

## CRITICAL REVIEW

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## A toolbox for improved recycling of critical metals and materials in low-carbon technologies

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The shift towards renewable energy sources combined with other factors, such as population increase, digitalisation, and a need to decrease carbon footprint, leads to increasing metal consumption. To meet this growing demand and avoid accumulation of waste in landfills, efficient recycling methods are needed. Current pyrometallurgical and hydrometallurgical methods achieve complete digestion of end-of-life materials using high temperatures and high consumption of chemicals, respectively. These methods can be applied to recover critical metals from end-of-life materials but suffer from inherent limitations when it comes to complex end-of-life materials made of interpenetrated layers of metals, inorganics and organics. This critical review describes a set of chemical and physical tools for improved recovery of metals from various waste streams, with a strong focus on the renewable energy sector (wind turbines, solar cells) as well as lithium-ion batteries and catalysts for hydrogen production. These tools target weaknesses at the interfaces between different layers to liberate the valuable metals. Physical methods used for size reduction and separation, ultrasound to process brittle materials, hydrogen decrepitation, selective dissolution and bio-metallurgical methods to process metals are among those reviewed. Management of inorganic and organic fractions is also emphasised, with pyrolysis and solvolysis to process organics and ways to recycle these materials. Limitations and future directions are discussed, providing a comprehensive guide to improve recycling of metals with versatile tools.

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## Sustainability spotlight

Climate action and access to clean energy is heavily reliant on metals. Recycling of metals from renewable energy production and storage devices is highly desirable but is challenging due to the complex architecture of these devices, made of interpenetrated layers of organics, inorganic and dispersed metals. In this critical review, we highlight and discuss different technologies that can potentially reduce the environmental impact of current recycling methods and improve the global recycling rates. This review falls under multiple UN sustainable goals such as goal 12 (responsible consumption and production), goal 7 (affordable and clean energy production) and goal 13 (climate action).

## 1 Introduction

Industry 4.0 coupled with an expanding world population is driving an increase in metal consumption and the complexity of material structures. This is exacerbated by the transition from fossil fuels to renewable energy sources<sup>1</sup> and the need for devices such as batteries, fuel cells, solar cells, wind turbines,

display devices, light emitting diodes (LEDs) lighting as well as the electronics that enable them. There is a need to transition towards Net Zero solutions, and reduce carbon emissions in all areas of our society. This will require widespread technological and societal change and technology transition in many different areas. Future transition pathways will require more sophisticated technologies to enable decarbonisation, with an increasing array of elements employed<sup>2</sup> in more sophisticated technological structures than incumbent technologies. The trend toward complicated architectures with thin, multi-layer coatings is causing issues with dilute, complex waste-streams containing dispersed technology critical metals (TCMs). Those elements are increasingly important to the economy due to the aforementioned reasons, and are at risk of supply disruption due to a combination of factors (geopolitical, geological, economic...).

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Bulk alloys based on iron and aluminium are mainly used for construction, automotive and machinery and are relatively easy to collect and separate by mechanical sorting and pyrometallurgical methods. This makes up the majority of the metal recycling market as these metals are a major component of construction and transportation infrastructures. The main limitation of pyrometallurgical methods is that they are innately energy intensive. Certain other metals such as lead or platinum group metals (PGMs) are used on a lower volume but their environmental toxicity, ease of recycling and high value ensures their recycling efficiency is higher as it is economically profitable to do so and legislation generally ensures they are processed in a circular economy.

Recycling of TCMs as presently conceived, generally includes their liberation from their initial casing using pyrometallurgical or other physical methods (comminution). Further purification and recovery usually requires the use of hydrometallurgical methods. Limitations of these methods are the high consumption of chemicals (particularly water) and the generation of wastes, which emphasises the need for circular processes, where chemicals and water are re-used over several (typically >10) cycles.<sup>3</sup>

Waste Electronic and Electrical Equipment (WEEE) is one of the fastest growing sources of waste worldwide due to the rapidly increasing disposal of malfunctioning or obsolete electronic devices. WEEE consists of an array of discarded electronic products such as computers, mobile phones, televisions, fridges, medical devices and other electronic appliances that have reached the end of their useful life. An abundant literature is available on metal recycling from WEEE, where authors mainly aim to recover precious metals from printed circuit boards (PCBs). However, WEEE suffers from poor levels of recyclability<sup>4,5</sup> which can be linked to high statistical entropy<sup>6–8</sup> and low metal content, and ultimately unfavourable economics. The amount of TCMs in WEEE is generally low, and taking into account the low collection rates, absence of detailed material stack information about various state-of-the-art products (adhesives and encapsulants), and difficulties to recycle the products, it unfortunately often leads to almost negligible recovery.<sup>9</sup> Recycling these products is however important from an environmental point of view and there is a clear need for an improved regulatory framework as well as technologies to improve collection, sorting and ultimately, recycling rates. To do so, digital products passports<sup>10,11</sup> based on blockchain and automatic sorting<sup>12</sup> using artificial intelligence are emerging tools that will certainly help improve recycling rates.<sup>13</sup>

The aforementioned issues are less pronounced for emerging sources of end-of-life materials from clean energy technologies. Solar panels or wind turbines can be found in a single location and do not move throughout their lifetime (their lifespan being predictable), facilitating their collection and producing more constant waste flows. Electric vehicles also provide a source of large concentrations of TCMs and although they are mobile, extended producer responsibility (EPR) is likely to enable their pre-sorting prior to processing when a mature market has established.<sup>14</sup> There are calls for EPR to be extended to all aspects of sustainable energy production including wind

and solar power, though clearly logistical and legal challenges exist with such long-lifetime products. Most western countries have targets<sup>15</sup> for recycling rates, such as those set by the European Union WEEE directive, but in most cases these fall short of defining who is responsible for recycling the products as well as reaching their national targets.

Increased content of critical materials and increased economic importance and value of the metals used (*e.g.* PGMs for hydrogen production) would certainly lead to economically profitable recycling, lower statistical entropy and higher recyclability of these products. However, valuable metals are also located within complex, multi-layer architectures which are not designed to be recycled. This is why efficient recycling processes with workflows for systemic removal/recycling of protective and encasing layers, and separation from bulk substrates and active materials (like semiconductors) are urgently needed, particularly to reduce demand for carbon-intensive mineral extraction. According to the international energy agency<sup>16</sup> and considering a “sustainable development” scenario, the demand for rare earth elements for wind turbines in 2040 would be higher than 10 kt (while it is lower than 5 kt in 2020). Demand for copper and silicon for solar cells would be close to 1000 and 800 kt, respectively (increasing from less than 400 kt for both copper and silicon in 2020). Cumulative demand for battery materials required for electric vehicles (lithium, nickel, cobalt, manganese and graphite) by 2040 could be close to 12,000 kt while it is lower than 1000 kt in 2020.

Moreover, increased use of these technologies obviously lead to a surge in end-of-life materials generation, with more than 1200 GW h<sup>16</sup> of spent batteries available by 2040 and a cumulative amount of up to 78 million tonnes<sup>17</sup> of end-of-life solar panels available worldwide by 2050. 421 MW of wind power was decommissioned in Europe in 2018.<sup>18</sup> It is estimated that 424 to 3305 GW of wind power will be installed by 2040,<sup>19</sup> each of those wind turbines having a lifetime of 20–30 years and containing 25 to 65 kg of NdFeB magnets<sup>20</sup> per GW and 10 tonnes of blade materials<sup>21</sup> per GW. A significant proportion of PGMs supply comes from recycling,<sup>22</sup> with 37 tons of waste PGMs produced from end-of-life vehicles in China alone in 2020.<sup>23</sup> The catalysts for proton exchange membrane (PEM) fuel cells need around 300 g of platinum and 700 g for iridium per MW, with an increasing global PEM manufacturing capacity of around 500 MW per year at the moment.<sup>16</sup>

With current metallurgical processes, it is challenging to deal with complex materials composed of different layers (metals, inorganics and organics wrapped together). Metal recovery and separation is not straightforward, as metal concentrations and compositions are unknown when compared to traditional ores. Current purification technologies have been developed for separation of critical metals from ores (often separated as by-products of other metals like zinc and copper). However, the composition of end-of-life materials differs greatly from primary minerals. Therefore, a process developed for metal purification within the frame of primary minerals extraction will need to be adjusted (in the best case scenario), in order to take into account differences in the concentration of target metal, nature of competing ions, presence of plastics. In



most cases, a new process will be required for each recycled product. Current techniques often oxidise metals, generally after shredding of the end-of-life materials, while the inorganic and organic layers are usually lost. TCMs are often further diluted and lost during the process, which contributes to lower recycling rates.

With both pyrometallurgy and hydrometallurgy, a large amount of energy and/or chemicals is needed to purify the metals, while large investments are needed to treat the hazardous gas or wastewater produced from the process. Both techniques are well-known and have been used for centuries to process minerals, but their application to metal-containing end-of-life materials is challenging, particularly due to the presence of layers of inorganics and organics.

From a sustainability perspective, the main issue with hydrometallurgy is that stoichiometric reagents (usually oxidants) are used. It should be highlighted that aqua-regia or other strong oxidising agents are often used, and this is primarily sulfuric acid which is often from waste sources and hence of lowest cost. A more sustainable and safe approach would be the use of catalytic oxidants, particularly using natural or earth abundant elements or compounds.

Knowing the growing importance of metals, the rise of a circular economy of metals is desirable to reduce the pressure on supply chains, but also to reduce the demand for mining. Although reducing and re-using strategies must be preferred over recycling ones, it is clear that recycling will play a major role in the coming decades, knowing the large number of devices used for clean energy production and storage that will reach their end of life. Reducing the environmental impact of recycling processes is necessary to favour the social acceptance of urban mining and the implantation of new recycling facilities.

This critical review highlights a “toolbox” of processes to deal with the different layers. The tools described herein aim at liberating metals efficiently, avoiding dilution and facilitating purification by targeting the weakness at the interface between different phases. These processes should be designed for circularity (reusing chemicals and/or energy) in order to limit energy and chemical consumption, and to reduce emissions (gases, dusts and volatile chemicals). This review discusses the merits and limitations of each tool and aims to provide guidance amongst the existing and emerging tools to improve recycling of TCMs.

Although most of these techniques could be used for WEEE recycling, the main focus of this paper is on emerging streams of end-of-life materials used for renewable energy production (solar cells, wind turbines), energy storage and electric vehicles (lithium-ion batteries (LiBs)), and PEM fuel cells for hydrogen production as they are more likely to be collected than WEEE. Moreover, a significant proportion of these devices contain organic (polymers) and inorganic (such as fibreglass) materials, which inhibit metal recovery. Their removal is often needed to access the metals, while they often bear little value and possibly have a high impact if dispersed in the environment. This review highlights tools that can be used to treat these non-metallic materials and improve global recycling rates of products.

## 2 Results and discussion

### 2.1 Architecture of metal-containing end-of-life materials

Recycling of metals from various end-of-life materials is complicated due to the presence of many diverse and inter-penetrated layers of materials. A representative example would be crystalline silicon solar cells made of different layers of inorganics (glass, silicon wafer, silicon nitride), organics (poly(ethylene-vinyl acetate) (EVA) polymer) and metals (aluminium and silver electrodes, copper wires, and aluminium frames). It is therefore necessary to deal with each of those layers by targeting weaknesses at the interface between different layers, using sustainable processes if possible.

The metals are generally reactive, and can be oxidised, although some of them will be more difficult to oxidise than others (high redox potential or formation of passivation layers). Inorganics are generally unreactive, but they are generally of brittle nature and physical methods can be used to remove them. Physical methods are less useful on organics, which are generally not brittle and are also difficult to oxidise. But organics can be removed by pyrolysis, solvent-assisted dissolution or other physical and chemical methods.

It should be mentioned that although metals are the target of the recycling process as they often bear most of the economic value, it is often vital to process the other layers in order to access the metals, as they are wrapped into multiple layers. This is particularly true for LiBs, solar panels, fuel cells, and wind turbine magnets, which share a complex architecture (detailed structure being shown in Fig. 1). This architecture is shared with many different sources of metal-containing end-of-life materials such as PCBs (composite materials and metals), thermoelectrics (ceramics and metals), laminate films (polymers and metals), and others.

There may also be opportunities to up-cycle some of the non-metallic materials into higher value products. It is also necessary to take into account the significant difference in value between different metals within the same waste streams. As most of the process focuses on the expensive precious metals, they often represent a low fraction of the total metallic content and there is a need for selective dissolution methods. An undiscerned dissolution of all metals leads to unnecessary consumption of chemicals and impure product streams. A prudent approach would be to oxidise and dissolve metals selectively, only when needed.

A common first step in most recycling processes would be to characterise the end-of-life material to identify the value and the weakness of the material. The weakness of the material would be a part that can be removed more easily than the others, allowing recovery of the valuable part without having to process the rest of the material.

### 2.2 State of the art of the recycling techniques

Current metal recycling facilities often combine pyrometallurgy and hydrometallurgy to treat the multi-component metal-containing end-of-life materials and recover TCMs.<sup>27,28</sup> Through the action of heat, pyrometallurgy is modifying the



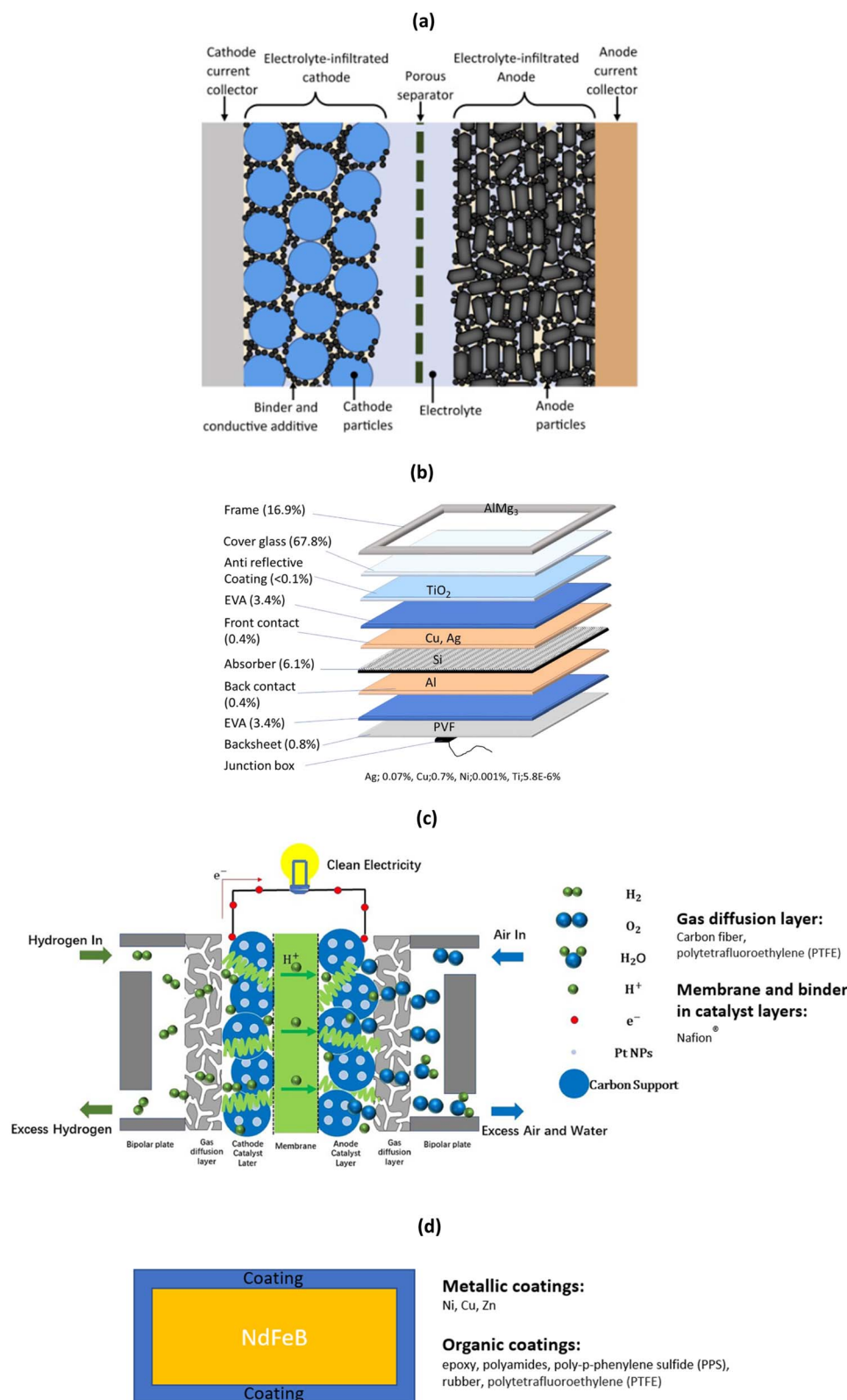


Fig. 1 (a) Detailed structure of LiBs (reproduced with permission from ref. 24), (b) solar panels with crystalline solar cells and typical mass percentage of various materials (reproduced with permission from ref. 25). (c) PEM fuel cells for hydrogen production where Pt NPs stands for platinum nanoparticles (reproduced with permission from ref. 26), and (d) coated magnets and commonly used coatings.





three layers and turning them into different products (which is summarised in Fig. 2). End-of-life materials are first treated at high temperatures in a copper smelter, where most of the metals are turned into a copper bullion containing precious metals and copper. The inorganics such as ceramics (as well as some metals) result in a slag which is further processed in a lead blast furnace to produce a lead bullion.

The organics are used as a source of fuel for the process. If the organic content is too high, removal of organics through physical methods may be required prior to pyrometallurgical treatment. Some of the by-products could be condensed to produce useful chemicals (such as sulphur dioxide to produce sulphuric acid).

Reductants may be present, or they may be added. For some waste streams, *e.g.* LiBs, the presence of aluminium in the waste stream will act as a reductant, but additional supplementary reductants may be added depending on the metal to be extracted. Hydrometallurgy is then applied for the leaching of the copper bullion, which allows dissolving copper and recovering it by electrowinning. The precious metal residue is then further treated through cupellation (blasting hot air over the molten metal) to recover Au, Ag, Pd and Pt. Similarly, the lead bullion is refined to recover special metals (In, Te, Se) and lead by-products (Sn, Bi, Sb).

Pyrometallurgical processes are energy intensive and they treat without discernment the inorganics, organics and metallic layers. The inorganics often end in the slag and a higher amount of inorganics increases its volume. A larger slag volume leads to higher losses of metals, particularly challenging during smelting of spent LiBs (losses of Li, Mn, Al...). Other elements

with a higher volatility (Pb, As, Sn, In, Cd...) can be found in the flue dust, which must be managed.<sup>29,30</sup> Degradation of organics at high temperature could lead to the production of valuable chemicals, but also to volatilisation of toxic and hazardous molecules such as brominated flame retardants or polychlorinated biphenyls, often encountered within WEEE plastics, condensers cables, housings, *etc.* Specific treatment of the gas is necessary to avoid release in the environment of hazardous substances. With this regard, the processing of end-of-life materials containing p-block elements (*e.g.* Cl, Br, As) is challenging using pyrometallurgy.

Other approaches involve first to shred the end-of-life material to reduce its size, then separate different fractions using physical separation methods prior to application of hydrometallurgical methods for metal purification.<sup>31</sup> The complexity of the material may necessitate disassembly of the end-of-life material as a prerequisite. Due to the variability of the feedstock, this step is often done by hand and is labour intensive, although it can be automated to some extent, using desoldering (using mostly high temperatures or oxidising agents), automatic disassembly<sup>32</sup> or other thermal, physical or chemical methods.<sup>33</sup> This depends on the value and concentration of the TCMs. Studies on LiBs have shown the importance of disassembly rather than shredding on the time and cost/profitability of recycling.<sup>34</sup> The complexity of the materials can also make manual disassembly too slow and too expensive depending on the cost of labour which adds a geopolitical aspect to recycling.<sup>13,35</sup> Some have posited that in the future "Industrial disassembling" will be a prerequisite to an efficient circular economy of TCMs, in preference to manual disassembly.<sup>36</sup>

The size of the materials is then reduced, which has multiple benefits. First, it liberates the TCM fraction which is often encapsulated within different layers of inorganics and organics. Secondly, it allows application of physical methods to separate and recover the organics and inorganics, based on the difference in their physical properties. Thirdly, the dissolution of metals from the metallic fraction is more efficient when the size of the particles is smaller. Hence, hydrometallurgy is applied to dissolve the metallic fraction, which is often carried out with harsh stoichiometric chemicals (strong bases, mineral acids, strong oxidisers, *etc.*). These types of reagents also need neutralising and produce gases which can affect the sustainability of the overall process. The preference for reusable reagents must again be stressed from a sustainability perspective. Some TCMs are found as minor constituents in devices, such as rare-earth elements (REEs) in windscreens, sensors, touchscreens, and headlights in vehicles. These may be too diffuse to warrant dismantling and recovering. This may be an aspect of where responsible innovation<sup>13</sup> is required to design out elements which cannot be used sustainably.

Similarly to pyrometallurgy, hydrometallurgical approaches suffer from the indistinct treatment of the different layers. First, the liberation of metals encased in plastics and inorganic layers leads to losses of metals. The size reduction step is energy intensive in common mineral processes, contributing for 36% of the energy consumption of the mining industry in Australia,

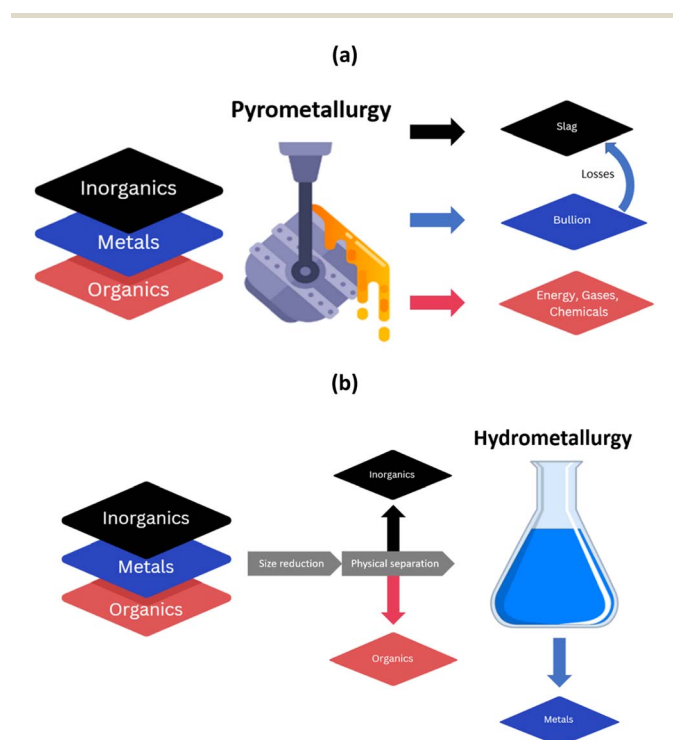


Fig. 2 Modification of different layers through the application of (a) pyrometallurgy, and (b) hydrometallurgy.



and more than 1% of Australia's total electricity consumption.<sup>37,38</sup> Energy consumption for comminution (crushing and grinding) of gold-containing rocks from underground mines is in the range 15–32 kW h per ton of ore.<sup>39</sup> Energy consumption for the crushing of end-of-life Li-ion batteries is in the range 4–12 kW h per ton, depending on the nature of the housing materials, size of the crushed materials and pre-treatment steps applied.<sup>40</sup> These numbers seem to indicate that crushing of end-of-life materials is expected to consume less energy than crushing of primary rocks, but size reduction will remain a major contribution to the global energy consumption of the recycling process.

Further losses of metals are possible during the physical separation steps. The dissolution step is often non-selective, which requires further separation of metals with chemical-intensive techniques (ion exchange resins, precipitation, solvent extraction, *etc.*). Wastewater treatment is then needed after the metals are recovered to deal with dissolved salts, suspended solids, organic solutes and pH issues. For all of these reasons, new processes based on a layer by layer approach are needed, preferably with selective dissolution. Complete digestion of base metals often makes them uneconomic to recover. Sequential delayering processes would be more efficient in order to improve recycling rates and recover not only metals but also inorganic and organic layers.

A typical smelter uses temperatures of 1200 °C,<sup>41</sup> while most hydrometallurgical processes are applied at lower temperatures (generally <100 °C). But electricity consumption of hydrometallurgical processes remains an important feature for size reduction and physical separation steps, heating and agitation of leaching solutions, and electrodeposition of metals. A comparative life cycle assessment of pyrometallurgical and hydrometallurgical approaches for the recycling of electronic waste shows that hydrometallurgical processes have a lower carbon footprint<sup>42</sup> (18 tons of carbon dioxide per kg of gold, compared to 58 tons of carbon dioxide per kg of gold using pyrometallurgy). Hydrometallurgical processes have a higher impact for most of the environmental factors considered (acidification, eutrophication, ecotoxicity), which is mainly due to the chemicals used such as acids and oxidising agents for the dissolution of metals.

## 2.3 Physical methods to concentrate metals

**2.3.1 Reducing the size.** Reducing the size of the materials facilitates post-processing of the materials. Leaching efficiency of metals using hydrometallurgy is heavily dependent on particle size and usually increases with reduced particle size.<sup>43,44</sup> From an health and safety perspective, the major issue with size reduction is the generation of hazardous fine dusts and particles which require specific treatment systems using dust collectors, filtration systems and dispositives to avoid emissions from the plant (tetracyclone, electrostatic precipitator, activated carbon, water spray, *etc.*).

It is useful to first identify which size-reduction method is the most appropriate. Hammer mills, shredders and granulators are the most common options. Shear, compression and

abrasion forces are used in these processes to break the interfaces and liberate the metals from their organic or inorganic casings. Shredding and grinding of WEEE is a common operation in existing WEEE recycling facilities. Within PCBs, metals like copper are found between layers of resins and their liberation by size reduction is easy. During the shredding of PCBs, aluminium is usually liberated in the coarser fractions. Copper and ferromagnetic metals are liberated completely when the size is further reduced. But interlocking of organics and metals is still an issue with other end-of-life materials like solar cells, where plastic-rich fractions still contain metals after shredding.<sup>45</sup> The smaller fractions (<0.25 mm) contain up to 70–80% of silver depending on the nature of the solar cell, which leads to losses in the range 20–30% for silver (the most valuable metal found in crystalline solar cells). The degree of liberation of metals from end-of-life LiBs is high as long as the particle size is reduced down to under 100 µm.<sup>46</sup> Reducing the size down to very low small particles may be energy intensive or not possible depending on the nature of the end-of-life material. Moreover, losses of precious metals as high as 40% have been reported during the size reduction steps of WEEE.<sup>47</sup>

Ball milling could combine size-reduction and oxidation or reduction of metallic species. This mechano-chemical approach allows redox reactions to occur without any solvent. The milling step is done by mixing the end-of-life material with solid chemicals like potassium persulfate, which allows the oxidation of metals from WEEE.<sup>48</sup> Other reagents like iron allow the reduction of cobalt from spent LiBs and facilitate metal leaching.<sup>49</sup> Other chemical reactions are possible with chemicals such as sodium carbonate to recover gallium from LEDs.<sup>50</sup>

Fracturing techniques are used in the mining industry to process rocks<sup>51</sup> and could be adapted to process end-of-life materials. Microwave heating,<sup>52</sup> cryo-milling<sup>53</sup> and high-voltage pulses<sup>54,55</sup> were reported, mainly to remove inorganics and organics fractions and to liberate the metals. The high voltage pulse crushing technique was reported to recover >90% of Ag and Cu from Si solar panels.<sup>56</sup> Other techniques like water jet pulverisation<sup>57,58</sup> or laser cutting<sup>59</sup> were reported to “cut” selectively the metal-containing parts from the end-of-life material. However, most of these techniques remain expensive and, so far, can hardly compete with the high-capacity industrial machines available for size-reduction.

Although comminution techniques are well established, it is sometimes possible to liberate valuable metals without shredding or grinding, which often leads to losses and dilution of the TCMs. “Soft” delamination techniques could be more sustainable than non-selective shredding to liberate the valuable metals, for example by weakening hydrogen bonding between current collectors and cathode active materials of LiBs,<sup>60</sup> or deagglomeration of cathode materials from binders and carbon using the high shear forces in a blender.<sup>61</sup> Liberation of the valuable metals from their initial casing is particularly challenging for renewable energy and energy storage materials. For LiBs, short-circuits and management of the potentially hazardous substances (fluorinated lithium salts, organic solvents used as electrolytes) are additional challenges. Preventing fluorinated polymers (like polyvinylidene fluoride



(PVDF) or Nafion<sup>®</sup>) degradation during the recycling process is important for LiBs,<sup>62</sup> fuel cells and the backsheet films of photovoltaic modules. Magnets are encased in organic binders<sup>63</sup> or coated with other metals<sup>64</sup> to prevent corrosion (which is particularly important for offshore wind turbine magnets); removal and separation of this coating is necessary. Solar cells can easily be crushed and sieved, which allows recovery of glass fractions<sup>65</sup> efficiently and separating out polymer-rich (>5 mm) and metal-rich fragments (<0.25 mm) according to particle size.<sup>45</sup> However, the silicon wafers are inevitably not recovered intact, preventing their direct re-use. "Hot-knife" cutting<sup>66</sup> employs elevated temperatures to soften

the EVA while a still blade removes the aluminium frame and the junction box from solar cells, allowing copper cable recovery from the junction box. This technology may allow to reduce losses of metals but EVA and backsheet removal is steel needed.

### 2.3.2 Physical separation of materials

**2.3.2.1 Density separation.** Physical separation methods are commonly used to separate materials using differences in their physical properties (Fig. 3). Density separation is a simple technique that allows the separation of low-density materials such as plastics from higher-density materials like metals. Perhaps the most efficient and obvious example of this is lead acid battery recycling, where the two solid components, lead

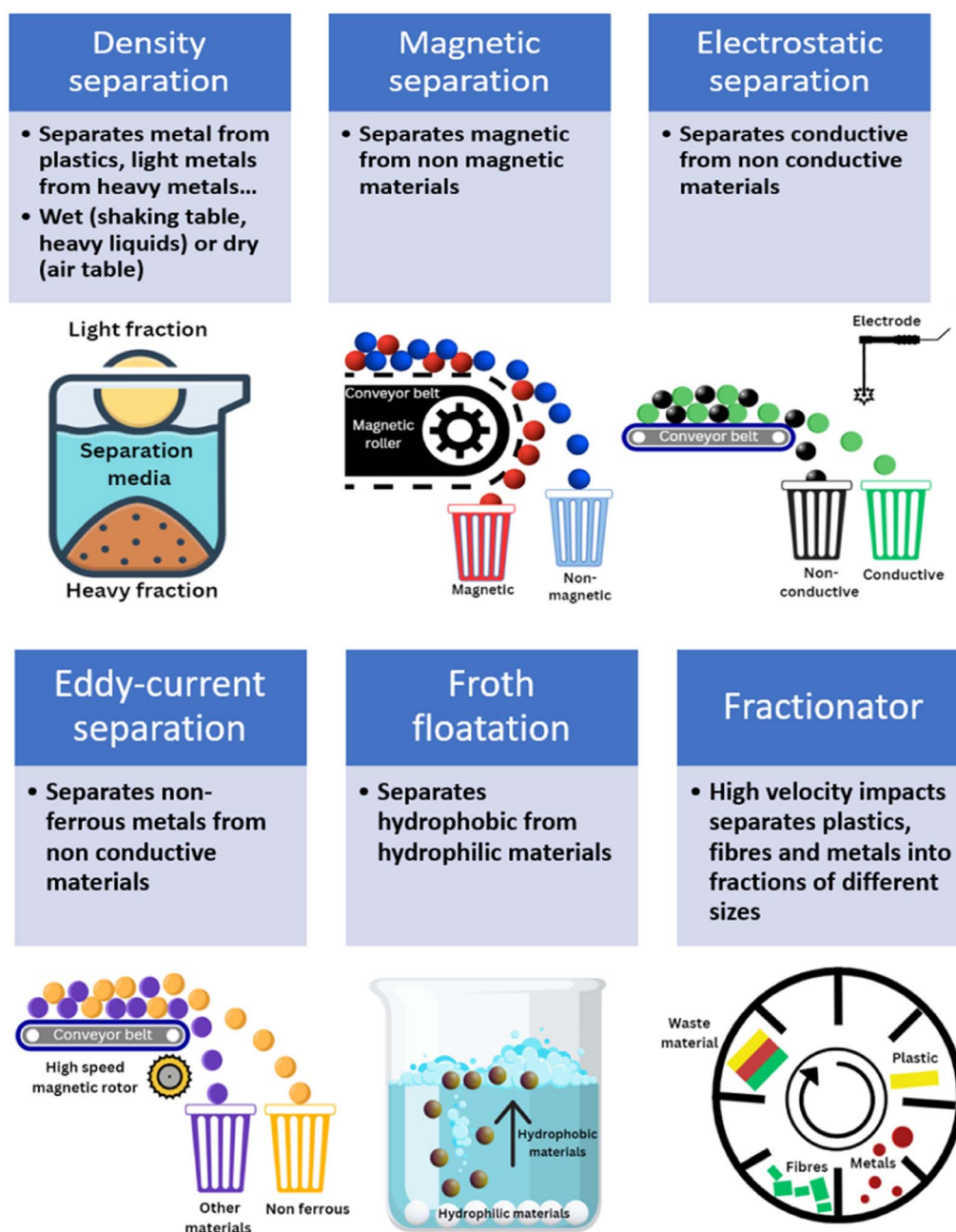


Fig. 3 Physical separation techniques and their application range.



and polypropylene (PP), have very different densities. Various techniques are available to sort materials by density. The feasibility of density separation can be estimated by calculating the concentration criterion<sup>41</sup> (CC), using eqn (1):

$$CC = \frac{\rho_{\text{heavy}} - \rho_{\text{fluid}}}{\rho_{\text{light}} - \rho_{\text{fluid}}} \quad (1)$$

where  $\rho_{\text{heavy}}$  and  $\rho_{\text{light}}$  is the density of the heavy and light material to be separated, respectively, while  $\rho_{\text{fluid}}$  is the density of the media used for separation. Separation is considered easy if CC is higher than 2.5, feasible if CC is below 2.5 but higher than 1.25 and only possible by carefully controlling the density if CC is lower than 1.25.

Air tables are the most common within the recycling industry. The materials are fed in the top of the air table, where air is blown vertically from bottom to top. While the high-density fractions will fall, lighter fractions will be blown away by the air stream and separated. Using a shaker table, materials flow through a liquid medium constantly shaken on a table that can be inclined. This wet separation media allows separation of most metals from plastic, while reducing the risks of dust dissemination; it has also shown some merits for concentrating metals from WEEE.<sup>67</sup> However, it consumes water and has a relatively low capacity (up to 1 ton per day). It is used more commonly for primary extraction than secondary but there is significant potential to expand the use of this technique.

Heavy liquids typically have a density  $> 2 \text{ g cm}^{-3}$  and examples include tetrabromoethane and diiodomethane. When separating materials with heavy liquids, materials with a lower density than the liquid will float at the surface, while heavier materials will sink at the bottom of the liquid. Separation of the different phases is easy and the liquid can be reused. However, the toxic nature of some of the heavy liquids used is detrimental to the application of this process. There is room for the application of this process with less hazardous liquids like sodium polytungstate, which is however expensive. Other liquids like sodium silicate<sup>68</sup> or some brines<sup>69</sup> can reach high densities and can potentially be used as a heavy media for density separation purposes, without the environmental burdens of halogenated solvents. There may be potential to use deep eutectic solvents (DESs) for this application, particularly with metal bromide salts. Both dry<sup>70,71</sup> and wet<sup>72,73</sup> density separation was reported to remove light components from end-of-life LiBs. Water and brines of different densities allowed glass, metal and plastics separation from crushed solar cells.<sup>74</sup> There are significant differences in the densities of materials found in end-of-life materials from clean energy production and storage (displayed in Fig. 1). Those contain high-density metals such as Ag (density<sup>75</sup> =  $10.5 \text{ g cm}^{-3}$ ) and lighter polymers (density<sup>76</sup> of EVA =  $0.95 \text{ g cm}^{-3}$ ). Separation of intermediate-density materials such as aluminium (density<sup>77</sup> =  $2.70 \text{ g cm}^{-3}$ ) is feasible but separating different polymers is more challenging.

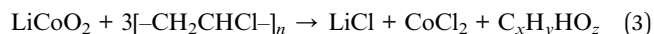
**2.3.2.2 Magnetic, electrostatic and eddy-current separation.** Magnetic separation uses the differences in magnetic susceptibility to separate different metals and materials. The magnetic attraction force<sup>41</sup> ( $F_M$ ) depends on the volume of the particles to be separated ( $V$ ), their magnetic susceptibility ( $\chi$ ), as well as the

magnetic field created by the magnetic separator ( $H$ ) and its gradient ( $\nabla H$ ) as described in eqn (2):

$$F_M = \chi \times V \times \nabla H \quad (2)$$

The process is easy to operate and allows separation of ferromagnetic/paramagnetic materials (attracted to magnets) from diamagnetic materials (slightly repelled from magnets). Separation of ferrous materials can be achieved with small magnetic fields ( $< 2 \text{ T}$ ), while attraction of paramagnetic materials requires stronger magnetic fields<sup>41</sup> (10 to 20 T). Magnetic separation provides excellent separation but there is a risk of agglomeration which must be avoided as it leads to loss of metals. Fine particles will increase the risk of agglomeration and reduce the separation efficiency. Dry, low-intensity magnetic separation is generally applied to coarse and highly-magnetic materials (in the range 0.5–6 mm for drum magnetic separators). Wet magnetic separation tends to be used for smaller fractions ( $< 0.5 \text{ cm}$ ). Size of the feed materials depend on the equipment, with overhead magnets used for ferromagnetic materials as large as 200 mm. Generally, a size reduction step is generally needed regardless of the type of end-of-life materials being considered for recycling.

Application of magnetic separation makes sense within the frame of magnet recycling to separate magnetic materials from other materials. It is also relevant for ferrous materials such as the steel casing<sup>78</sup> found in spent LiBs. The presence of cobalt allows magnetic separation of cathode active materials from organic polymers.<sup>79</sup> It is also feasible to increase the magnetic susceptibility of cathode active materials to facilitate their separation. By grinding waste lithium–cobalt oxide in the presence of waste polyvinyl chloride (PVC), dechlorination of the polymer is accompanied by the formation of lithium chloride and cobalt chloride,<sup>80</sup> according to eqn (3):



The addition of zero valent iron to the waste PVC and  $\text{LiCoO}_2$  improves PVC reductive dehalogenation by abstracting chlorine atoms from PVC. Li is preferentially removed from the oxide materials to form lithium chloride (because of its higher charge density than cobalt and its helium-type double shell), leaving iron and cobalt oxide that can be treated thermally to form magnetic oxides ( $\text{CoFe}_x\text{O}_y$ ), which can be magnetically separated.<sup>81</sup>

Electrostatic separators use the differences in conductivity and resistivity of materials to separate them. Shredded materials (with a size generally lower than 5 mm) are usually sent on a conveyor belt and arrive on a rotating drum connected to the ground. In a roll-type corona electrostatic separator, a high-voltage electrostatic field is applied between this drum and one or several electrodes. The materials to be separated are charged by ion bombardment.<sup>82</sup> While the conductive materials discharge quickly and are ejected away from the rotating drum, the non-conductive materials remain charged and attracted to the rotating drums which allows their separation. The non-conductive particles are pinned to the electrode by the electric





image force ( $F_i$ ) detach from the rotating electrode if the difference between the centrifugal force ( $F_C$ ) reach a certain value defined by eqn (4):

$$F_C - F_i > F_g \cos(\alpha_d) \quad (4)$$

where  $F_g$  is the gravitational force and  $\alpha_d$  is the detachment angle. The electric image force can be calculated with eqn (5):

$$F_i = \frac{Q^2}{4\pi\epsilon_0(2a)^2} \quad (5)$$

where  $\epsilon_0$  is the permittivity of air,  $a$  is the diameter of the particles and  $Q$  their charge. The charge decreases over time but the maximum charge of the particles is defined using eqn (6):

$$Q_{\max} = 4\pi E \epsilon_0 a^2 \frac{3\epsilon_r}{\epsilon_r + 2} \quad (6)$$

where  $E$  is the local electric field and  $\epsilon_r$  the relative permittivity. The separation efficiency depends on the high-voltage applied and configuration of the electrode system, speed of the conveyor belt, ambient conditions, and nature of the materials. Within clean energy and energy storage end-of-life materials, metals are conductive (conductivity  $>10^6 \text{ S m}^{-1}$ ). Intermediate conductivity materials can be found (graphite is in the range<sup>83</sup>  $3 \times 10^2$  to  $3 \times 10^5 \text{ S m}^{-1}$ ) but glass and most polymers have a conductivity lower than  $10^{-10} \text{ S m}^{-1}$ .

While corona electrostatic separators do not require contact between the materials and the electrodes, triboelectric separators charge the materials by contact, leading to materials losing or gaining electrons depending on their position on the triboelectric series. Positively and negatively charged materials can then be attracted to electrodes. Electrostatic separators generally separate metals from plastics due to their different conductivities. The position of a material in the triboelectric series depends on the amount of negative or positive charge it accumulates, which depends on the nature of the materials and other factors such as size and size roughness of the materials. While polymers like PVC and polytetrafluoroethylene (PTFE) are generally negatively charged, other polymers are usually positively charged, which could allow their separation, although the exact position of PP, polyethylene terephthalate (PET) and polyethylene (PE) in the triboelectric series is still debated.<sup>82</sup> Separation of Al and other positively charged polymers from negatively charged PVC and PTFE is possible<sup>84,85</sup> and could be interesting within the frame of end-of-life materials from clean energy and energy storage. These materials must first be liberated from their initial casing, which can be achieved with a size reduction step (particle size for triboelectric separation is in the range 1–10 mm).

Eddy-currents allow separation of non-ferrous materials from other materials. By applying a high-frequency magnetic field with a rotating magnet, induced currents create magnetic fields opposite to the first one; non-ferrous metal will be repelled from the magnet. Ferrous metals must be removed prior to eddy-current separation as their temperature tends to increase when they are submitted to eddy-currents. Eddy current separation is applied to particles with size in the range

3–150 mm, and the highest separability is achieved for materials with high conductivity and low density, and was proposed for the separation of current collectors from cathode materials from spent LiBs,<sup>86</sup> or Al from solar cells.<sup>87</sup>

Magnetic separation techniques are often encountered to separate steel casings and remove any magnetic contamination from spent LiBs.<sup>70,88</sup> Electrostatic separation has a broader range of application and can be used to separate current collectors or polymers from spent LiBs.<sup>89,90</sup> It can also be used to recover 95% of the conductive metals from milled solar cells and separate them from non-conductive fractions (glass and silicon).<sup>91</sup> However, glass tends to accumulate with the conductive fraction and polymer separation is challenging, requiring an additional separation step using density or hydrophobicity differences.

**2.3.2.3 Froth floatation and fractionation.** Within the mining industry, froth floatation is historically used to concentrate metals from low-grade minerals. It allows separation of materials having different hydrophilicity and, as such, has the potential to separate organic layers from end-of-life materials. In froth floatation, hydrophilic particles, wetted by water, stay in the liquid, while more hydrophobic particles will be attached to air bubbles blown in the liquid and can be collected at the surface. Surfactants (called collectors) are used to stabilise air bubbles and enhance the hydrophobicity of target solids through modification of their surface energy. Electrical double layer at the solid/water interface is of primary importance as it controls the absorption of collectors. High surface charge can inhibit the chemisorption of chemically adsorbed collectors, while absorption of physically absorbed collectors depends on the sign and magnitude of the surface charge.<sup>92</sup> Froth floatation is a mature technique that can be used for plastics recycling.<sup>93</sup> Its use for WEEE recycling concentrates precious metals prior to their purification.<sup>94</sup> It has found applications for recovery of cathode materials<sup>95,96</sup> and graphite<sup>97,98</sup> from spent LiBs, to separate aluminium from silicon<sup>99</sup> or recover lead<sup>100</sup> from end-of-life solar cells. Polymers like PVDF are used as binders and therefore firmly attached to the cathode active materials. Thermal treatment is therefore needed to remove it. The residue of the thermal treatment can be removed by ultrasonication prior to flotation of hydrophobic materials.<sup>101,102</sup> Similarly to all physical separation techniques, complete liberation of the materials is a prerequisite to achieve high separation efficiency. Glass fibres were reported to float during a froth floatation process, possibly due to entrainment related to their morphology or presence of residual hydrophobic resin attached to the fibres.<sup>103</sup> Similarly, carbon supports (used as catalysts in fuel cells) will probably float, entraining with them the PGMs attached.

Most physical separation techniques will use one physical property to separate the target fraction from the other components. Fractionator mills will use several of these properties to combine delamination and separation of different fractions. Shredded materials are introduced in a rotating apparatus which submits the materials to high accelerations and decelerations. These high-frequency impacts break the interface of the different layers. While plastics keep their shape, brittle



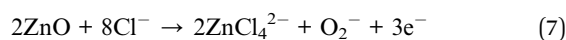
inorganics like fibres are fragmented, and ductile metals aggregate. The output materials have different shapes and sizes, facilitating the subsequent separation of the coarse plastic fraction from the smaller metallic fraction, while precious metals can be found in the finer fractions. Fractionators were proven effective for the processing of conventional WEEE<sup>41</sup> (mostly for the separation of plastics, copper and precious metals from PCBs), their use for renewable energy and energy storage end-of-life materials is yet to be demonstrated. Using high accelerations (similar to fractionators), the use of centrifugation was reported to separate lithium-containing materials from carbon-black of spent LiBs.<sup>104</sup>

Physical techniques are promising for the recycling of emerging end-of-life materials as they are mature and easy to implement on an industrial scale. However, these techniques are used for pre-concentration purposes and the purity of recovered metals is often too low after their application. Moreover, metal losses are inevitable during size reduction and physical separation steps, and emerging waste streams are complex in nature. Breaking metal oxides (the “black mass”) from the cathode of spent LiBs certainly requires hydrometallurgical or pyrometallurgical methods. Physical methods are certainly useful to recover the black mass from the spent vehicle, but other processes are required to recover high-purity lithium carbonate from the black mass.

## 2.4 Chemical methods to process metals

### 2.4.1 Selective dissolution of metals and their oxides.

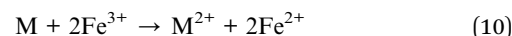
Metals and their oxides are present in many different forms of end-of-life materials, from swarf or pyrometallurgical bullion, to mixed oxides in batteries and catalytic converters. In some cases, the metals are present in a complex arrangement, such as being layered individually onto an unreactive substrate, such as PCBs, fuel cells, solar panels, and thermoelectric devices, or even embedded into a polymer matrix, such as X-ray protective garments. During the recycling process, metals are commonly oxidised in order to form soluble complexes in whichever solvent is being used. Oxidation can be carried out *via* chemical or electrochemical methods, depending on the form and composition of the metals to be recycled. During electrochemical oxidation, solid metals are in contact with the anode (possibly as a slurry “painted” on the electrode<sup>105</sup>), while electrodes are immersed in the solvent. The electrochemical oxidation of metal oxides occurs possibly through formation of superoxide<sup>106</sup> ( $\text{O}_2^-$ ), for example in the case of zinc oxide, as illustrated in eqn (7):



In the meantime, metals dissolved in the solution are electrochemically reduced at the cathode. Anodic dissolution coupled with cathodic reduction of target metals was applied for precious metals recycling from e-waste<sup>107</sup> with DESs as solvents combining complexing ability and electrochemical stability.

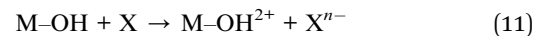
Commonly, chemical dissolution is carried out using conventional hydrometallurgical processes, such as either nitric

acid or hydrogen peroxide as an oxidising agent in combination with sulfuric acid. Oxidation of the target metal (M) with an oxidising agent such as hydrogen peroxide occurs<sup>108</sup> according to eqn (8) and (9), while its oxidation with ferric sulphate can be described with eqn (10):

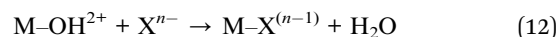


Oxidation with hydrogen peroxide is attractive since the decomposition products are benign, and the oxidising agent doesn't contaminate the metals. However, ferric ions can be regenerated anodically during an electrodeposition process where the target metal is recovered at the cathode, which leads to lower environmental impact than hydrogen peroxide.<sup>42</sup>

Metal oxides are also commonly processed using acidic media, with leaching effectiveness based on a combination of proton concentration and the coordinating ligands available. The proton acts on the oxide moiety, and the ligand acts on the metal ion to weaken the metal-oxide bond sufficiently to achieve dissolution. During metal oxide dissolution, with DESs as solvents, an organic acid (HX) is commonly used as a hydrogen bond donor (HBD). The organic acid is first absorbed on the active OH sites of the hydrated metal,<sup>109</sup> according to eqn (11):



The metal-ligand complex is then formed in solution according to eqn (12), provided that the metal ligand complexes formed are more stable than the metal-OH complex:



Using aqueous solutions, large amounts of acids and/or additives are often required to achieve the desired chemistry and reactivity of the target metals, due to competition with the high concentration (55.5 M) of water ligands. These approaches generally lead to non-selective dissolution of metals, consuming a large amount of chemicals but can be applied to a wide range of feedstocks, and is often used when processing low-grade materials. Hydrometallurgical processing is often carried out after pyrometallurgical processing to recover the individual metals from the bullion. Selectivity is usually achieved through the recovery methods, *e.g.* precipitation, cementation, electrowinning, solvent extraction, or ion exchange. Another option to impart selectivity into a metal dissolution process is to “force” surface passivation onto the less desirable metals. This is a common process that is applied to steels to prevent iron oxide ‘scales’ from forming, but could potentially be expanded to other areas of metal processing. These pickling solutions often involve aqueous solutions made from nitric, sulfuric, or citric acids.<sup>110</sup>

A greener and more sustainable approach would use a benign and catalytic oxidising agent such as iron chloride or iodine, or to tailor the solvent system to impart selectivity in



reactivity, leaching, or solubility. Ionometallurgy uses systems formed of ionic fluids, such as ionic liquids (ILs) or DESs, to process metals. These systems have the benefit of minimal or no water being present, minimising the effect of oxide chemistry.<sup>111</sup> They also have inherently high concentrations of ligands, permitting the chemical oxidation of noble metals such as gold,<sup>112–114</sup> or palladium.<sup>115</sup> Alternatively, the ionic environment can stabilise oxidising species such as iodine<sup>116</sup> or bromine<sup>117</sup> without the need for additional anions that would be required in aqueous media.

In cases where metal oxides are to be dissolved in DESs, the type of HBD present significantly affects the dissolution process, as acidic HBDs provide both the necessary protons to react with the oxide and ligands to coordinate the liberated metal ions. Depending on the coordination ability of the different HBDs, different metal ion complexes are formed,<sup>109</sup> with different stability constants. This can result in either the selective dissolution (or non-dissolution) or selective precipitation of metal salts, as in the case of the processing of LIBs cathode active materials,<sup>118,119</sup> or the recovery of metals from flue dust,<sup>120,121</sup> lamp phosphors,<sup>122</sup> and NdFeB magnets.<sup>123</sup> When using ionic fluids to dissolve metal oxides, one of the critical parameters to metal solvation is the anionic component of the system.<sup>124,125</sup> However, the downsides to these systems is the high viscosity<sup>126–128</sup> and inevitably higher material costs relative to aqueous media. If ILs or DESs are to be used, care must be taken to ensure that the 'green' credentials are not overstated, and that the economic benefits are maximised, either by a decrease in the energy needed for the process, reducing the amount of chemical additives required, or through the processing of small, concentrated amounts of high value materials. Systems containing materials where oxide/hydroxide passivation must be minimised will also benefit from the use of ionic fluids, however the high viscosity of ionic fluids will lead to physical losses which must be factored into the overall sustainability of the process.

A middle ground between aqueous solutions and ionic fluids is concentrated brines, which are formed from water and a high concentration of organic or inorganic salt. The benefit of using a brine over an ionic fluid is that the high ligand concentration is maintained, but at viscosity values closer to aqueous solutions. The presence of water does raise the possibility of oxophilic metals passivating, which may be detrimental to complete metal dissolution. However, in some cases, it is interesting to dissolve a base metal layer, which can then liberate less reactive metals from unreactive organic substrates. For example, thermoelectric devices are assembled by sandwiching semiconductor legs between ceramic plates, using copper connectors and tin-based solders. Tailoring the chloride concentration of the brine allows targeted leaching of the solder, liberating the semiconductor legs, as the other elements present would passivate below a certain chloride concentration.<sup>129</sup>

This selectivity through passivation can also be generated by the addition of water to an ionic fluid containing iodine as the oxidising agent. The metallic portion of PCBs is primarily composed of copper; however multiple layers of different metals

are also present in surface coatings and solder. A challenge exists as different generations of PCBs have used different material combinations; however, the most common currently found in WEEE are hot air solder levelling (HASL), which is a tin/lead alloy and electroless nickel immersion gold (ENIG), which contains nickel and a thin layer of gold.<sup>130</sup> When no water is present, all PCB metals are easily oxidised together, albeit very slowly due to the poor mass transport caused by the high solvent viscosity. When 40 wt% water is present, the rate of gold oxidation is improved due to the decreased solvent viscosity, but the nickel layer passivates and protects the copper underneath from further oxidation for a period of time.<sup>131</sup>

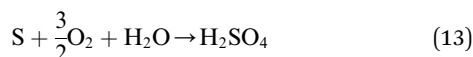
Electrochemical dissolution methods take advantage of the differences in reactivity between the target metals. Metals with cathodic redox potentials, such as copper or iron, are much easier to oxidise than those with anodic redox potentials, such as platinum or gold. In aqueous systems, redox potentials are well-characterised,<sup>132</sup> however by adding a large number of ligands or through the presence of strongly complexing ligands, the reactivities of the metal complexes formed can be altered. At the extreme end of this ligand concentration scale are the ionic fluids, where the change in chemistry can lead to increased stability of metal ions in solution with respect to the reduced form of the metal. For example, in a system formed from choline chloride and ethylene glycol,  $\text{Cu}^+$ ,  $\text{Ag}^+$ , and  $\text{Au}^+$  form anionic chloride complexes due to the high chloride environment,<sup>133</sup> which means that the redox potentials can be up to 1.0 V more cathodic in the ionic fluid compared to in aqueous medium,<sup>134</sup> allowing a greater range of metals to be accessible under relatively mild conditions. However, selective electrochemical dissolution is generally not employed in the literature, with the selectivity focus being on the metal recovery step, which is often a chemical method such as precipitation. The bulk of the literature focuses on electropolishing, so this particular "tool" is underutilised and further research must be carried out before it can be applied to complex metal-containing systems.

Importantly, the solvent selected must match the process with regards to material cost of reagents and value of product. The main challenge to selectivity will be to favour oxidation or passivation selectively with cheap, environmentally friendly, and readily available chemicals. Additionally, one must consider the composition of the material when designing the process; if the most valuable metal is embedded in another matrix, then the whole matrix must be dissolved, or some form of comminution must be employed. If the most valuable metal is on the surface of the material, such as with PCBs, then minimising dissolution of the underlayers is important to reduce chemical consumption. Alternatively, the more reactive metals could be oxidised to undercut the valuable and unreactive metals, causing them to slough off and be recovered by simple filtration.

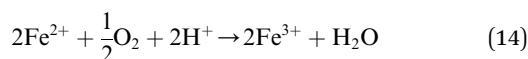
**2.4.2 Biometallurgical processing of metals.** Recycling and purification of metals can be achieved *via* the interaction of microorganisms with metals, the field of these biotechnological processes being called biometallurgy. Dissolution of metals is feasible using bacteria, based on three mechanisms:



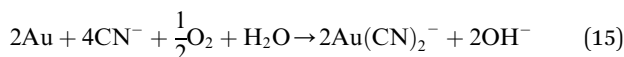
complexolysis, redoxolysis, and acidolysis.<sup>135</sup> Acidolysis involves chemolithoautotrophic bacteria and fungi able to produce organic and inorganic acids, which in turn can protonate oxygen atoms at the surface of metals and solubilise them. The same microorganisms are also involved in redoxolysis mechanisms, where electron transfer provides energy for microbial growth. These reactions are mostly used for the microbial oxidation<sup>136</sup> of sulphur to produce sulphuric acid, as described in eqn (13):



It is also possible to oxidise metallic iron ( $\text{Fe}^{(0)}$ ) and ferrous ion ( $\text{Fe}^{2+}$ ) into ferric ion ( $\text{Fe}^{3+}$ ) using microorganisms, as described in eqn (14).



Sulphuric acid is then able to dissolve metals *via* the acidic mechanisms already described, while ferric ion is able to oxidise metals. Complexolysis is possible with fungi and cyanogenic bacteria. It mainly involves the decarboxylation of glycine to produce cyanide ions, which are then able to form complexes with precious metals like gold. These complexes are then oxidised with oxygen<sup>137</sup> according to eqn (15):



As opposed to conventional cyanide leaching, some bacteria are able to turn toxic cyanides into  $\beta$ -cyanoalanine, which facilitates wastewater treatment. Gold recovery from WEEE was extensively studied using cyanogenic bacteria such as *C. violaceum*,<sup>138,139</sup> *B. megaterium*,<sup>140</sup> or *P. fluorescens*.<sup>141</sup>

The leaching of metals from end-of-life LiBs is feasible by reduction of  $\text{Co}^{3+}$  (within the insoluble  $\text{Co}^{3+}$  oxide) into  $\text{Co}^{2+}$ , which can be achieved using cheap pyrite ( $\text{FeS}_2$ ) and sulphur. Both chemicals will be oxidised with oxidising bacteria to produce sulphuric acid and  $\text{Fe}^{3+}$  ions. The subsequent reaction of  $\text{Fe}^{3+}$  ions with pyrite releases  $\text{Fe}^{2+}$  ions which in turn allow formation of soluble sulphate salts of Co and Li.<sup>136</sup> Similarly, lanthanide elements (Nd, Pr, Dy) can be leached from end-of-life magnets<sup>142</sup> using *L. ferrooxidans* bacteria, sulphur, and iron. High leaching rates ( $\geq 90\%$  for Dy, Nd and Pr) can be obtained in 14 days, ferric ions playing a major role in the leaching efficiency. Multiple studies reported the leaching of PGMs<sup>143</sup> from catalysts using bacteria. PGMs having a high redox potential, their leaching is feasible by cyanide leaching,<sup>144</sup> similarly to gold leaching depicted in eqn (15). Other strains like *Aspergillus niger*<sup>145</sup> can produce oxalic acid and favour Pt leaching, but the recovery rates remain low (below 40%). Gallium and arsenic leaching from thin film solar cells containing gallium arsenide (GaAs) is feasible using bacteria which can produce organic acids (including amino-acids), which helps solubilise gallium and arsenic in 20–30 days.<sup>146</sup>

Most of the dissolution reactions involved require nutrients, careful control of the pH and bubbling of compressed air to provide oxygen and  $\text{CO}_2$  for microbial growth. These experimental conditions will therefore contribute to significant consumption of chemicals for the leaching process. However, comparison of techno economic assessments between bio-metallurgical and hydrometallurgical processes indicate that the former could be cheaper than the latter for the recovery of metals from end-of-life PCBs<sup>147</sup> (0.159 € per kg for bio-metallurgy, 0.224 € per kg for hydrometallurgy). This process can be made even more economical and sustainable when waste materials are used as nutrients.

Some technological issues need to be addressed for bio-metallurgical processes, the main ones being potential formation of toxic chemicals like hydrogen cyanide, the presence of organic compounds and heavy metals detrimental to bacterial growth, and precipitation of iron species through iron hydrolysis or jarosite which decreases the leaching efficiency. Bio-metallurgical leaching suffers from slow kinetics; it typically requires several days to reach significant leaching rates. Different strategies were developed to accelerate the leaching kinetics such as the application of a voltage<sup>148</sup> to improve electron transfer reactions, using a weak magnetic field,<sup>149</sup> ultrasounds,<sup>150</sup> or ultraviolet irradiation<sup>151</sup> to induce mutations on the bacteria, this last approach being the only one so far to provide a clear improvement of the leaching kinetics. A more promising approach is the addition of a catalytic agent like activated charcoal,<sup>152</sup> or silver.<sup>153</sup> In that case too, improvement of kinetics is limited and addition of catalysts could have a negative effect on the growth of microorganisms.

Bacteria can also be used to precipitate or separate metals which have been previously dissolved by bio- or hydrometallurgical means. Multiple bacteria, fungi, plants and extracts are able to precipitate metals through reduction reactions.<sup>154</sup> This approach is promising to up-cycle base metals into valuable nanoparticles, or to produce PGM-based catalysts from dissolved PGM solutions,<sup>155,156</sup> which can improve the performance of the catalysts as compared to commercial ones.<sup>157,158</sup> Bacteria can also be used to produce biomolecules useful for purification of metals. Proteins like lanmodulin have a very high affinity for lanthanides.<sup>159</sup> Hence, lanmodulin can be obtained from bacteria, purified, and used for the selective biosorption<sup>160</sup> of REEs from WEEE or minerals, eliminating the need for solvents or ion-exchange resins.

Natural molecules like reducing organic acids, sugars or antioxidants can be easily obtained from waste fruit peel,<sup>161</sup> waste tea<sup>162</sup> or grape seed.<sup>163</sup> These molecules were shown to be as efficient as synthetic reducing agents (like hydrogen peroxide) for the dissolution of metals from LiBs cathodes. Hence, microorganisms could be used indirectly through metabolic processes such as fermentation to produce useful chemicals able to solubilise metals. Within this frame, food waste can be considered as a sustainable and cheap source of chemicals for recycling processes.

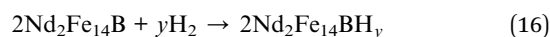
Overall, biological processes are cheaper and make sense for large volumes of low metal content materials where processing time is less of an issue. Low grade ores such as mine tailings are





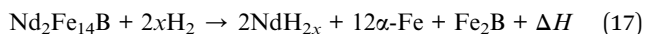
particularly suited as they represent a high volume and could contain small but significant amounts of critical metals. For the same reasons, low-grade WEEE falls into this category. The biohydrometallurgical purification and recovery of metals from leach liquors is less hampered by slow kinetics and looks promising for reducing chemical consumption during the metal recovery stages. While increasingly used for primary extraction, application to secondary extraction is only in its very early stages of research.

**2.4.3 Hydrogen decrepitation.** Magnetic materials rely heavily on REEs. While samarium–cobalt magnets are preferred for high temperature applications, NdFeB-type magnets are mainly used in wind turbines, electric vehicles and electronics such as hard drive disks (HDDs), accounting for the majority of Nd, Pr and Dy consumption. Within these products, magnets are often encased within complex architectures. Submitting the complete equipment to a flux of hydrogen (under moderate pressure of 1 atmosphere) decrepitates the magnet,<sup>164</sup> while other materials are barely affected by the process. Decrepitation is caused by volume expansion of metal lattices caused by formation of solid solution and/or hydrides.<sup>165</sup> Within magnets, Nd rich regions and Nd<sub>2</sub>Fe<sub>14</sub>B matrix phases absorb hydrogen near room temperature according to eqn (16):



During hydrogen decrepitation (in the temperature range 25–400 °C), formation of Nd-hydrides and hydrogen diffusion causes expansion of Nd rich regions and cracks within the materials, finally resulting in a demagnetised powder with a size in the range 6–600 μm.<sup>166</sup>

The hydrogenation–disproportionation–desorption–recombination differs by the higher operation temperature used (in the range 750–950 °C), and the smaller particle size of the powder obtained (down to 0.3 μm (ref. 167 and 168)). In that case, further hydrogenation of Nd<sub>2</sub>Fe<sub>14</sub>B leads to its disproportionation into NdH<sub>2</sub>, Fe<sub>2</sub>B and Fe, according to eqn (17):



Desorption of hydrogen takes place under vacuum at higher temperatures (150 °C for NdFeB phases, up to 700 °C for Nd rich regions). Even higher temperatures also allow recombination of decomposed phases into the original Nd<sub>2</sub>Fe<sub>14</sub>B. The obtained product is a powder which is easily separated from the initial casing of the magnet and can then be turned into new magnets by sintering, hot pressing, or pelletising followed by melting and casting.<sup>169</sup> Vacuum sintering can take the place of the degassing step. Removal of surface oxides and reduction of oxygen content may be needed since a high oxygen content reduces the sinterability and the magnetic properties of the final material.

The size of the powder obtained depends on the temperature of the hydrogen decrepitation process with room temperatures leading to fine powders (<250 μm) with some larger particles (>500 μm). Hotter hydrogen has a lower solubility in Nd<sub>2</sub>Fe<sub>14</sub>B, the lower volume expansion generates larger particles when the

process is carried out at 150 °C.<sup>164</sup> Milling is sometimes needed in order to reduce particle size and favour densification of the magnet.

When applied to magnets from HDDs<sup>169</sup> (coated with Ni or Ni and Cu), hydrogen decrepitation produces a fine, demagnetised powder containing REEs, while metals from the coating can be sieved and separated from the powder. But depending on the nature of the coating and its thickness, pre-processing of the magnets may be needed to allow hydrogen to access these Nd phases and decrepitate them.

As compared to production of magnets from virgin materials, comparative life cycle assessment (LCA) clearly shows a much lower footprint of the hydrogen decrepitation process.<sup>170</sup> However, it should be mentioned that applying a nickel coating on the magnet accounts for a significant proportion of the overall environmental footprint, while shredding of the materials leads to massive losses of neodymium (90%). The decrepitation process is so far limited to magnets, but can in theory be applied to other intermetallic compounds for which volume expansion following hydrogen absorption is accompanied by powder formation<sup>165</sup> (e.g. LaNi<sub>5</sub>, SmFe<sub>3</sub>, TiFe, SmCo<sub>5</sub>, NdCo<sub>5</sub>, PrCo<sub>5</sub>, LaCo<sub>5</sub>). Other metals show a large volume expansion under hydrogen but either form coarse powders (e.g. Ti, Zr) or keep their integrity (Pd).

## 2.5 Methods to process inorganic and organic fractions

**2.5.1 Breaking brittle inorganics and delaminating with ultrasound.** The perturbation of ultrasonic waves through a solution leads to the formation of agitated microbubbles in solution. This process is known as cavitation. The bubbles compress and shrink in cycles, until reaching a critical size where they undergo a violent collapse. This phenomenon is called cavitation,<sup>171</sup> and according to the ‘hot spot theory’, it leads to micro-environments where temperature and pressure could reach several thousands degrees and hundreds of atmospheres, respectively.<sup>172</sup> The effects of cavitation in bulk solution can be attributed to electric charge at the surface of the bubble which creates a large electric field when the bubble collapses. This symmetrical collapse causes shockwaves to propagate through the solution, inducing turbulent flow in the solution, and acting to remove surface films at solid–liquid interfaces. These shockwaves have been shown to be important during material delamination.<sup>173</sup> If cavitation occurs in close proximity to a rigid surface however, the collapse will occur asymmetrically, sending forceful jets into the surface.<sup>174</sup> Jetting phenomena have been shown to be important in the removal of surface passivating layers as well as causing micron sized pitting in the surface of metals.<sup>175</sup>

Ultrasound was first used to improve the leaching rate of lithium into water from spent cathodes following reductive roasting,<sup>176</sup> or leaching of the same metals with organic acids<sup>177</sup> and hydrogen peroxide. Cavitation was found to improve flotation,<sup>178</sup> comminution,<sup>179</sup> and leaching of metals from end-of-life PCBs. Leaching of indium and tin from layered materials like liquid crystal displays (LCDs) was improved using either sulfuric<sup>180</sup> or hydrochloric<sup>181</sup> acid in combination with



ultrasound. Application of ultrasound improved precipitation of lithium carbonate by accelerating nucleation and reducing polymerisation of lithium carbonate crystals.<sup>182</sup> Leaching rates and/or kinetics of metal dissolution are clearly improved with ultrasound<sup>183</sup> in most cases, although it remains unclear if this improvement is of thermal, mechanical, or sonochemical origin. Energy consumption is an obvious limitation during the prolonged use of ultrasonic processes, particularly with high-power, low-frequency ultrasound.

The mechanical effect of ultrasound is particularly useful to break brittle inorganic materials. One could use ultrasound to pulverise the brittle semiconductor legs from thermoelectric materials.<sup>129</sup> In that case, water is used as a propagation media for the ultrasound. The process does not produce wastewater, as no metals are dissolved in the propagation media. Contrary to conventional size reduction methods, ultrasound only reduces the size of the most brittle materials (avoiding mixing of highly-brittle semiconductors, brittle ceramics and ductile metals<sup>184</sup>), which reduces the risk of diluting valuable materials. Hence, the number of steps required for metal purification are lower, while it also reduces the risk of hazardous dust formation.

Ultrasound can also be used as a powerful delamination method. Comparative techno-economic analysis of different processes showed that shredding of end-of-life LiBs was less economically attractive than disassembly followed by delamination of the cathode active materials<sup>34</sup> with ultrasound. Delamination is feasible due to the high acoustic pressure induced by cavitation. Using either citric acid or ethylene glycol as wetting agents (which also help the collapse of bubbles), cathode and anode active materials are separated in a few seconds from their current collectors.<sup>185</sup> This is a clear advantage, as shredding and leaching of both current collectors and cathodes require additional chemicals and further separation steps to remove aluminium. Removal of lithium-iron phosphate materials from aluminium current collectors was demonstrated using ultrasound,<sup>186</sup> which proves that the technique works independently of the nature of the active materials. Interestingly, ultrasound-assisted Fenton oxidation forms hydroxyl radicals, which can possibly degrade the PVDF binder and help recover the cathode materials. Ultrasonic delamination works even better when the binder is partially dissolved, but this objective should not be achieved with toxic or costly solvents like *N*-methylpyrrolidone<sup>187</sup> (NMP). This ultrasonic delamination approach could be helpful for multi-layered end-of-life materials such as fuel cells.<sup>188</sup> Application of ultrasonics to solar cells immersed in various solvents like acetic acid, formic acid, butanone and benzyl alcohol,<sup>189</sup> allowed removal of polymeric backsheet.

**2.5.2 Dissolving organics.** Organics often represent a major fraction of end-of-life materials, and can be found under different forms such as polymers to provide mechanical support, protect and insulate (EVA for solar cells, epoxy resins for bonded magnets and composites), binders or membrane between electrodes (PVDF in LiBs, Nafion® in fuel cells). Spent LiBs and fuel cells contain well-known polymers like PET (gaskets of fuel cells), PE or PP (used as separator between LiBs electrodes), for which recycling schemes are well established,

although they represent a much lower volume than used bottles for example, and may be contaminated with hazardous substances. Removal of organics is a prerequisite of most recycling processes, but management of fluorinated polymers, epoxies, or EVA increases complexity for these specific end-of-life materials. Organics are generally ductile but their dissolution isn't as straightforward as for metals. Some polymers are however soluble in organic solvents, which allows the design of circular processes where the polymer is dissolved and the solvent regenerated by distillation (such process is schematised in Fig. 4(a)). Alternatively, solvents can be used to break the polymers into monomers, possibly leading to value-added chemicals.

Different polymers such as PVC or PET were recycled at the industrial scale by dissolution with organic solvents.<sup>190</sup> Care must be taken to avoid hazardous solvents, and select solvents with a low impact on health and the environment.<sup>191</sup> Polymer solubility in different solvents can also be predicted up to a certain extent by using the Hansen and Hildebrand solubility parameters.<sup>192,193</sup> Evolution of regulation must also be taken into account. The ban on the use of certain plasticizers such as bis(2-ethylhexyl) phthalate made PVC recycling more complicated, as it involves dissolving a banned substance that requires specific authorisation to use.<sup>194</sup> In the VinyLoop® process, a butanone-hexane-water mixture was used to dissolve PVC, while the azeotrope formed was regenerated by distillation.<sup>190</sup> PET recycling is generally achieved by hydrolysis or glycolysis. PET dissolution from waste textile was achieved at the pilot scale by Worn Again.<sup>190</sup> Multiple solvents can be used to dissolve PET (such as aromatic esters or aldehydes), benzyl acetate and ethyl benzoate probably being the best option for health and safety reasons.

Within renewable energy and energy storage end-of-life materials, the most challenging organics are Nafion®, PVDF, EVA and epoxy resins (their structure is shown in Fig. 4(b)).

Nafion® is a sulfonated fluoropolymer (having a PTFE backbone), which is used to conduct cations in PEM fuel cells. This membrane is coated with a catalyst, often made of carbon black particles coated with platinum group metals. Catalyst recovery is possible by dissolution of the Nafion® polymer, which can be achieved with water/alcohol mixtures<sup>195</sup> at elevated temperatures (100 °C) but requires working in an autoclave. However, this approach does not take into account the flammability<sup>196</sup> of some catalysts (in particular dry and finely divided platinum group metals) which should not be combined with flammable solvents. Moreover, it is possible to dissolve platinum from the catalysts with hot sulfuric acid<sup>197</sup> or hydrochloric acid<sup>198</sup> and hydrogen peroxide, which may degrade the membrane (particularly the sulfonate groups) but not dissolve it. It is also possible to detach both the hydrophobic catalyst from the membrane and the Pt from the carbon particles by using hydrophobic ILs. Amongst the ILs investigated, the commercial trihexyltetradecyl phosphonium chloride IL allowed high recovery of Pt (>90% after 6 hours at 120 °C) through specific interactions, swelling and wettability of the membrane, the ionomer and the carbon particles composing the catalyst layers of fuel cells.<sup>199,200</sup>



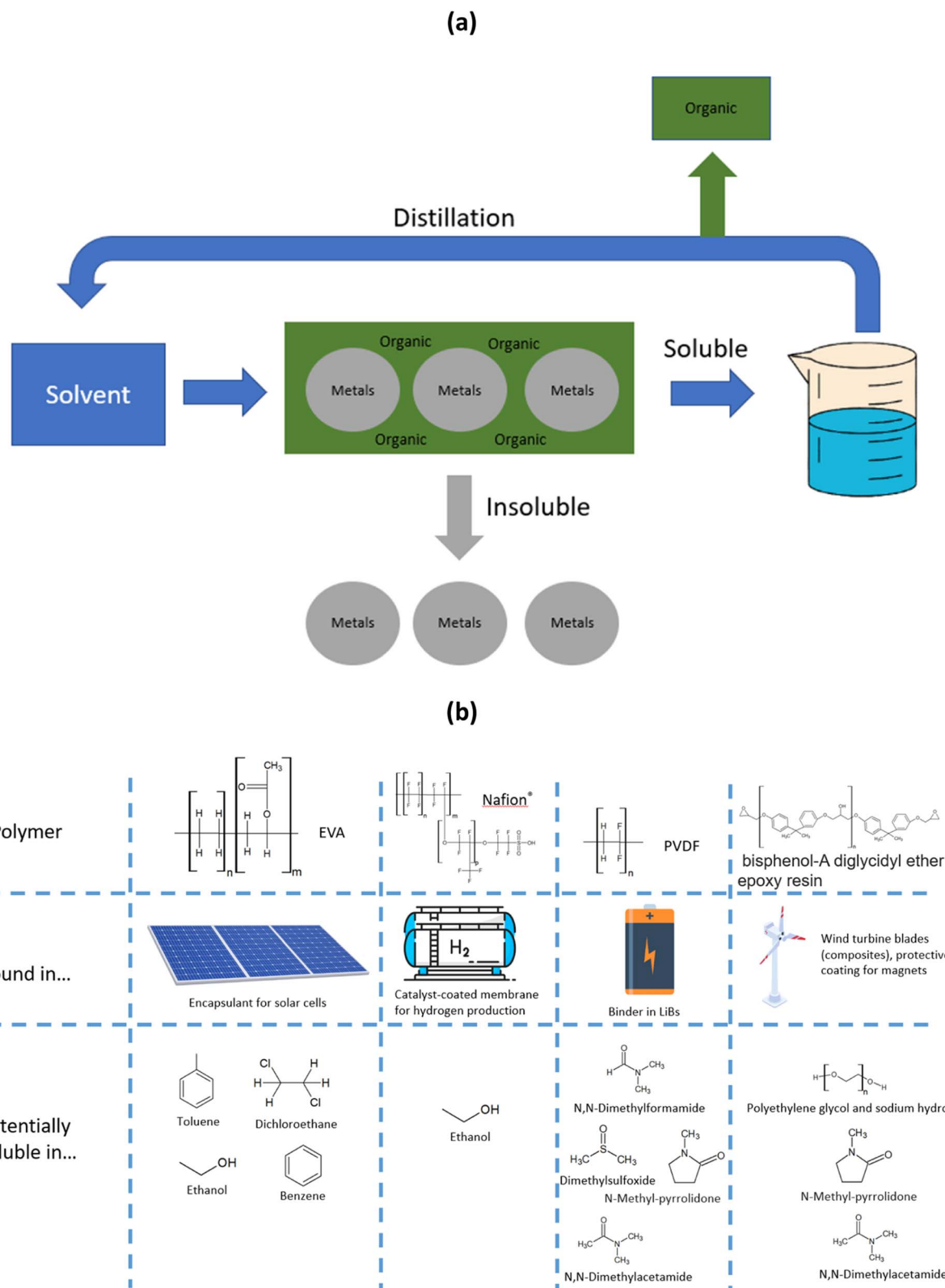


Fig. 4 (a) Scheme of organics dissolution process within the frame of metal recycling, and (b) structure and potential solvents for polymers found in end-of-life materials.

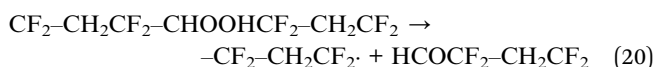
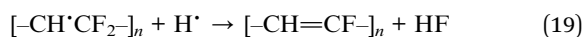
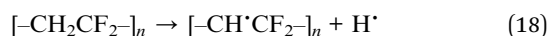
PVDF is often used as a binder between the current collectors and the cathode active materials of LiBs. Its dissolution has been proven effective to liberate the black mass and separate it

from the current collectors. Dissolution can be achieved in 20 minutes at 190 °C using waste cooking oil.<sup>201</sup> The fatty acid methyl esters are good solvents for PVDF but they must be



modified by transesterification reactions with methanol and sodium hydroxide as a catalyst. However, the process temperature exceeds the melting point of PVDF (around 160 °C). PVDF could also be dissolved in a range of polar solvents<sup>202</sup> like *N,N*-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), NMP, or dimethylacetamide (DMAc). The aforementioned solvents cause problems due to their toxicity, with the exception of DMSO, which is considered safer, but is not innocuous.<sup>203</sup> Sustainable alternatives like dihydrolevoglucosenone<sup>204</sup> (Cyrene<sup>®</sup>) have been identified as promising solvents. In that case, large scale availability and cost of the solvent could be an issue. Use of sustainable solvents like gamma-valerolactone for PET recycling by dissolution was demonstrated.<sup>205</sup> Both economics and environmental metrics of the process depend on the solvent used but also on the polymer recovery and solvent regeneration method (distillation, anti-solvent precipitation...).

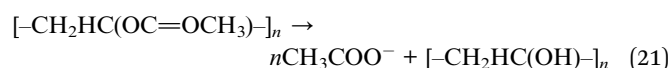
Managing waste fluorinated polymers is complex,<sup>206</sup> and their degradation into toxic fluorinated perfluoroalkylated substances is of particular concern. Mineralisation of PVDF (and other fluorinated polymers), where it is decomposed into fluorine ions and carbon dioxide, can be achieved with supercritical water at elevated temperatures (250 °C or more) using either oxygen,<sup>207</sup> hydrogen peroxide,<sup>208</sup> or potassium permanganate<sup>209</sup> as oxidising agents. Degradation of the PVDF starts by hydrogen abstraction from the polymer backbone (eqn (18)), which leads to the formation of a C=C double bond and the release of HF (eqn (19)). The presence of potassium permanganate promotes hydrogen abstraction, while its reaction with water leads to hydroperoxide formation ( $[-\text{CHOOHCF}_2-]_n$ ), which in turns leads to backbone scission (eqn (20)):



The acid fluoride is easily hydrolysed, subsequent oxidation of end groups and carbon-carbon bond cleavage leads to reduction of PVDF backbone and release of fluoride ions and carbon dioxide.

EVA removal is necessary to liberate the solar cells which contain most of the value (silver electrodes, silicon wafers). Solar cells are completely encapsulated in EVA which protects the wafer from the environment while ensuring mechanical support. EVA dissolution is feasible in a wide range of organic solvents such as acetone, ethanol, toluene, and dichloroethane.<sup>210</sup> However, complete dissolution seems too long to be implemented at industrial scale (7 days at 80 °C with most solvents), and initial EVA swelling breaks the wafers, making it impossible to re-use them directly. Process can be made faster (30 minutes at 70 °C) with benzene or toluene (3 mol dm<sup>-3</sup>) diluted in ethanol as a solvent, and using high intensity ultrasonication<sup>211</sup> (900 W). It also allows recovery of the wafer without cracks, as long as the solvent is not degraded. But degradation of the solvent may occur due to the high

temperatures reached during ultrasonic irradiation and the formation of radicals during the collapse of bubbles formed under ultrasound. Therefore, clear investigation of the solvent degradation is needed, as well as a clear comparison of electricity consumption, environmental impact and overall cost of the process compared to other EVA removal methods like laser irradiation<sup>212</sup> or pyrolysis<sup>213</sup> (both techniques allowing recovery of the wafer without breaking it). For all these reasons, delamination of solar cells with gas-expanded liquids<sup>214,215</sup> seems more appropriate than dissolution of EVA, although it does not allow intact recovery of wafers. Solar cell delamination is also possible with KOH in ethanol, this solvent allowing a better wettability and ensuring penetration of the hydroxides ions.<sup>216</sup> Corrosion of the glass with the same hydroxide ions favours their diffusion between the layers, while EVA is hydrolysed to yield acetic acid and polypropylene, according to eqn (21), separating the glass from the silicon wafer.



Composite materials are made from the interpenetration of a matrix material (such as epoxy resins), reinforced with fibre-glass or carbon fibres. Their lightweight and mechanical properties made them a material of choice for PCBs or wind turbine blades. Epoxy resins are one of the biggest challenges of wind turbine recycling, with an estimation of around 225 000 tons a year of fibre-reinforced polymers from wind turbine blades reaching their end-of-life.<sup>217</sup> This is also relevant to metal recycling, as magnets are often coated with epoxy resins.<sup>218,219</sup> Multiple attempts have been made to break the structure of the epoxy and to recover valuable monomers from the epoxy without decreasing the mechanical performance of the embedded fibres. Epoxy can be decomposed by oxidation with hydrogen peroxide in acetone, which takes 30 minutes at 60 °C and allows to recover clean carbon fibres<sup>220</sup> with high mechanical strength. The degradation products formed (which include explosives) are however problematic. Supercritical water and gas-expanded liquids<sup>221</sup> can degrade epoxy (probably through hydrolysis), but they could also lead to toxic degradation products like hydrogen sulphide. Epoxy resins can be degraded by a variety of solvents associated with catalysts like polyethylene glycol and sodium hydroxide, which allow ester hydrolysis in less than an hour, at 180 °C and atmospheric pressure.<sup>222</sup> Benzyl alcohol and K<sub>2</sub>CO<sub>3</sub> allow depolymerisation of epoxy resin at ambient pressure,<sup>223</sup> but at temperatures of 180 °C for ten hours. ZnCl<sub>2</sub> in water will preferentially cleave the carbon-nitrogen bonds,<sup>224</sup> which would take 6 hours at 250 °C. Concentrated nitric acid (4 M) at 80 °C for 100 hours, allows carbon nitrogen cleavage and nitration of the benzene ring.<sup>225</sup> Other solvents like monoethanolamine lead to solid degradation products.<sup>226</sup>

Brominated epoxy resins from WEEE were dissolved at around 100 °C in NMP<sup>227</sup> and DMSO, higher temperatures with DMF,<sup>228</sup> or mixtures of propanol and heptane,<sup>229</sup> microwaves helping to achieve better extraction rates. Similarly, subcritical acetic acid<sup>230</sup> (220 °C) can be used to swell and ultimately





decompose brominated epoxy resins. Some ILs such as 1-butyl-3-methyl-imidazolium acetate in combination with ethylene glycol showed promising results for the dissolution of epoxy resins thermosets.<sup>231</sup> Other ILs like tributyl ethyl phosphonium diethyl phosphate were unable to dissolve epoxy resins, but were efficient for polymer removal (polyamides, poly-*p*-phenylene sulfide) from bonded magnets.<sup>63</sup>

Chemical dissolution of organics could be a powerful separation technique, but only if a non-toxic, recyclable and cheap solvent is available in large quantities to dissolve the polymer under mild conditions, and does not produce harmful by-products. In this regard, dissolution of PVC and some fluorinated polymers seems favourable to avoid formation of gaseous HCl and fluorinated compounds, respectively, when these polymers are submitted to high-temperatures. PVC swelling or dissolution seems particularly indicated to recover copper from PVC-coated copper cables.<sup>232</sup> Dissolution of brominated flame retardants for decontamination purposes seems also relevant. For epoxy resins, use of solvents and high temperatures for a prolonged time leads to comparable, if not higher, environmental impact of dissolution as compared to thermal treatment.<sup>233</sup>

**2.5.3 Pyrolysing and gasifying organics.** Depending on the organic considered, pyrolysis and gasification could be a useful alternative to dissolution. As opposed to incineration, pyrolysis heats (at 500–550 °C in most industrial plants<sup>234</sup>) and decomposes organic materials under an inert atmosphere, creating syngas, liquid tar, and solid char. Gasification heats organic materials at 500–900 °C using an air atmosphere or 1000–1600 °C using pure oxygen or oxygen-enriched steam,<sup>235</sup> which aims to produce high-calorific value gases as syngas (a tar and solid residue are also inevitably produced). For the management of WEEE plastics, pyrolysis was shown to have a lower environmental impact than incineration.<sup>236</sup> Although a detailed comparative LCA is needed for each end-of-life product, re-use of the degradation products (potentially as a fuel for the process) is key for both pyrolysis and gasification. For municipal waste, gasification was shown to have the lowest impact, as compared to incineration and pyrolysis. Incineration was shown to have a lower global warming impact than pyrolysis due to a more efficient use of heat.<sup>237</sup> Efficient modern flue gas cleaning methods (including electrostatic precipitator and acid gas neutralisation) and ash recycling help reduce the impacts of incineration, which however remain significantly higher than pyrolysis and gasification in terms of impacts of acidification, terrestrial eutrophication and photochemical ozone formation (notably due to the contribution of acidic gases NO<sub>x</sub>, SO<sub>2</sub>, HCl and HF).

Pyrolysis of fluorinated polymers leads to fluorinated degradation products which are difficult to handle. Acid scrubbing is needed to deal with potential HF formation, while high temperatures are needed to ensure complete destruction of CF<sub>4</sub>, which takes 1 second at 1440 °C.<sup>238</sup> So far, incineration remains the only large-scale solution to ensure complete degradation of fluorinated polymers. Irradiation can also be used to degrade such polymers, which can also be mechanically reprocessed or chemically turned into their monomers.<sup>206</sup>

Physical processes can be applied to remove fluorinated backsheet from solar cells prior to thermal treatment. This can be achieved using milling machines made for cutting or engraving, which are able to remove the backsheet through superficial abrasion.<sup>239</sup>

Pyrolysis under nitrogen atmosphere of EVA and PET from solar cells leads to complete conversion of the organics in 30 minutes at 500 °C.<sup>240</sup> Liquid decomposition products were mostly long-chain olefins and alkanes (like 1-tridecene, 1-nonadecene, 1-tricosene, and 1-tetradecene), while gaseous products were short-chain olefins and alkanes (like ethane, butane and propane). Pyrolysis possibly allows recuperation of intact silicon wafers,<sup>241</sup> which is an additional advantage, but attention must be taken to hazardous and volatile degradation products when dealing with gallium-arsenide solar cells, or the fluorinated polymers from the back sheet of most solar cells.<sup>242</sup> Gasification of EVA from solar cells was used to produce cedrene, a medical intermediary used for cancer treatment.<sup>243</sup> However, the feedstock used in most studies was shredded solar cells; intact recovery of wafer was demonstrated with pyrolysis, not with gasification.

For epoxy resins, two degradation pathways were identified depending on the temperature of the pyrolysis process. Degradation products will be mostly composed of olefins, alkanes, aromatic compounds, and carboxylic acids.<sup>244</sup> The pyrolysis process could be catalytic<sup>245</sup> (to break bonds selectively), oxidative,<sup>246</sup> or based on superheated steam.<sup>247</sup> The degradation temperature and time needed will depend on the atmosphere used, but superheated steam advantageously removes the char produced during pyrolysis, which decontaminates the carbon or glass fibres from composite materials. Another important parameter is the mechanical resistance of the recovered fibres, which is often reduced after thermal treatment. Gasification of epoxies in supercritical water was showed to be promising due to production of OH<sup>•</sup> radicals which improve ring opening and C–O bond rupture.<sup>248</sup>

Pyrolysis and gasification seems to be more indicated if the degradation products are not harmful, and if there are very large volumes of end-of-life materials to deal with. Both pyrolysis and gasification have been hindered by the use of heterogeneous feedstock. End-of-life solar cells for example could offer a relatively homogeneous feedstock for these processes. So far, high-temperature processes and mechanical shredding are the only relevant large-scale options available for wind turbine blades. Pyrolysis and gasification produces syngas and tars as products which can be used to fuel the process, produce heat, electricity or other chemicals, which is key to reduce their environmental impact. Offshore wind turbines could be pyrolysed near ports and produce valuable chemicals for the chemical industry, often located nearby. Recycling rates, mechanical properties and value of the recovered products must inform techno-economic and life cycle assessments to select which process can be used to deal with the organic fractions. It is however necessary to find relevant options to treat the residue of these processes, like fibreglass.

**2.5.4 Valorising low-value fractions.** After physical treatment of end-of-life materials (comminution and physical



separation) or high-temperature processes, high value metals are liberated and separated from low-value fractions (various polymers, graphite from LiBs, fibreglass from composites). These low-value fractions represent a large proportion of the mass of the end-of-life materials, for which a relevant solution should be found to avoid landfill. While some of these products have a relatively high intrinsic value (carbon fibres) and can be reused “as is”, other products should be upcycled to increase their value (fibreglass, particularly if broken during recycling process). It is however necessary to remove potentially hazardous substances (such as brominated flame retardants from WEEE plastics) to avoid their dispersion in the environment. So far, most studies focused on WEEE plastics, but can provide valuable information that can be transposed to treat polymers and other materials remaining after metal recovery from LiBs, wind turbines, magnets, solar cells, or fuel cells.

Waste polymers (plastics) can be mechanically reprocessed,<sup>249</sup> burned to produce energy, or up-cycled into value-added products. The last option seems the most promising from an economic point of view. Up-cycling could lead to products such as graphitic or activated carbon,<sup>250,251</sup> carbon fibres, foams,<sup>252</sup> or silicon carbide,<sup>253</sup> for which the raw materials are pyrolysed WEEE. Fig. 5 regroups the value-added and cheaper products that can be obtained from waste graphite, fibreglass, or plastics. Up-cycling of these materials is an interesting option, but the value-added products are often niche applications with a small market. Cheaper products can also be produced using the waste as a raw material, but they offer a much larger market.

Waste plastics from WEEE have been investigated for construction materials produced in bulk quantities like asphalt binder.<sup>254</sup> In that case, plastics like acrylonitrile butadiene styrene improved the stiffness and elasticity of the asphalt,

possibly due to covalent bonding between plastics and the asphalt promoted by treatment with cumene hydroperoxide. However, high melting temperature plastics like high-impact polystyrene reduced the beneficial interfacial bonding with asphalt. WEEE plastics were also used as coarse aggregate<sup>255</sup> for concrete production, but adding more than 10 wt% reduces the mechanical strength of the concrete. Although it reduces the strength of the obtained cement, WEEE residue used as an aggregate could improve the workability and durability<sup>256</sup> of the concrete. From a circularity perspective, plastics are immobilised for a long time into construction materials, preventing them from being re-used more than once. The long-term behaviour of such materials is also concerning. It is necessary to anticipate the end of life of concrete containing plastic aggregates, and avoid leakage of microplastics from ageing materials.

Injection moulding can be used to make wood–waste plastic composites<sup>257</sup> with improved stiffness. 3D printing filaments<sup>258</sup> can be made from WEEE plastics, which gives more flexible filaments and significantly reduces their carbon footprint (around 30% less CO<sub>2</sub> when using the waste plastics). WEEE plastics were also investigated as cell culture substrates and showed promising results for cell growth and differentiation due to their surface properties.<sup>259</sup>

Overall, recycling of waste polymers from WEEE was more studied than polymers from end-of-life materials from renewable energy and energy storage technologies. Recycled EVA was investigated as a raw material to produce bituminous binders.<sup>260,261</sup> The incorporation of high contents of EVA (higher than 1 wt%) leads to the formation of polymer rich phases and seems detrimental to the mechanical properties of the bitumen. Incorporation of EVA and its blend with low-density polyethylene (LDPE) and ground tyre rubber could also produce

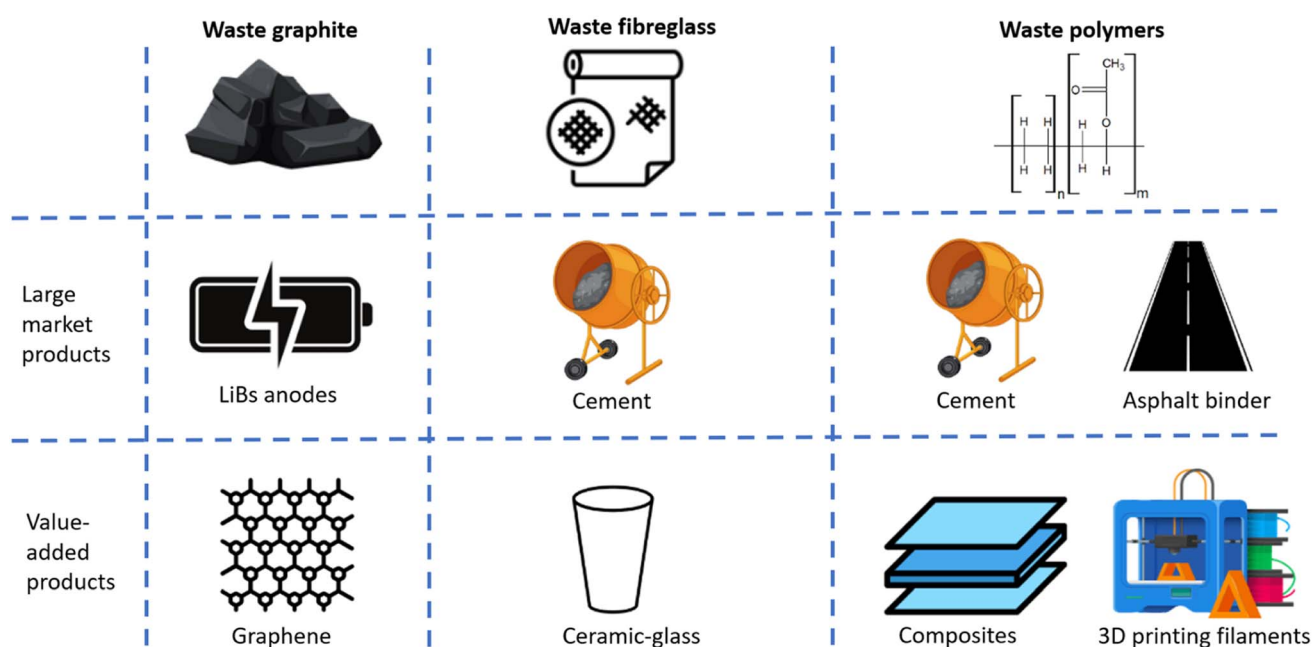


Fig. 5 Waste materials and potential recycled products.



thermoplastic elastomers<sup>262</sup> by injection moulding. While LDPE reduces the viscosity of the melt, the EVA copolymer improves the elastomeric properties of the material. Waste EVA could also be turned into vitrimers through a chemical<sup>263</sup> or mechanochemical<sup>264</sup> approach. Vitrimers are an emerging class of polymers with a dynamic covalent bonding. Exchangeable reactions (like transesterification) occur upon heating the vitrimer at specific temperatures, leading to easily recyclable materials with good mechanical properties.<sup>265</sup> Vitrimerisation of epoxy is promising to produce easily recyclable composite materials,<sup>266</sup> or recycle epoxy thermosets.<sup>267</sup>

Fibreglass is another high-volume and low-value material for which direct re-use is sometimes not possible due to reduction of size and mechanical properties during the recycling process. Fibreglass can be treated at high-temperatures (melting with Na<sub>2</sub>O, then sintering) to produce a glass-ceramic material.<sup>268</sup> Fibreglass were also investigated as reinforcement material<sup>269</sup> in concrete. Fibres with spherical particles, large size dust or splinters were shown to reduce the mechanical properties of concrete. The presence of oxides like CaO, Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> in glass fibres could improve adhesion and binding of concrete and sometimes improve its mechanical properties.<sup>270</sup> However, the incorporation of a high proportion of fibreglass reduces the mechanical properties of mortars.<sup>271</sup> 3D printing reinforced filaments<sup>272</sup> are amongst the products that can be made with waste fibreglass.

Waste graphite was investigated to make conductive electrodes and produce triboelectric generators.<sup>273</sup> Graphite can also be up-cycled to higher-value graphene.<sup>274</sup> But knowing the large amounts<sup>16</sup> of graphite needed for LiBs anodes, the most straightforward market for recycled graphite would be new anodes.<sup>13</sup>

Amongst the biggest challenges faced for low-value fractions recycling is the lower mechanical properties of recycled products, mismatch between the volume of waste available and the size of the market for recycled products, potential dispersion of hazardous substances, and value of recycled products. While valorisation of WEEE plastics has been studied, management of uncommon polymers such as fluorinated ones or EVA (to a lesser extent) is yet to be addressed and is particularly important knowing the potential environmental damages and the evolution of regulations.

## 2.6 Limitations and future directions

**2.6.1 Manufacturing processes and designs conducive to recycling (eco-design).** There are numerous recycling aspects that can be simplified by the design of the original device, some of these are listed in Table 1. For recycling to be economic and even profitable it requires systematic and automated process flows similar to the current manufacturing assembly lines. Despite the significant progress made to address the urgency of TCM recovery, there are limited industrial scale processing facilities and many processes still remain at low technology readiness levels (TRL), due to diverse and evolving device structures and materials. There is room for more automation in current recycling practices. Use of robots able to perform many

operations (pick and place, cutting, unscrewing...) have been proven effective, particularly for disassembly of battery packs,<sup>275,276</sup> and dismantling of WEE.<sup>277</sup> But lack of standardisation hinders the automation of many recycling processes. Apple's recycling lines can only deal with specific models of iPhones<sup>278</sup> since different models use different designs, different screws and fixings. The lack of flexibility and difficulty in adapting automated recycling to different models and objects can be partially solved if similar fixings are used. Similarly, favouring physical connections (for *e.g.*, mechanical clamps for encasing), when possible, over soldering or glueing could make automation easier. The presence of multiple layers and different interfaces also hinders recycling. Reducing the number of interfaces decreases the number of fixings and the amount of adhesives used, while larger units decrease the amount of casing, materials and seals used.

As such, it is worth addressing why commercial deployment of recycling processes hasn't kept pace with the rapid improvements in research and development over the past 10 years. Although regulation, environmental and health concerns play a role in the development of a circular economy, it is clear that a viable economic model must exist to recover TCMs. Therefore, greater emphasis must be placed on how to incentivise recyclers to recover materials. As materials become more widely used and scarce, it is quite possible that market forces will play a role in encouraging recycling; for example, if the demand for virgin materials escalates, so too will the cost, thus reducing the price differential between virgin and recovered materials. However, relying on market forces can lead to perverse and undesirable consequences and, as such, government intervention may be required to ensure that (i) new products are designed with recycled content, (ii) materials are recycled as a public service, or even (iii) taxes on virgin materials to make recycled materials more economically desirable are introduced.

In addition to the standardisation of recycling methods for existing commercial products, it is important to identify module and device structures that simplify metal recovery from devices without affecting their efficiency and lifetime. For *e.g.*, EVA from silicon solar cells involves peroxide, silane or radiation crosslinking which imparts its high thermal and mechanical durability, but at the same time increases the difficulty in recycling. EVA vitrimers were shown<sup>263</sup> to retain 90% of their Young's modulus after 5 iterations of the recycling process as compared to 72% retention of Young's modulus by the thermoplastic EVA. This directly increases the end-of-life value of the panels. At the same time, the modified processes must not affect the key material parameters (in this example, the transmittance of the EVA sheets), so that it is not detrimental to the efficiency of the device (solar cell). If the decrease in the recycling cost fares better than the increase in manufacturing complexity (and hence cost) of the modified process, then it can be a major incentive towards recycling.

**2.6.2 Adapting to emerging technologies and economical aspects.** The complexity of the devices in core electronics as well as in energy harvesting/storage devices has increased continuously, both in terms of structure and materials. Even very



Table 1 Design for recycle approaches and associated issues

Approach	Issue
Base metal substrates	Co-deposition of base metals with TCMs could enable selected release through catalytic oxidation <i>i.e.</i> a debondable substrate
Debondable adhesives	Permits judicious separation of components without compromising structural integrity
Water-miscible adhesives	Enable TCMs to be separated from binders and circumvents high temperature pyrolysis
Biocomposites	Enable specific biocatalytic digestion of binders
Standardised fixings	Negates the need for unnecessary re-tooling during automated disassembly
More physical connections	Allows more rapid disassembly and repair
Fewer interfaces	Decreases the number of fixing and amount of adhesive required to construct devices
Larger units	Decreases the amount of casing materials and seals

mature technologies such as size reduction or physical separation methods will require some form of adaptation to apply them to relatively new waste streams like spent LiBs, which come with their own challenges, such as safe dismantling and discharge. Further, more and more electronic devices like batteries and supercapacitors are integrated onto non-conventional fibre-based substrates, which frequently use Cu, Ag and Ni fibres.<sup>279,280</sup> These devices are moving towards commercialization and recycling processes must adapt accordingly.

For solar panels, most recycling processes aim at recovering glass and the Al frame only. The current mechanical waste sorting practice is to shred the whole module including the frame and junction box or to remove the frame and junction box and shred the solar cell, recovering glass, Al, plastic, some metals and Si. Current plants claim 95% recovery rates, most of this is low grade glass that is used for glass fibre insulation or fillers, the rest being primarily Al from the frame. Future recycling should target recovery of intact silicon wafers and metals, which require a minimum of 30% cost reduction for both mechanical and thermal recycling processes.<sup>281</sup> Further, thin-film and flexible solar cells devices might emerge which will require modifications in the recycling process flow. Various solvent based techniques are reported to remove the organic and metal halide layers from perovskite solar cells.<sup>282</sup> As the perovskites and other organics are dissolvable in various solvents, mechanical crushing to reduce particle size and subsequent step by step dissolution in various solvents can recover the metal electrodes from these, but the recycling processes must adapt to take into account the environmentally toxic materials (like Pb and Cd) present in these cells.

For wind turbines, decommissioning and transport of such large objects is an economic challenge, particularly the on-site decommissioning of offshore wind turbines. For spent LiBs, the value lies in the recovery of high-purity cobalt, nickel, manganese and lithium salts but their purification is difficult and currently based on complex processes.<sup>283</sup> For PEM fuel cells, value lies in the PGM catalysts, the challenge being to liberate them from the anode and cathode catalyst layer, while management of fluorinated polymers is another (potentially costly) issue. In any case, techno-economic assessment is a very important tool to assess which processing route is the most suited for each material and metal content.

In addition to TEA, LCA is an important tool to choose between multiple processes and select the one with the lower environmental impact, which is unfortunately made difficult by the multiple methodologies available and lack of standardisation. Another drawback is the lack of precision when the LCA is performed in the early stages of process development,<sup>284</sup> when real-world data from a plant do not exist yet. Furthermore, optimising LCA metrics does not lead to the best performing or lowest cost solutions so methods that combine environmental impact and performance should be considered.<sup>285</sup> For chemical processes, it is however possible to get more accurate data from stoichiometric reactions by combining reaction yields with the average values for utility demands.<sup>286</sup>

As the ease of removal of the polymers directly translates into easier and cheaper access to the metal underneath, the recycling complexity, costs and environmental impact of each alternative/emerging class of encapsulant materials must be compared. In the case of solar cells, non-crosslinking encapsulants like thermoplastic polyolefins and polyvinyl butyral<sup>287</sup> (PVB, extensively used in safety glass laminates in automotive industry) or silicone<sup>288</sup> based encapsulants are potential alternatives that are possibly easier to dissolve<sup>289</sup> or degrade.<sup>290</sup>

The choice and design of adhesives can reduce the complexity of the recycling process, especially for adhesives that can be removed easily using a trigger mechanism such as ultrasound, magnetic field, electric current, mechanical stress or temperature.<sup>291</sup> Adhesives that can be easily dissolved with a chemical are particularly attractive, but would require significant volumes of solvent. At the same time, these alternatives put a limitation on the environments where the devices can be used. For *e.g.*, water-miscible adhesives (particularly as blends) would only require water, but their use would be limited to applications where humidity levels remain low throughout the lifetime of the product. Stress or ultrasound triggered adhesives cannot be used for flexible devices which are expected to withstand high bending stresses in their working environment. Debondable adhesives were shown to improve reparability of small electronics such as smartphones,<sup>292</sup> and have a major role to play in other devices such as solar cells<sup>293</sup> or LiBs.<sup>294</sup>

**2.6.3 Newer chemical processes and degradable materials.** The leaching-based hydrometallurgical recycling processes being developed offers a selective separation between metal and nonmetals but tend to require long processing times, involve





significant volumes of ancillary chemicals, and can produce product streams of lower value and purity. Moreover, certain metals (in particular precious metals) are hard to oxidise and require problematic, noxious and/or expensive oxidising agents (e.g. cyanides, nitric acid, aqua regia, iodine). Using base metals to link the precious metal to its organic or inorganic substrate can reduce consumption of these chemicals. Base metals (such as aluminium) can be easily dissolved with common household chemicals (such as sodium hydroxide), leaving the precious metals to be filtered off. This approach was proven feasible for thermoelectric materials,<sup>129</sup> where a semiconductor leg (containing Bi, Sb, Te and Se) is attached to an inert ceramic plate by a layer of copper. Copper can be selectively oxidised using  $\text{Cu}^{2+}$  salts, separating the semiconductor leg from the ceramic plate.

Biocomposites are composites materials made of biodegradable polymer matrices (starch, cellulose, *etc.*) and/or bio-fibres of vegetal, animal or mineral origin (hemp, bamboo, *etc.*). These materials are very attractive since they could make recycling easier.<sup>295,296</sup> In addition, there is the emerging area of biodegradable electronics that might also simplify recycling processes,<sup>297</sup> enabling recovery of higher value metals and components. In biocomposites, matrix material is selected to avoid epoxies or other petroleum based and non biodegradable polymers, allowing specific biocatalytic digestion of binders, and easy degradation of the polymer matrices. Combustion of these materials does not release additional carbon dioxide, while residual fibreglass after thermal treatment is eliminated. Careful selection of the matrix and reinforcement composite is needed to ensure good mechanical properties to the materials, long durability and low cost.

**2.6.4 Reuse in high performance applications and responsible innovation.** A major challenge relates to the purity of metals and semiconductors needed for high performances, particularly the refinement of the recovered semiconductor for electronics or solar cells. For example, refinement is needed with a minimum purity of 6 N, though some applications require 9 N purity (99.999999%). The refining process needs to remove dopants, coatings and any other contaminants that might have been introduced during the usage stage. Many of these refinement processes are at TRL 3–4 and are not commercialised, because of the economics (energy use).

Finally, the concept of responsible innovation should be highlighted.<sup>298</sup> Many items are created using TCMs which are designed for a short linear life; an example of this would be disposable vapes. They are created on a large scale with sufficient lithium to be hazardous but insufficient to make recycling viable. There are other less hazardous batteries that could be used, but irresponsible innovation has led to a large, worldwide, complex waste issue.

### 3 Conclusion

The transition to a zero-carbon energy system will require the development and deployment of new energy generating and storage technologies. These technologies will use a wider range of critical materials and will be more complex than the technologies used in the era of hydrocarbon-fueled thermal power

generation. Increasing dependency on technologies such as wind turbines, solar cells, lithium-ion batteries or catalysts for hydrogen production leads to increased consumption of critical metals. Better recycling processes for these technologies are urgently needed to recover metals more sustainably and with minimal environmental impact.

To do so, a set of tools could advantageously complement the conventional hydro- and pyrometallurgical methods by targeting interfacial weaknesses between the different layers. Traditional physical methods of size reduction (comminution), associated with physical separation methods, can be easily implemented to liberate and separate metals from organics and inorganics fractions. High-intensity ultrasonication is useful to delaminate materials and process brittle materials. Organics can be dissolved, pyrolysed or gasified, while metals can be selectively oxidised and dissolved, possibly with biological processes. Hydrogen decrepitation is particularly indicated for magnet recycling. In order to improve recycling rates, low-value fractions like plastics can be turned into construction materials or up-cycled into value-added products. Ultimately, selection of the process applied will depend on techno-economic and life cycle assessments. Given the enormous volumes of technology end-of-life materials that will need to be processed in the future, it could be imagined that future approaches to the treatment of sophisticated technology end-of-life materials, might leverage generic knowledge about the archetypes of mixed material end-of-life materials, combined with automated disassembly and sorting and the application of AI and digital technologies to enable high throughput, high value intelligent recovery from a wide array of materials.

The concept of “Digital Passports”, where physical assets are associated with a digital record that contains details about the material that they contain, are another potential technology that could enable high value sorting of TCMs. Some form of digital asset code, whether it be RFID or QR code, could potentially provide information to future intelligent systems, which could then apply a toolbox approach to select suitable processes for recycling materials streams. Analysis by SystemIQ predicts that such a digital passport could increase profitability for recyclers of lithium ion batteries by 20–30%,<sup>299</sup> by optimising pre-processing, material sorting, and aiding in reduced sorting and automated dismantling.

Application of design for recycle principles and responsible innovation are clearly needed to improve recyclability of products before manufacturing. Aspects of design can also substitute components with TCMs for more Earth-abundant alternatives, often with negligible effect on performance.

This paper focuses on a toolbox of technical approaches that can be applied to the challenges of materials criticality and circularity. However, there is also enormous potential to shape the flows of critical materials through a circular economy by shaping the social dimension of the socio-technical systems that utilise them. In some cases, innovation in business models, product service-systems<sup>300–302</sup> and new modes of consumption could bolster efforts to commercialise and implement technical improvements to their conservation and recycling. With the application of different approaches being applicable to different



materials and production-consumption scenarios, it could be envisaged that a complementary toolbox of business models and product service systems could be synergistic with the push to improve recycling technology, and in many cases, the enabling innovation that facilitates the new approaches outlined in this paper, may be social and not technical in nature.

## Abbreviations

CC	Concentration criterion
DESs	Deep eutectic solvents
DMAc	Dimethylacetamide
DMF	<i>N,N</i> -dimethylformamide
DMSO	Dimethyl sulfoxide
ENIG	Electroless nickel immersion gold
EPR	Extended producer responsibility
EVA	Poly-(ethylene-vinyl acetate)
HASL	Hot air solder levelling
HBD	Hydrogen bond donor
HDDs	Hard drive disks
ILs	Ionic liquids
LCA	Life cycle assessment
LCDs	Liquid crystals displays
LDPE	Low-density polyethylene
LEDs	Light emitting diodes
LiBs	Lithium-ion batteries
NMP	<i>N</i> -methylpyrrolidone
PCBs	Printed circuit boards
PE	Polyethylene
PEM	Proton exchange membrane
PET	Polyethylene terephthalate
PGMs	Platinum group metals
PP	Polypropylene
PTFE	Polytetrafluoroethylene
PVB	Polyvinyl butyral
PVC	Polyvinyl chloride
PVDF	Polyvinylidene fluoride
REEs	Rare-earth elements
TCMs	Technology critical metals
TRL	Technology readiness level
WEEE	Waste electronic and electric equipment

## Conflicts of interest

There are no conflicts of interest to declare.

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