Research article

Designing lithium-ion batteries for recycle: The role of adhesives

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A B S T R A C T

While electric vehicles are seen as an important tool in the decarbonisation of transport, pack and module architectures make disassembly and recycling slow and complex. Removal of physical fastenings such as clips, screws, welds and adhesives are the rate limiting factor in pack to cell disassembly. This study investigates the types of polymeric adhesives which are used in various battery components and shows how careful choice of components can speed up disassembly and circumvent the need for shredding and increase the purity and value of the recycled material.

1. Introduction

The demand for lithium-ion batteries (LIBs) in electric vehicles (EVs) has increased significantly due to their potential in decarbonisation of energy production. However, the scale of the projected electric vehicle market also indicates the exponential amount of battery waste that will be produced in the coming years, with conservative estimates stating that 1 million EVs have the potential to produce 250,000 tonnes of battery waste. As the market share of EVs grows, having surpassed 16 million in 2022, it is clear that the development of a circular economy model now whilst the scale of end-of-life products is still manageable is essential to not only treat this waste but also recover the critical materials used in battery manufacturing. [1,2] Additionally, life cycle analysis (LCA) shows that recycling processes have the potential to reduce greenhouse gas emissions of the entire battery life cycle by 20 kg CO_2-eq/kWh_bc. [3] This industry will develop in concert with the processing of manufacturing scrap, and whilst in the early stages the recycling industry will have to contend with existing battery designs, in the future better designed batteries could unlock enhanced and more efficient recycling methods. A comprehensive roadmap for LIB recycling has recently been published, which summarises the key developments in LIB recycling, including design for recycle, and issues that still need

to be addressed to establish a scalable recycling methodology capable of establishing a circular economy. [4]

The primary concern of battery design is to ensure safe and long battery performance with a high-power density to efficiently store electrical energy. Many of the design features currently employed make efficient and economic recycling challenging. Recycling has previously been seen as an end-of-pipe process, with little thought for end-of-life dismantling or processing. The approaches currently used are similar to those used in other waste recycling and primary metal extraction from ores. [1]

As with all complex devices, the components can be assembled using a variety of methods to contact distinct phases for structural integrity, strength and electrical continuity. These joining methods can either be physical (clips, screws, springs etc.), metallic (welds and solders), inorganic cements or organic adhesives. Unfortunately, the in-service properties are generally at odds with the end-of-life requirements. In service the joint needs to be durable and non-reactive whereas at end-of-life it needs to be soluble or reactive. Most recycling processes start with a disassembly of the battery pack down to either module or cell level. From there, most physical and hydrometallurgical recovery start with comminution (shredding) to break joints and separate the different phases which results in cross-contamination between components and results in low value product streams. This is particularly an issue for

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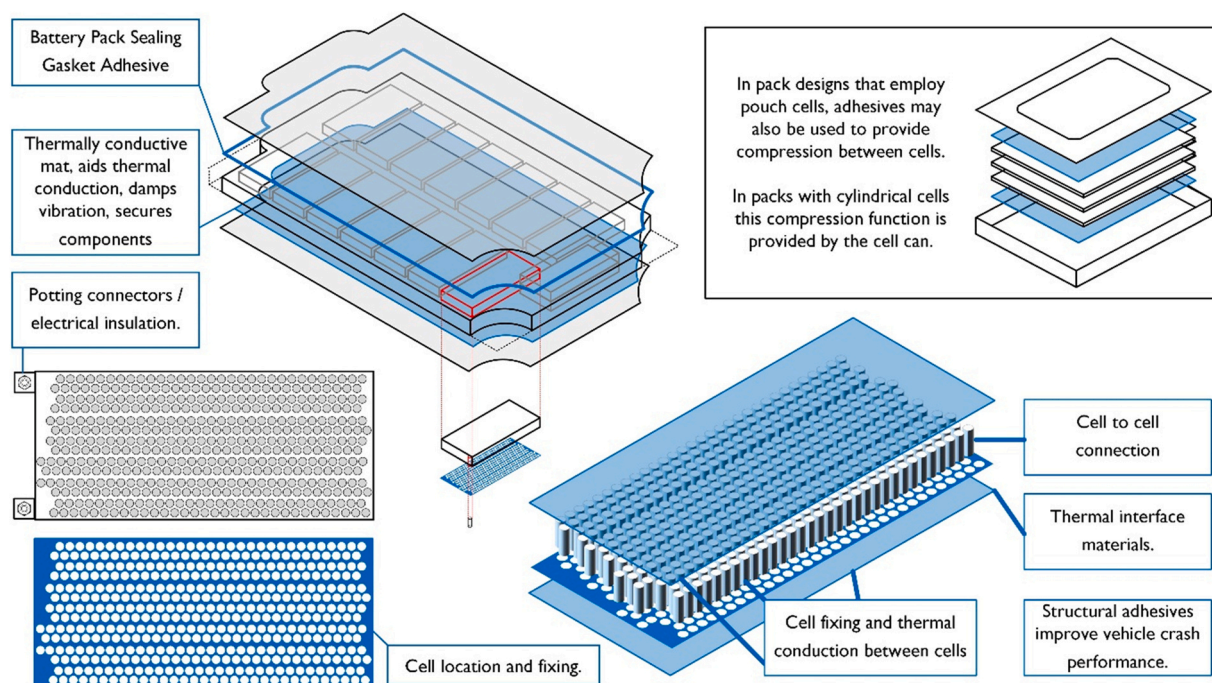 *E-mail address:* apa1@le.ac.uk (A.P. Abbott).

Fig. 1. Examples of where structural adhesives are used within a battery pack using cylindrical cells.

polymeric and soldered components. A recent study on LIB recycling highlighted the difference in recycling economics depending on recycling method, geographical location and battery chemistry. [5] Dismantling the cell down into individual electrode materials as an alternative to shredding, can significantly improve economics and product purity but it is hampered by the complexity of the component joining techniques. [6] The cost of manual and automated disassembly has been estimated from the fixing and connector types found in a range of battery packs. [7,8] The study showed that economic recycling of battery packs requires automation which in turn depends on pack, module and cell design. Automated industrial disassembly has been argued to be a key enabler of a circular economy of EV batteries. [6] Current designs make disassembly complex due to the array of connectors used, the scale and packing of the cells and mechanical and chemical damage to the components during service. [9,10] It was noted in particular that the biggest barriers to disassembly were the number of screws when disassembling from pack to module and the number of welds and structural adhesives as well as the number of modules when going down to cell level. Fig. 1 shows some of the locations and application for polymeric adhesives used for in battery packs. For the structural and longevity reasons listed above, thermoset resins and unreactive fluorinated polymers have been extensively used throughout battery structure.

The logistics of moving waste is also important and it was shown that reducing transport and disassembly was important to the overall economics of recycling. [5] The geospatial configuration of a future recycling system will have a bearing on the impacts that arise at the end-of-life. The scale of the recycling plant is also important as are the hazard classification of end-of-life cells as it will affect the transportation costs if they are classified as hazardous. The configuration of technologies used for end-of-life processing, will have a bearing on the topology and structure of this future industry. Simple pre-processing that can take place near to the point of disposal may aid in reducing the quantity of material moved around the system, and the distances material must travel reducing the environmental burden of the end-of-life phase. Batteries that can be disassembled more easily close to point of disposal may unlock "hub and spoke" recycling models.

A variety of studies have investigated the technoeconomic assessment of shredding and disassembly. It was found that cost saving (with

respect to using virgin material) of up to 20% could be achieved using shredding whereas cell dismantling could recover material with up to 80% cost reduction. [11,12] The advantages of shredding are that it rapidly reduces the active battery into a safer format. It is a process that is easily scaled, although the atmosphere around the shredder does need to be controlled. However, shredding does not separate aluminium from lithium metal oxide efficiently and attrition milling down to sub-mm scale is required to get reasonable separation. [13] Studies have shown that impurities incorporated into recycled cathode material can significantly affect the performance of cells. [14] There are, however, caveats with these studies which, in most cases, have not considered the cost of disassembly and, for those which have, they do not dismantle beyond module level.

Design for recycle is not a topic which has been discussed in detail but a recent critical review highlighted many of the issues. [15] Design for disassembly has been discussed for removal of lithium ion batteries from PC laptops and although much simpler, issues of structural adhesives and fixing types are common with the automotive sector. [16] Product disassembly and material liberation is frustrated by the use of non-reversible adhesives in products. [17] In some cases, with thoughtful design and strategic placement, non-reversible adhesive bonds can potentially facilitate recycling, where they enable a "path of preferential breakage" which aids material recovery. The concept of disassembly sequence planning has also recently been introduced. [18] Moves to make the battery pack a structural element of the vehicle have led to an increased use in structural adhesives and permanent welds to increase pack rigidity. For example, the use of thermoset resins leads to the necessity for shredding rather than dismantling. A further disadvantage of thermosets is that they cannot be recycled and can only go to energy recovery. Thermoset resins are a high surface area foamed material, which also complicates separation and leads to entrapment of active material in streams destined for combustion.

Shredding has the disadvantage that the majority of electrolyte cannot be recovered and hydrolysis of salt can lead to the production of hydrofluoric acid (HF). It also leads to the necessity to scrub organic solvent from wastewater streams. While lithium is a minor value component, it is a significant mass component and lithium recovery efficiency is being targeted by battery legislation in many regions. In most cases the PF_6 anion has a significantly higher value although it is lost in most recycling processes.
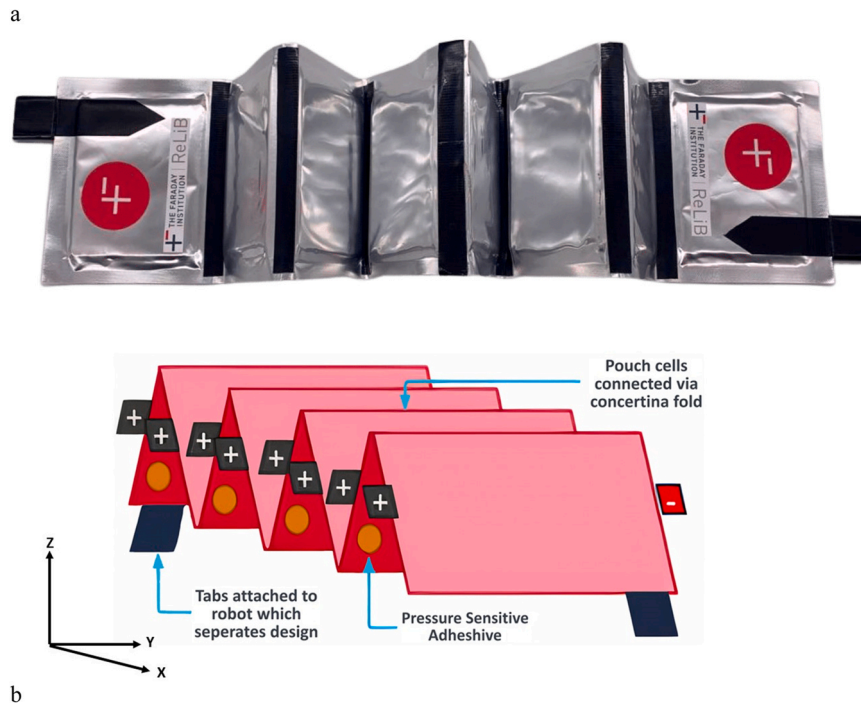
Fig. 2. The proposed zigzag conformation for pouch cells. a) image of the dummy cell used in these experiments and b) the schematic diagram for the conformation showing placement of the adhesives.

The aim of this study is to demonstrate some simple design modifications that could be adopted to improve the ease of pack disassembly. Using dummy cells and modules the efficacy of disassembly is shown using, at times, robotic manipulation and the timings are compared to those proposed by Lander et al. [8].

2. Design for disassembly

Returning any device to its constituent components is fundamentally restricted by the numbers of interfaces and the methods chosen to join those interfaces. Recycling is made more challenging when dissimilar materials with incompatible properties are joined permanently using non-reversible adhesive bonds, making them difficult to separate. The three basic joining approaches; thermal welding, chemical adhesion and physical connection have implications for the cost, speed and permanency of the join and its subsequent release. The complexity of battery architectures has been highlighted in several recent reviews. [1,15,19].

There are numerous aspects of pack, module and cell design which can be adopted for simplified disassembly and recycling:

- Fewer, but larger cells
- Minimal use of thermoset adhesives
- Fewer fixing types
- Cells that are more easily opened
- Electrode binders that can be fully dispersed using water.

This article will review each of these ideas showing the extent to which each is viable. It will also analyse a case study currently being carried out to disassemble and recycle lithium iron phosphate (LFP) cells and the design aspects that can be assimilated from this process to other battery chemistries. The polymeric components can be split into two types depending on their applications:

- Extracellular: These hold the cells, modules, cooling components and the BMS together and are chosen primarily for their strength.
- Intracellular: These are chosen largely for their inertness and flexibility and maintain the active material in contact with the current collector.

2.1. Extracellular Adhesives

Disassembly tends to lend itself to form factors with larger amounts of battery material per cell, i.e. prismatic and pouch rather than cylindrical. This is for two main reasons: the time taken to open the individual cells and the mechanical separation of cells from each other in the module. The Tesla Model S P85 battery pack, for example, has 16 modules, containing a total of 7104 cells whereas the BMW i3 Mk 1 has 8 modules, each containing 12 cells (96 in total). Cell opening is viable with the latter as each cell opening process yields approximately 2 kg of material but with the former, less than 50 g of material are obtained. The disassembly of cylindrical cells is further complicated by their geometry, with active materials, foils and separators being spiral wound into a "swiss roll" like configuration, rather than the planar geometry of electrodes in pouch and prismatic cells.

In most pack and module designs currently used in the automotive sector, structural adhesives provide rigidity and strength to the assembly. Their use is logical, as they are inexpensive to apply and irreversibly provide the strength needed to minimise movement of cells during use. Most of the adhesives used are thermosets, based commonly on epoxides or polyurethanes. A recent critical review explained the possibility of using debondable adhesives which incorporate an element which enable depolymerisation or bulk delamination but it highlighted the difficulties of using heat, light or electrical potential as debonding stimuli in a battery pack. [19].

An important factor in the cost of LIB recycling is the complexity of pack disassembly. Manual disassembly is too slow and costly in many regions due to labour costs. It has recently been estimated that the manual disassembly time for many makes of electric vehicles are currently in the range 8–10 h. Automated pack disassembly down to modular level has been estimated to take 1–2 h and this is severely limited by the retooling of robots and the time taken to remove welds and glues. Most fixing methods are permanent, so servicing and end-of-life are not part of the pack design. Nine joining methods for metals in batteries have recently been identified, [20] and of these, only mechanical assembly (screws and clips) is seen as a process which makes disassembly easier, however mechanical fixtures add to the weight of the battery pack.

An important design for disassembly would be to avoid using structural adhesives as the only form factor imparting strength to a module or pack. One method to decrease the use of structural adhesives between cells could be to create a permanent link between pouch or prismatic cells and strategically place a small amount of adhesive at a point where selected directional movement could physically break the bond. One such arrangement could involve hinging the cells at alternate ends to create a zigzag conformation as shown in Fig. 2. This decreases the degrees of freedom that each cell can independently move in and generates levers between the cells. This could significantly decrease the amount of adhesive that needs to be applied to impart structural rigidity. If the properties of the adhesive are tuned correctly, separation of the cells into a linear configuration (Fig. 2) could be carried out effectively with programmed robots as only a simple extension of the design is required in the x-axis for the disassembly to occur.

2.1.1. Pressure sensitive adhesives

Pressure sensitive adhesives (PSAs) are a versatile class of viscoelastic materials which form bonds using initial pressure and flow, unlike conventional adhesives, which bond once they have hardened through a chemical or physical process. PSAs do not require additional agents such as heat, water, or solvents to activate. Due to this, the rheological properties of the adhesive must be adjusted specifically to their application. [21] The three main characteristics to be considered are ultimate adhesion, shear resistance and initial tack. Ultimate adhesion is the measure of the strength of the fully formed bond once the adhesive has set, shear resistance correlates to the adhesive resisting forces parallel to its surface and initial tack corresponds to the property that controls the instantaneous formation between the adhesive and adherend. [22] Contact adhesives are easier to apply than thermoset resins and are already used in small aspects of pack construction, their application over large surface areas make debonding slow and necessitates large volumes of solvents.

An investigation was carried out to compare the effectiveness of different types of commercially available PSAs including glue dots, double-sided tape, and Velcro. A peel test with tensile testing apparatus was used to simulate how a real module of cells using the zigzag conformation would be pulled apart. For this investigation two pouch cells of total weight 1.6 kg were created with a hinge between them by heat sealing a large pouch (41.5×23.5 cm) in the middle. Initially no adhesive was used to set a base level for the different PSA's and to accurately compare their loading and extension which in turn corresponds to the energy required to break the connection. Then a PSA was applied onto the test sample and planted in our universal testing system. All adhesives were able to resist shearing motion in the z-axis and provided reasonable strength in the x and y axes as shown in Table 1.

As seen by the data in Table 1, double-sided tape is the strongest adhesive of the set closely followed by Velcro. Both provide viable solutions to holding a zig-zag-configuration cell together, however weight and spacing must be considered alongside the ease of removal of adhesive. In contrast to glue dots or double-sided tape, using Velcro increased the thickness of the sample design by nearly 35%, decreasing the power density. Additionally, a weight calculation was carried out to

evaluate each adhesive for the test sample, where it was found that Velcro contributes a relatively high amount of additional weight, while the glue dots contribute almost no weight.

Comparing the ease of removal of the adhesives, the double-sided tape was found to be the most difficult to remove, due to the sticky residue left behind. However, even this residue can be removed relatively easily with an acetone wash or by hand. Both Velcro and the glue dots did not leave residual material behind, thus making them a preferential choice in this respect as they would not require the additional removal step and therefore provide the simplest disassembly procedure. This is particularly useful in the area of pack or module repair and may decrease the proportion of cells being scrapped during production.

The amount of force required to separate the pouch cell design is also dependent on the position and orientation of the pressure sensitive adhesive. To demonstrate the applicability of using this approach, a dummy battery pack was constructed using 8 cells, each scaled down to 110×140 mm weighing ~193 g each, which was about 20% of the mass of a pouch cell that would be used in the Nissan Leaf. This was done due to limitations of the lifting capacity of the robots used in these tests. 'Franka Emika Research 3' robots were used to lift the 8-cell module from a box using two tabs. Two glue dots were adhered on alternate sides of the line of joined cells such that they could be aggregated into a zig-zag pattern. This approach enabled the ensemble to have mechanical strength but allowed some movement within the confines of the module container. The robot was able to lift the module contents and extend the string of cells into a straight line, as shown in Fig. 3, in only 15 s and this can also be viewed in video format. This automatic process could resemble how industry standard disassembly of a full battery pack could take place.

Alternative ways in which this could be carried out would be using strapping, commonly used in commodity transportation. These straps are usually made of polypropylene or polyester with either a thermal or metal crimped seal. This provides a physical join which is quicker to unfasten/refasten. Cutting this band would provide instantaneous cell release and simplify disassembly. Some test modules were made with single sided adhesive tape across the cell stack. These are reinforced tapes, but allow quick release when the tape is cut.

Cell opening can be easily automated with mechanical processes, however the material composition of the cell container has a significant impact upon which method can be utilised. A list of different methodologies that can be used are given in Table 3. The pouch cells are laminates of aluminium with polyethylene terephthalate (PET) and polypropylene (PP). [23] These are easily cut open with ceramic blades, leaving little residues or extra contamination. Other opening methods such as using lasers or plasma, produce heat during the cutting process,
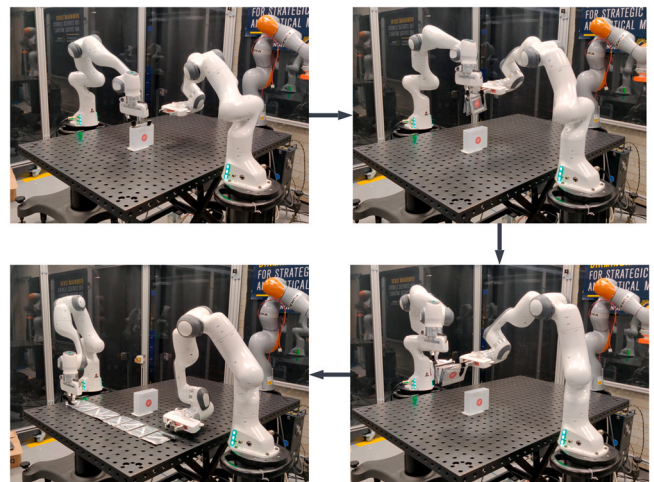
Table 1
Comparison of the forces and associated energy required to open the modules using the zigzag conformation when different pressure sensitive adhesives are used.

Pressure Sensitive Adhesive	Average Load (N)	Maximum Load (N)	Energy Usage (J)
No Adhesive	6.02	9.76	2.12
Glue Dots	15.83	22.65	2.82
Velcro Strips	31.90	67.89	3.43
Double Sided Tape	58.67	103.77	4.49

Fig. 3. Images showing the stages of robotic module opening when the zigzag cell conformation is used. This process takes place over the course of 15 s.

but this can be optimised through fast, short pulses allowing for heat dissipation. [24] Any heat or sparks may ignite the flammable electrolyte. The sparking and heat removal may be further reduced with cooling fluids, which can also have a second benefit for passivation of the components as the cell is opened. [25] In addition, ultrasound could be utilised for pouch cell opening, as this tool can cut through multi-layers, and is used for materials which melt if exposed to heat, such as the laminate.

As mentioned, large format pouch and prismatic cells are preferred for this pack configuration due to their ability to stack easily within a module. The cells are stacked and glued together in the stack, with adhesive pads at the stack ends, to ensure rigidity in the module design. Pressure sensitive adhesives are already used in some aspects of battery construction in the form of double-sided sticky pads inside the module case, to maintain an intimate bond between the cell stack and the module case. However, the large contact area makes debonding problematic for disassembly. Removing the cell stack and separating the individual cells from their epoxy resin can take up to 2 h and require several litres of solvent. This disincentivises careful dismantling and the cells are usually shredded instead. Strategically placed, judicious use of adhesive pads may enable mechanical dismantling and also contribute less mass to the battery module. This could significantly improve the possibility for repair and reuse. While there may be concerns about the structural integrity of EV packs bonded in this way, this sort of approach could be ideal for static battery packs, where mechanical action is less of an issue.

2.2. Intracellular polymer binders

Once the cell is opened and the electrodes are separated, the next challenge is to separate the active material from the current collector and the polymeric binder. Polymeric binders provide adhesion and interconnectivity between electrode components, but they cause significant issues when left as a residue within battery waste streams obtained via shredding, known as the 'black mass'. Interactions between the binder and the other electrode components (active materials and additives) occur during slurry mixing via two mechanisms: direct binding, where the binder is physically adsorbed to adjacent particles forming interparticle bridges; Fig. 4a gives an example of this binding mechanism for a gelatin binder. The other mechanism is via indirect binding, where the polymer forms a chemically inert network which constrains the particles, seen in Fig. 4b for a polytetrafluoroethylene (ptfe) example system. [26] Sufficient dispersion of particles is also imperative in the formation of homogeneous slurries and is dependent on numerous factors, such as the density, flexibility and polarity of polymers, to promote electrostatic repulsion. Usage of appropriate solvents for a given polymer facilitates dissolution and aid in dispersion

of particles within the slurries. For instance, the conventional polyvinylidene fluoride (PVDF) binder possesses a high dipole moment necessitating the use of polar solvents, such as N-methyl-2-pyrrolidone (NMP), to dissolve the polymer and resist flocculation within the electrode manufacturing process. [27] Binders also play an important role in electrochemical performance as key attributes of the binders such as flexibility and oxidation/reduction resistance, can dictate the degree of structural changes and chemical decomposition, impacting the amount of capacity fade and consequently the lifetime of these batteries. [28].

Recently, alternative water miscible binders have been the focus of research to reduce the usage of toxic solvents used in conventional electrode slurries, such as NMP. However, implementation of alternative binders is also essential to facilitate simplified and low energy separation of the electrode materials during battery disassembly. Fluorinated binders, such as PVDF, require high temperature pyrolysis ($>400\,°C$ in air) to be removed, which produces toxic gaseous products such as HF during decomposition. [29] In-service breakdown products from PVDF, such as HF, are capable of reacting with transition metal oxides within the cathode active materials, decreasing their capacity. [30] These conditions could be significantly improved if an alternative binder is used in manufacturing. At present, the use of alternative binders has been largely limited to current and next generation anodes, with the most common example being carboxymethyl cellulose (CMC)/styrene butadiene rubber (SBR), due to CMC being dispersible in water and SBR possessing good thermal stability, flexibility and adhesion. [31,32] Additionally, other water miscible binders, such as guar gum, [33] gelatin, [34] sodium alginate [35] and chitosan [36] have been investigated, showing similar properties to the CMC/SBR binders, with the possibility to be further enhanced via modification. [37–39] These water dispersible binder systems eliminate the need for the intensive conditions required for conventional battery separation steps, allowing facile separation of the active material and current collector, promoting the production of higher purity waste streams and simplifying subsequent recycling procedures.

Implementation of these alternative binder systems into cathode materials has been limited, as the resulting electrodes experience extensive cracking, and poor adhesion. The source of this cracking has been attributed to multiple factors, with corrosion of the aluminium foil considered to be the primary source. Aluminium corrosion is an issue in most industrial applications, however corrosion rates are limited, unless pH is significantly increased or decreased. [40].

$$2Al + 6H^+ \rightarrow 2Al^{3+} + 3H_2 \tag{1}$$

Water-based, positive electrode inks encourage the reactivity of the surface of the cathode materials, forming hydroxides which can dissolve in the ink, creating an alkaline slurry. If the pH is not controlled,
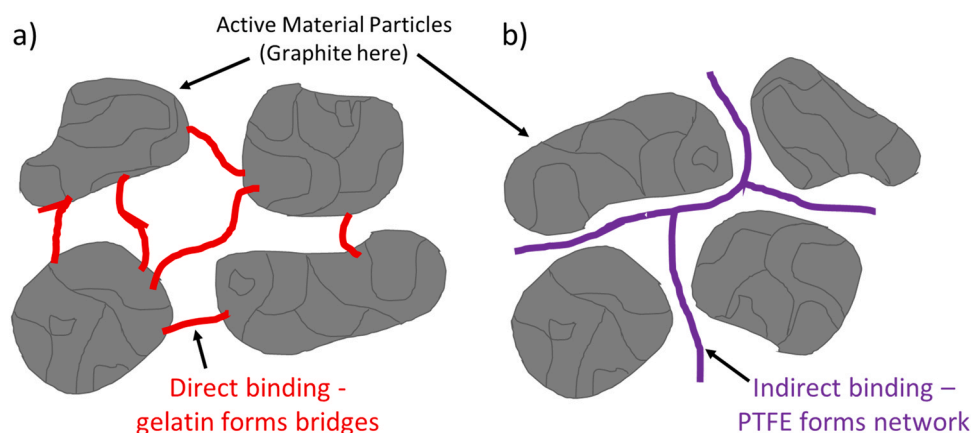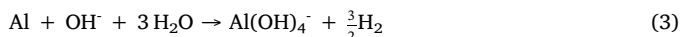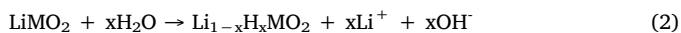

Fig. 4. Diagrams showing the two possible binding mechanisms for composite electrodes. The examples here show graphite particles as the active material. a) Direct binding - adsorption of particles to form interparticle bridges, b) Indirect binding - polymer network is formed and 'traps' active material particles.

the alkalinity of the slurry causes corrosion on the aluminium current collector, producing hydrogen gas via Eqs. 2 and 3. [41,42] Additionally, secondary sources of cracking can be brought about during the drying step, where the elimination and diffusion of the solvent can induce significant internal stress within the coatings, unless a relatively thin wet coating (thickness ca. 200 μm) and low temperatures (20 °C) are employed. [43].

$$LiMO_2 + xH_2O \rightarrow Li_{1-x}H_xMO_2 + xLi^+ + xOH^- \qquad (2)$$

$$Al + OH^- + 3H_2O \rightarrow Al(OH)_4^- + \tfrac{3}{2}H_2 \qquad (3)$$

To replace PVDF, minimise NMP usage and avoid cracking in the electrode coatings, whilst enhancing cathode separation at the end-of-life, novel binder systems different from those currently being investigated for the anode may be required. Miscibility in water, if only partial, is still desirable in reducing the solvent and power requirements of end-of-life processing, but the cracking observed in manufacturing is a direct result of the use of water-based slurries. A possible compromise could involve use of co-polymerisation or blending of hydrophilic and hydrophobic polymers, allowing use of non-aqueous solvents during manufacturing, as well as a route for water-based electrode delamination processes to be used during battery recycling. An example of a possible system that could be employed uses starch and polyethylene to form a polymer blend that could be solubilised in deep eutectic solvents and was found to degrade in boiling water. [44] Usage of a polymer blend or co-polymer similar to this example would still result in a simplified electrode delamination process, similar to that observed when purely water miscible binders are used in the anodes. Incorporation of alternative binders into the cathode, as well as the anode, would simplify subsequent recycling procedures, minimise the use of harmful solvents, additives and high-power consuming processes, as well as attaining better recovery of the cathode active materials, which currently make up the majority of the value of end-of-life batteries.

2.3. Case study

End-of-life battery disassembly has been demonstrated on a commercial scale by the Swiss company Kyburz, who build light weight electric vehicles for private individuals, companies, municipalities, and delivery companies. Their vehicles are powered by LFP cells, and the recycling protocol is claimed to recover > 90% of the battery materials. These materials are then reused in secondary and tertiary applications, such as in 'used vehicles' (~ 85% capacity) and power storage (between 65% and 85% capacity), respectively. [45] Kyburz uses cylindrical cells in a prismatic conformation, encased in a rigid steel casing. This recycling approach is only viable because no structural adhesives are used between cells, instead the internal components are held together via mechanical fixings, allowing for a simpler, albeit manual and more time-consuming, module-to-cell separation strategy. This approach is justified, given the relatively small sizes of the battery packs. It does, however, show that disassembly, rather than shredding, can be used if structural adhesive use is minimised.

Once the cells are separated, they are opened in an inert atmosphere, using a bandsaw to cut off the terminals and battery management system. The electrode roll is ejected from the casing by drilling a hole in the opposite end of the casing material and applying force using a compressed gas. The polypropylene separator between the electrodes is rewound vertically onto a spindle, allowing the anodes to fall in one direction and the cathodes to fall in the other. These electrodes are immersed in water, allowing a complete delamination. While no specific information exists about this process in the literature, this study replicated the process conceptually, where it was found that both the anode and cathode delaminated in under 30 min, with 100% recovery of material. In both instances, it is thought that intercalated lithium reacts slowly with water as there is evidence of gas evolution at both electrodes and the solution pH rises from 3.3 to 5.2 during delamination,

indicating lithium hydroxide formation (Eq. 3). It should be noted that the acidic starting pH arises from the formation of acidic species, such as hydrofluoric acid (equation 5), when the electrolyte reacts with the water. [46] Both observations can be related to the aqueous delamination solution leaching lithium from the active material and this provides a facile method for lithium recovery from the aqueous solution using ion exchange, precipitation, or evaporation of the solvent. The hydrogen forces the active layer to separate from the current collector, but it does not break apart the active material from the binder, so the active layer remains as a continuous phase. While the binder used in the batteries dismantled by Kyburz is unknown, thermal characterisation and IR carried out on commercial LFP cells have shown that they contain CMC/SBR as the binder for both anode and cathode. While this binder can be applied with water during slurry making, washing the electrodes with water is not sufficient to separate the binder from the active material. This residue is only fully removed with thermal treatment. An additional study, conducted by the authors compared the removal of CMC/SBR and sodium alginate (NaAlg) binders from a graphitic active material, after soaking in water and applying 10 s of high intensity ultrasound. Thermogravimetric analysis of the electrode material before and after ultrasound found that only 7.5% of the CMC/SBR was removed, while 76% of the NaAlg was lost. It should be noted that in the case of CMC/SBR, neither polymer was removed preferentially as a two-phase decomposition with the same mass loss ratio of 1:1 for each phase was seen before and after ultrasound processing. The study demonstrates the importance of novel intracellular binders, where the desired binders are soluble in water during both, manufacturing, and end-of-life processing, simplifying the disassembly procedure through further separation of the constituent parts of the battery materials.

While this approach seems applicable to this specific pack type and battery chemistry, there are aspects which could be applicable to other pack form factors. Most automotive packs have small cylindrical cells and, while the structural adhesives prevent segregation of the individual cells, they do provide rigidity and so a bandsaw approach could be used to open and separate cell contents from containers and prevent the cross-contamination observed in shredded materials.

3. Environmental impacts of utilising alternative adhesives

The use of alternative structural adhesives and electrode binders have been identified, both in this work and in previous investigations, to have a significant impact on simplifying battery disassembly. [19,39] Changes to cell design and the use of novel separation techniques, such as electrode ultrasonication, can form purer waste streams during end-of-life processing. [47] In this section, the use of alternative adhesives and binders on the global warming potential and the processing capacity of recycling are compared.

A potential battery dismantling route is shown in Fig. S1. The impact assessments carried out in this work focus on the steps highlighted, as these will be mainly affected by the change in adhesive components. Environmental impact was assessed through global warming potential (GWP), which measures the ability of a greenhouse gas to trap heat within the atmosphere, relative to carbon dioxide, over the course of 100 years. [48] This was measured in reference to the IPCC 2013 impact assessment method, with the results given in kg CO_2-equivalents (kg CO_2-eq), which describes the amount of CO_2 that has the equivalent GWP of an emitted amount of greenhouse gas. [49] Modelling and calculations were completed using Umberto LCA+ (v. 10.0) software and the Ecoinvent 3.7.1 database was used to acquire necessary data regarding the production and distribution of electricity and any solvent formulation.

3.1. Environmental impact of different structural adhesives during module opening

Table 2 shows that the choice of adhesive will influence the module opening time, which affects both the power requirements for module opening (measured in Wh) and the processing capacity. This study

Table 2

Physical properties of the pressure sensitive adhesives used in this study including the dimensions and weight of the adhesives and the time taken for removal.

Pressure Sensitive Adhesive	Dimensions	Weight (g)	Average time taken to remove manually (s)
Glue Dot	0.3 cm radius	0.016	20
Velcro Strip	2 cm × 2 cm	0.434	25
Double Sided Tape	2 cm × 2 cm	0.052	50

assumes that that the modules are opened with robots as shown in Fig. 3, technical information for the robotics was obtained from their data sheet and is listed in Table S1, along with other key parameters used in this assessment. [50] The reference scenario considered here is based on data for the Nissan Leaf module-to-cell disassembly previously assessed by Lander et al. This reference scenario permits comparison between the conventional cell design with structural adhesives, which are assumed to be epoxide based, and the zigzag conformation utilising alternative adhesives. [8] In the case of the alternative adhesives, the time taken for debonding was estimated based upon the manual debonding times in Table 2. When robots were used on cells joined with glue dots the time required for debonding reduced by 25%; it was assumed in these scenarios that the automation of disassembly provided a similar reduction in the time taken. Fig. 5 shows the GWP, energy usage and process capacity graphs obtained for these module opening scenarios and Table S2 gives the data behind this figure.

The first detail that should be noted from this data is the small GWP of this process, regardless of the adhesive used, as it has been shown in g CO_2-eq rather than kg CO_2-eq. In this instance, the low GWP correlates to the relatively low power requirements of the robots in opening the modules and the fact that these values have been determined based on the processing of a single module. The primary difference between the scenarios is the amount of time required to open up the module, which is dependent on the module design. As seen in Fig. 5, a significant reduction in GWP is observed between the reference scenario and the scenarios using the zigzag module design. The time discrepancy between the designs is related to the lack of physical fastenings like screws, simplifying the process required to dismantle the module and subsequently the amount of operation time the robots need for each module. The choice of adhesive used with the zigzag module is also important for minimising robot processing time and the associated GWP. All of the adhesives tested debond on the timescale of a few seconds. However, debonding of the glue dots was shown to be the quickest and hence shows the smallest GWP, at 250% lower than that of double-sided tape and 125% lower than the Velcro strips.

The reduction in disassembly time will not only affect the environmental impact of battery separation but can also facilitate a higher processing capacity for end-of-life battery treatments by improving the rate of battery disassembly. Processing capacity was estimated for each scenario based on their respective processing time and the amount of time required for the robot to reset and be ready to open another module, which was found to take around 20 s for the robots used in this assessment. Less processing time and a higher throughput of modules, paired with purer waste streams acquired through dismantling batteries over shredding, could have a significant impact on the profitability of recycling facilities, just through simple changes to module design and adhesive materials used in manufacturing.

3.2. Environmental impact of alternative binders during ultrasound delamination

The novel ultrasonic delamination technique proposed by Lei et al. was selected as the separation method in this study, to assess the effect alternative electrode binders have on battery disassembly. This has already been shown to have a beneficial technoeconomic analysis compared to many hydrometallurgical processes. Only delamination was assessed, so that the environmental impact of replacing the conventional binders can be emphasised. To obtain the input materials for this process from the opened module, the cells acquired in the previous step would have to be opened before the cell components, i.e. electrodes, separators, electrolyte and packaging, are separated into distinct waste streams, ideally via an automated process such as that demonstrated in the Kyburz process. [51] Cell opening and separation will also contribute GWP to battery recycling and future work will incorporate these steps into a full LCA study.

Two scenarios were investigated for electrode delamination, one being a reference scenario, using PVDF and CMC/SBR as the cathode and anode binders respectively. This scenario was compared to a hypothetical pouch cell using water miscible binder systems in both electrodes. It was assumed that the cathode binder was a hybrid system, like the example described in Section 2.3. Data related to the power, solvent and the time required to delaminate the electrodes was obtained experimentally. Table S3 shows the parameters assumed for this assessment. Electrode delamination of the cathode and anode will result in the formation of four distinct waste streams; the separated anode/cathode active materials, and their respective current collector foils.
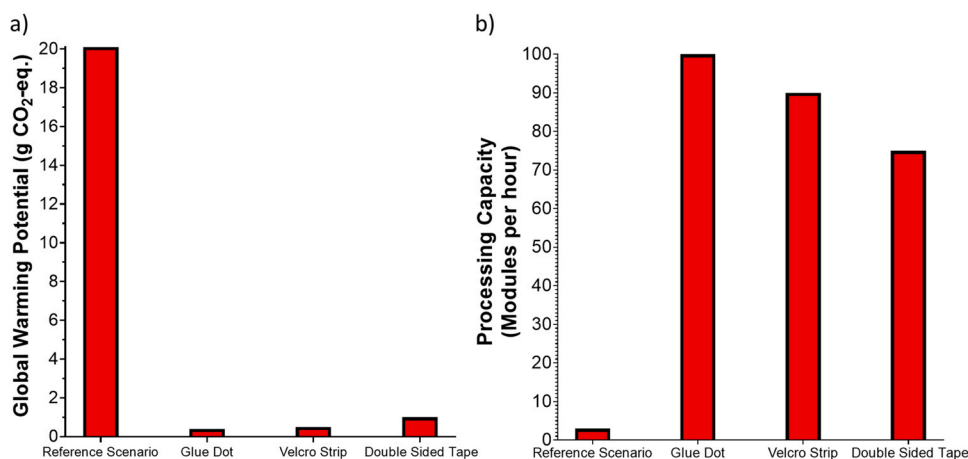

Fig. 5. Graphs showing the changes in global warming potential (a) and processing capacity (b) values corresponding to the opening of a battery module. The reference scenario assumes the use of a conventional module design and adhesives. The other data assumes the use of the zigzag module conformation with three different adhesive mechanisms.
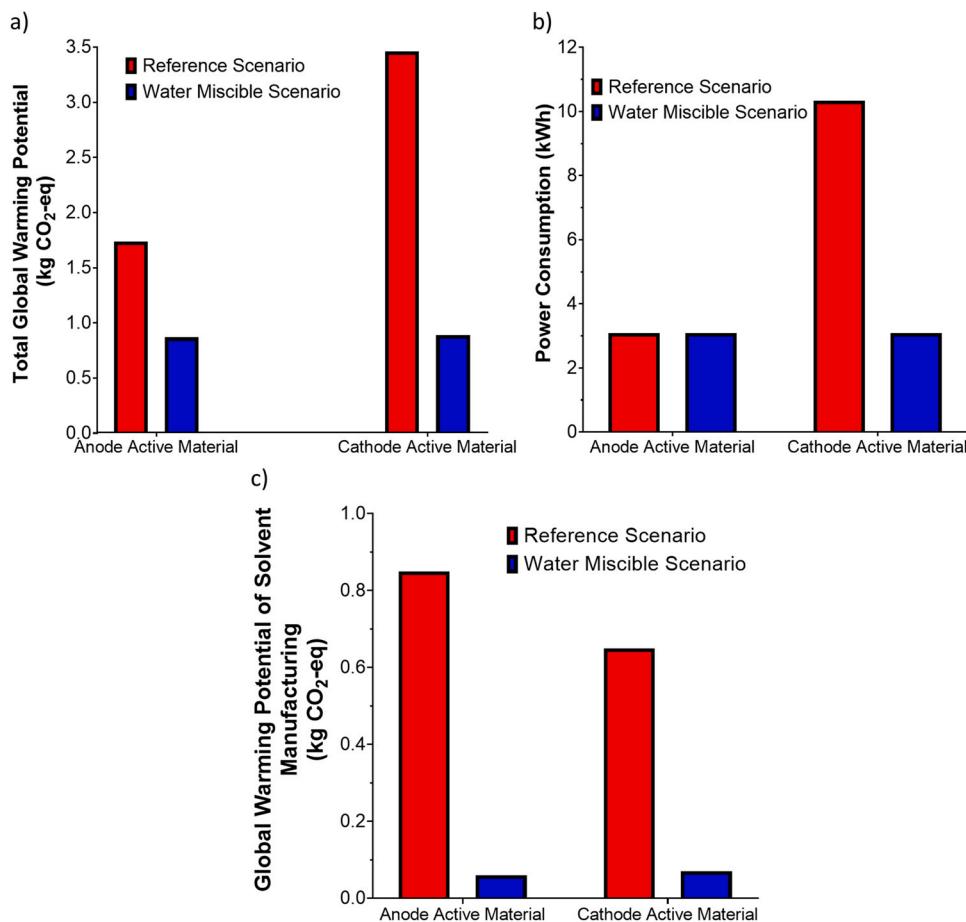
Fig. 6. Graphs showing the changes in global warming potential (a), energy usage (b) and GWP from solvent usage (c) values corresponding to the ultrasonic electrode delamination step. Reference scenario assumes the use of electrodes containing the conventional binders (PVDF and CMC/SBR). 'Water Miscible' scenario assumes the use of hypothetical electrodes containing water miscible binders.

The results from this assessment are given with respect to two of these waste streams, the anode and cathode active materials, given in Fig. 6, while all the data acquired, including that related to the current collector foils, is given in Table S4.

Fig. 6a shows a significant reduction in the GWP of ultrasonic delamination when alternative binders are utilised within the electrodes, with the recovery of both types of active material reducing their environmental impact. Fig. 6b and c show the power requirements and the GWP associated with solvent usage of ultrasonic delamination. This allows determination of whether the changes to the power output of the ultrasound or the solvents and additives used will reduce GWP the most. Since the anode already uses a water miscible polymer, CMC/SBR, the reduction in GWP is not as extensive as that seen for the cathode, where PVDF is utilised. It is predicted that the only contributing factor to the reduction in GWP of the anode is from no longer requiring the solvent additives. This is because, unlike the binder assumed in the water miscible case, the commercial anode materials require additives, as well as water, to allow for complete and efficient delamination via ultrasound. Table S3 gives the type and amount of additives used for each electrode and the associated environmental impact data was retrieved from the Ecoinvent database. Although the impact that changing the anode material has on GWP is smaller than for the cathode, the elimination of additives and associated manufacturing routes can be significant, when dealing with the considerable amount of battery waste which will be seen in the coming years.

It has been shown in previous studies that while ultrasonic delamination is effective at removing the active material from the current collectors, the binders are still adhered to the active material particles, requiring high temperature processing in order to remove the binder. Anodes using CMC/SBR have the same issue as discussed in the aforementioned case study in this paper, where the use of NaAlg left significantly less binder residue when ultrasound is conducted using water as the solvent. This means that, as well as reducing the energy consumption of the disassembly processes, it also reduces the number of steps required, which would be beneficial to the overall economics and GWP.

Overall, when comparing the reference scenarios to the best alternative scenarios, i.e. using the glue dot and 'water miscible' scenarios, the % reduction in GWP in producing the separate anode and cathode material is 150% and 173%, respectively. It is also thought a similar reduction will be observed in processing costs of battery recycling, when comparing the reference and alternative scenarios. It is reasonable to assume that novel cell designs, new structural adhesives and water miscible binders will minimise recycling processing costs, with the aim to bring the cost towards the $45 per pack suggested by Lander et al. [8].

4. Conclusions and future perspective

This study has shown that the biggest challenges faced by the emerging LIB recycling business is the complexity and diversity of the feedstock. The current LIB recycling market is dominated by small packs/cells from consumer electronics which is slowly being matched by automotive production scrap. Recycling will have to deal with legacy vehicles already on the market and new designs unlikely to come to end-of-life for at least 15 years. Future recycling plants will also need to deal with alternative electrode chemistries and structures. These include solid state batteries, nanostructured electrodes, [52] sodium ion batteries, [53] lithium sulfur batteries [54] and binder-less cathodes. [55] Some of these are more problematic than others, for example all solid state batteries (ASSBs) employ lithium anodes, which have adhesive properties that make traditional cutting methods challenging.

Table 3

Techniques which can be employed to open up lithium-ion battery cells once they have been removed from the battery pack structure.

Cell Opening Methods	Advantages/Disadvantages
Ceramic Blade	Mechanical cutting with ceramic blades, is simple. However, the tooling parts may require replacing frequently which adds to the cost and any HF released from the cell will exacerbate wear considerably. Cutting fluids can be used to reduce wear, and to improve throughputs.
Ultrasound	Suited to materials which cannot be exposed to heat, such as plastics which melt. Owing to lack of heat, this process may be the safest option for pouch cells.
Plasma	Requires electrically conductive materials and is often used for metals. However, a significant level of heat is produced that would need to be dissipated during long-term usage.
Laser	In most laser cutting applications, a robot is loading/unloading metal sheets to the bed of a laser cutting machine. Because lasers produce a large amount of heat, this could damage these robotics unless heat dissipation is carefully controlled.
Water Jet	High pressure water jets can be used for a wide variety of materials but may react with or cause contamination of the products if mixed with abrasive ingredients.

Conversely, binderless systems may lend themselves to simple ultrasonic delamination if the porosity is suitable.

Binders and adhesives are one of the most problematic aspects of recycling and the clear take home messages from this article are that the use of:

- larger cells,
- fewer structural adhesives such as epoxy resins,
- fully water dispersible binders,
- reversible physical connectors
- and alternative cell configurations

can all significantly simplify pack disassembly. The case study discussed in section 2.4 shows that cell opening and electrode separation can be achieved and coupled with intelligence-assisted predesign leading to purer product streams. [15,56] The range of opening techniques discussed in Table 3 show that this approach could be tailored to specific battery chemistries. These design aspects may be easier to apply to batteries used for different applications. For example, static packs used for energy storage experience negligible external mechanical stress so adhesion between packs components may be handled differently from those used in motorsport.

Improved battery design, particularly in vehicles results in easier repair and recycle and can results in improved brand reputation and increased residual values. Design for recycling has the potential to create environmental as well as commercial value. This is seen clearly in Section 3.0, where changes to both the adhesives and pack design used were found to have a significant impact on both energy consumption and processing capacity, minimising environmental impact and the cost of disassembly processes. Furthermore, emerging business models such as PAAS (Product as a service), which may aid in advancing circular economies, have the potential to unlock synergistic benefits for producers, if they become responsible for regenerating and recycling products once their initial service life has expired. We have argued that a clear legal extended producer responsibility (EPR) that balances the interests of end-of-life materials recovery with the requirements of second-life, [57] coupled with a servitisation model, [58] is the best route for promoting many of the goals outlined in this paper.

Declaration of Competing Interest

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nxener.2023.100023.

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