries based on organic redox-active materials seems to undergo an even more rapid progress than their solid-state counterparts. At the same time, novel research approaches – not least the combination of high-throughput calculations, machine-learning tools, artificial intelligence and advanced robotics – are also likely to have a significant impact on this field.

This review has shown the advantages and challenges of several of these organic electrode materials. For redox polymers, perhaps the major advantage is their cycling stability, where they seriously outcompete most organic counterparts. The redox kinetics also seem to be of little problem, allowing for often very good rate performance in practical cells, where 100C has been achieved for nitroxide radicals or heteroaromatics as redox-active groups. It should be acknowledged that the type of polymer backbone greatly influences the specific capacity, rate capability and cycling performance of redox polymers. Therefore, apart from selecting the best redox-active group, the design of the redox polymer structure is of high importance. Small molecular counterparts, and metal-organic salts, instead hold promise of high capacity materials and energy-dense cells. The energy density can be tuned through adjustment of their redox potentials (molecular design) and/or optimization of specific capacities. While cyclability is still clearly an issue for these compounds, recent and promising examples have shown that this can be mitigated through molecular design, the use of solid or gel electrolytes, or optimization of the electrode through binders and inactive components. Another promising approach in a similar context is the exploitation of organic electrodes in hybrids with the inorganic materials, where their drawbacks are often straight-forwardly mitigated.

In context of the Strategic Energy Technology (SET) Plan launched by the European Commission, where specific performance targets are required "chemically neutral", it is easily seen where the main advantages and challenges exist for solid-state organic electrode materials. For example, these have already shown that they can reach the gravimetric energy and power density required, albeit not yet at cell level where the field is still too immature. However, the volumetric targets are considerably more difficult to approach considering the low density of most of these compounds. For applications where volume is of concern, for example passenger vehicles, these targets will likely be very difficult to meet. Moreover, while cycle life is not a major problem, at least not for redox-active polymers, calendar life according to the SET plan will be difficult to achieve. On the other hand, it could be speculated that many of these issues could potentially be resolved through employment of novel electrolyte systems, tailoring of the interfaces, and improved electrode fabrication strategies.

While the SET plan also addresses economical cost targets, it should be acknowledged that very little such data truly exists for organic electrodes. This could, however, be forecasted to not be much of a challenge considering the general cost levels for non-exotic organic materials, not least if biomass precursors can be utilized. Moreover, recycling and sustainability are likely, as described, the major advantages for these storage systems, and will constitute the strong selling point for moving to organics.

These organic materials and systems are thereby still awaiting their dedicated part of the energy storage market. It is, for example, noteworthy that organic electrodes could be useful to power single-use (disposable) devices [264]. While an application in conventionally sized vehicles is likely too challenging, and beyond the possible powering of small devices, stationary storage is often mentioned as a niche. For these applications, calendar life needs to be significantly improved.

It should also be acknowledged that this has to be done without large compromises in energy density, which is not always addressed in the discussion. Large-scale batteries for stationary storage will require costs for maintenance, installation, etc., while they also most likely will consume large amounts of non-active materials – the lower the energy density, the larger will be these costs [268]. Furthermore, if the cost is small for these storage devices with upscale of production, and volumetric energy density can be kept at a reasonable level, it will open up for utilization also in consumer electronics and similar devices where life-time is not as critical.

CRediT authorship contribution statement

Birgit Esser: Conceptualization, Writing - original draft, Visualization. **Franck Dolhem:** Conceptualization, Writing - original draft, Visualization. **Matthieu Becuwe:** Conceptualization, Writing - original draft, Visualization. **Philippe Poizot:** Conceptualization, Writing - original draft, Visualization. **Alexandru Vlad:** Conceptualization, Writing - original draft, Visualization. **Daniel Brandell:** Conceptualization, Writing - original draft, Visualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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