

1	To shred or not to shred: A comparative techno-economic assessment of lithium ion
2	battery hydrometallurgical recycling
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13	techno-economic analysis,
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15	Abstract
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17	Present techniques for recycling lithium-ion batteries (LIBs) tend to employ shredding as
18	a preliminary step. This results in size reduction and passivation of reactive components.
19	However, it also delivers lower purity products, decreasing process economics. We
20	propose that disassembly followed by delamination retains product value and simplifies
21	downstream chemistries. A retro-economic analysis shows the theoretical cost of
22	reprocessing for a hypothetical \$100 / kWh battery. Ten different hydrometallurgical
23	approaches to LIB recycling are contrasted through techno-economic analysis of the wet
24	part of the process. We show that shredded material can be recycled into new cathode
25	material with a cost saving of up to 20%. Comparable processes using disassembled cells
26	enable up to 80% cost saving (not accounting for the actual step of disassembling the cell).
27	In the light of these results, we set out the barriers to disassembly of LIB cells,
28	recommending the importance of design for disassembly as key to improving the
29	circularity of LIB supply chains, ensuring that greater value is retained within the system.
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#### 32 **1. Introduction**

Recycling of materials is often essential to creating a sustainable, circular economy and maintaining the availability of resources. Whilst recycling for bulk materials such as steel, glass and cardboard is well established, there is an urgent need to improve recovery rates of processes used to recover strategic elements, critical materials and technology metals that are often finely distributed through modern hi-tech products.

38 Most current recycling processes start with mixed feeds and render material accessible through 39 shredding, by using cutting, pressure, impact or abrasion. The suitably sized pulverised materials can then be separated depending on their physical, optical, magnetic or mechanical 40 properties. Several direct and indirect methods exist and many municipal waste processing sites 41 42 integrate a variety of these techniques to produce bins of segregated waste with varying values 43 depending on the purity of feedstock and efficiency of the segregation steps. The main sorting methods and the properties on which their separation is based are listed in Table 1 (Gundupalli 44 45 et al., 2017; Sommerville et al., 2020; Bi et al., 2020; Bi et al., 2019; Al-Thyabat et al., 2013).

Table 1: Common separation methods, the properties they act upon, and materials theytarget.							
Method	Property	Separates					
Magnetic drum	Magnetic attraction	Ferrous metals					
Eddy Current	Magnetic repulsion	Non-ferrous metals					
Jigging	Density	Metals from plastics					
Hydrocyclone	Density	Organics					
Froth flotation	Hydrophobicity/ density	Hydrophobic from hydrophilic					
Electrostatic	Induced charge	Plastics					

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For mixed streams e.g. from municipal solid waste, shredding is the only method available to liberate/separate complex mixtures, but this tends to yield bins of simple materials of lower purity and lower value e.g. plastics, paper, glass and fabrics. This results in producing new products of lower value, so-called down-cycled materials. From the perspective of the waste management hierarchy, processes vary in their desirability. It is preferable to maintain the quality of materials wherever possible. 53 Materials with unique properties, e.g. ferrous metals, are easy to separate from complex 54 mixtures. A preliminary series of methods are often required to get streams of materials of 55 similar chemistries i.e. metals, organics and inorganics. These material chemistries tend to have 56 significantly different properties in terms of surface wettability, density, charge, and magnetic 57 properties. The success of the liberation/separation methods, however, also depends on how 58 well the materials of different chemistry are separated. This is particularly an issue for 59 composite or bonded materials, where adhesives can change the way in which metals or 60 inorganic compounds behave during separation stages such as froth flotation or eddy current 61 separation. This is a particular issue for technology metals such as those found in printed circuit 62 boards, battery electrodes and composite magnets. Shredding is also an issue with hazardous 63 waste, as dusts are particularly problematic in a working environment. More research into 64 mitigation of these hazards will naturally be necessary (Sommerville et al., 2020).

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#### 66 **1.1 Recycling lithium-ion batteries (LIBs)**

Lead acid batteries are one of the most efficiently recycled products due to their simple design 67 68 and the efficiency of the shred-and-settle method of separation (Gaines, 2014; May et al., 69 2018). Recycling other battery chemistries is much less efficient and accordingly recycle rates 70 are much lower due to economic and efficiency factors (Melin, 2020). A big challenge is arising 71 for lithium-ion battery recycling due to the predicted waste of 300 million tons of LIBs from 72 electric vehicles due to be produced between 2015-2040 (Mossali et al., 2020). There is an 73 enormous amount of potentially valuable resources available within LIBs that are not being 74 used (Diekman et al., 2016). In the UK alone, it is estimated that there will be a cumulative 75 total of up to 106,000 obsolete LIB packs reaching end-of-life by 2040 (Skeete et al., 2020). 76 Reclaiming materials back from LIBs would reduce the reliance on mining for fresh minerals. 77 Some techno-economic analysis of LIB recycling has been carried out (Rohr et al (2017), 78 Steward et al (2019), Spangenberger et al (2018), Ciez and Whitacre (2019)) but these tend to 79 be on specific processes and most of the hydrometallurgical processes start with the concept of 80 battery shredding, followed by black mass (mixed anode and cathode active material) 81 separation.

There is also the potential for LIBs to be used in second-life applications such as stationary grid storage, although the high energy density from chemistries such as NCA (nickel cobalt aluminum oxide) and NMC (nickel manganese cobalt oxide) technologies is not necessarily required. Note that the decision to employ a LIB in a second-life application can introduce a lag of 5 to 10 years before the raw materials are re-introduced to the battery supply chain via
recycling, depending on the application (Kamran et al., 2021).

88 The recycling of LIBs presents great challenges, but also great opportunities (Ghadimi et al., 89 2019; Harper et al., 2019). There are numerous approaches for recycling LIBs but they can 90 broadly be split into three main categories; pyrometallurgical, hydrometallurgical, and 91 physical. These have been reviewed in depth in a variety of recent articles (Sommerville et al., 92 2020; Lv et al., 2018; Or et al., 2020; Assefi et al., 2020). Pyrometallurgy (followed by 93 hydrometallurgical recovery) is favoured for its simplicity. As a technology, it is mature, and 94 it can accept a wide variety of feedstocks with minimal pre-processing. The process is mechanically simple, with little required in the way of disassembly or pre-treatment. The 95 96 challenge with the technology is that several components such as manganese, aluminium, 97 carbon, the electrolyte, polymers and lithium are consumed and go to either energy production 98 or slag. The other metals, such as cobalt, nickel and copper, are recovered as alloys, and 99 additional hydrometallurgical steps are required for their separation (Ekberg and Petranikova, 100 2015; Melin, 2018). Therefore, in terms of both the quantity and value of the materials 101 recovered, these are lower than for hydrometallurgical processes. Furthermore, there is perhaps 102 limited scope for future improvement in the pyrometallurgical treatment of LIB cells 103 (Thompson et al., 2020). Whilst pyrometallurgical recycling of LIBs conserves some of the 104 value of materials in the LIB supply chain, some value is destroyed as materials are lost to 105 states from which they are not economically recoverable.

106 By contrast, hydrometallurgical processes can recover more material from recycled batteries, 107 both in terms of value and quantity, with the advantage of metal extraction occurring in the 108 first phase. There is also the opportunity to recover the material as a precursor for LIB 109 production (Melin, 2018). The full potential of the technology is currently unrealised. More 110 sophisticated approaches to the pre-treatment and disassembly of batteries have the potential 111 to unlock improved recovery rates, which will become increasingly important as the price of 112 LIBs continue to fall and primary supply chains become more agile and nimble, reducing their 113 costs. The promise of improved hydrometallurgical processes is that more value can be retained 114 within a circular LIB supply chain, improving the sustainability of LIB manufacturing by 115 conserving materials, and the energy and resources invested in their production. This review does not intend to compare the economics of Pyrometallurgy and hydrometallurgy as this has 116 117 already been published (Spangenberger et al. (2018)).

#### 119 **1.2 Aim of Analysis**

120 Initially, most approaches to lithium ion battery (LIB) recycling used shredding as the initial 121 stage of waste handling, but this leads to lower purity products which tend to be down-cycled 122 (Melin, 2018). While this is preferable to incineration or landfill, ultimately this continual 123 downgrading of functionality is not sustainable and leads to a linear model of consumption 124 (Baars et al., 2020). Recycling has the potential to be a future source of raw materials for 125 electric vehicle (EV) batteries (Crabtree, 2019) and decrease some of the issues associated with primary extraction of elements such as lithium and cobalt (Alonso et al., 2007; Katwala, 2018; 126 127 Harper et.al., 2019; Bertau et al., 2017, Rajaeifar et.al. 2020). Retaining value in the recycled 128 chain can also ameliorate challenges with price volatility and political instabilities in some 129 supply countries (Mann et al., 2019).

130 Product purity is essential for LIB recycling, as very low levels of contamination could render a product unusable for EV batteries (Li et al., 2017; Rothermel et al., 2018). There is a growing 131 132 school of thought that disassembly can lead to higher purity and higher value products (Harper et al., 2019). The aim of this paper is to perform an economic comparison of the cost and value 133 134 of shredding vs disassembly as methods of recycling using previously published pilot scale 135 studies. The analysis also estimates the permissible costs of recycling a hypothetical \$100 per 136 kWh battery system to achieve gate fee-free recycling. One caveat of the analysis is that labour, 137 equipment and sundry operating costs are omitted. These have been previously modelled 138 (Steward et al., 2019) but, given the future scale will require robots for battery handling and 139 disassembly, previous assumptions are felt to be inappropriate for this analysis and so the 140 results presented are purely comparative to highlight the differences between shredding and 141 disassembly.

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### 143 **2. Economic analysis**

144 The initial part of this analysis starts with the commonly stated premise that a battery system 145 target price of \$100 per kWh<sup>1</sup> enables price parity between electric and gasoline vehicles 146 without subsidies (Harper et al., 2019). This is a useful benchmark as it enables the permissible

<sup>&</sup>lt;sup>1</sup> A stated goal of a number of international programs, including the U.S. Department for Energy's Vehicles & Technology Office sub-program <u>https://www.energy.gov/eere/vehicles/batteries</u>, the Faraday Battery Challenge <u>https://www.cenex-lcv.co.uk/storage/seminar-</u>

programme/sessions/presentations/tony\_harper\_introduction\_to\_the\_faraday\_battery\_challen ge\_1537351167.pdf, Page 10

147 costs to be estimated knowing the value of the product (retro-economic analysis), not only of the cost of the individual components, but also the target costs for recycling end-of-life cells. 148 149 At the \$100 per kWh target price for automotive battery systems, mass production enables 150 economies of scale to be achieved in cell manufacture. Given that the energy density of LIBs is in the range of 100-265 Wh kg<sup>-1</sup>, the cost of the battery itself needs to be in the range \$10 to 151 26.5 kg<sup>-1</sup>. In this range, the lower cost likely refers to chemistries such as lithium iron phosphate 152 153 (LFP), whereas the higher cost range refers to lithium cobalt oxide (LCO) and the different 154 NMC chemistries. As Wang et.al. (2014<sup>2</sup>) observe, LFP cathode batteries have materials values 79% less than cobalt containing formulations. 155 Figure 1 shows the composition of a LIB using NMC111 (1 nickel: 1 manganese: 1 cobalt, all 156

157 as oxides) as a benchmark, both from a mass and a cost perspective. Assuming that economies 158 of scale apply equally to all materials, then typically 65 to 70% of the cost lies with the cathode 159 material and this explains why many recycling processes have focused on this stream as the 160 important one to recover. The aim of this article is to probe deeper into the economics of the 161 recycling process and determine whether gate fee-free recycling is viable.

162 Patry et al. (2015) devised an algorithm for determining the overheads and other fees in cell manufacture. They also calculated the process costs, which were in the range 22 to 24% of 163 164 overall costs, the remainder of which relate to the material costs and overheads. It was found 165 that the cost per kWh for LFP (\$402) was greater than NMC (\$307) and these are significantly 166 higher than the target of \$100 per kWh for automotive battery systems. These figures, however, are predicated on relatively high cathode material costs. Cost of the cathode active material 167 168 varies quite considerably depending on the cell chemistry, the purity and the scale of 169 production. Spangenberger et al. (2018) stated that the costs of the common cathode 170 chemistries were \$62 (LCO), \$42 (NMC811) and \$32 (LFP) per kg in 2018. Prices in July 171 2020 span a wide range depending on purity and scale, but prices as low as \$16/kg (NMC811) 172 and \$10/kg (LFP) are quoted, showing that these could take material costs into the realm of 173 \$100 per kWh overall price for an automotive battery system if the scale and purity were 174 appropriate.



Figure 1: Composition of a typical LIB currently being recycled, as a function of cost and mass. Raw data taken from Patry et al. (2015)

Patry et al. (2015) showed that the percentage of material costs were similar between battery chemistries (NMC 57% vs LFP 54%). Using this, an approximate cost for battery material and cathode active material can be calculated for the theoretical \$100 per kWh cell. These approximate calculations are shown in Table 2. If the cost of the electrode materials are in the range \$4.42 - 11.73 per kg of battery and the cathode material makes up 25% of the cell by mass, then the cost of the cathode material should ideally be in the range \$16 - 44 per kg. This shows that the value of the cathode material is in the range of 44% of the total battery cost.

183 In order for recycling of LIBs to be economically viable, without significant gate fees, the 184 overall processing costs need to be collectively cheaper than buying virgin materials and the 185 recycled material needs to have the same performance as unrecycled material. Table 2 shows 186 a breakdown of the material costs in a battery, where the battery cost per kg was calculated 187 earlier and shown to be a range of \$10-26.50 /kg due to the costs of LFP and LCO. All materials 188 (i.e. active materials and other materials) are 56% of the total battery production cost, where 189 the remaining cost lies with overhead fees and process costs. Of the materials costs, 79% of 190 the cost comes from the electrode materials, with the cathode material contributing to 68% of 191 the electrode material costs. Table 2 shows that if the process only recovers cathode material, 192 then the total processing costs (energy + labour + chemicals + on costs) will realistically need 193 to be in the range  $\frac{2-6}{\text{kg}}$  of battery to yield a recycled product which is significantly cheaper 194 than buying a product containing virgin material. This range is an assumption of a reasonable

195 threshold whereby the cost of recycling should be at least half the value of the electrode materials, to ensure some profit gain. It is also to be noted, that as the battery manufacturing 196 197 industry develops, it is to be anticipated that further cost reductions will result due to economies 198 of scale and efficiencies through learning. Of course these are tensioned against supply and 199 demand in the marketplace, but on an assumption that the industry will work to develop cheaper 200 batteries, and develop the supply chain to produce the resources to produce these batteries more 201 cheaply, the LIB recycling industry will need to show commensurate improvements in 202 economic efficiency to compete as a source of materials supply. The next section attempts to 203 calculate a value for various recycling processes described in the literature depending on 204 whether the feedstock material was first shredded or obtained by dismantling the module and 205 cell.

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Table 2: Approximate cost ranges for cells in a theoretical \$100 per kWh battery system.							
	Cost (%)	Cost per kg of battery (Steward et al., 2019)					
Battery Cost		\$10 - \$26.50					
All materials	56%	\$5.60 - \$14.85					
Electrode materials	79%	\$4.42 - \$11.73					
Cathode material	68% of electrode materials	\$3.80 - \$10.10					
Anode material	9% of electrode materials	\$0.50 - \$1.33					

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## 209 2.1 Cost of Recycling

The analysis in Table 2 is useful in that it sets approximate costs for components which are chemistry independent. The figures are also important as it gives a framework around which

- the economics of recycling can be based, assuming a gate fee-free recycling process.
- 213 There are several LIB recycling plants around the world with capacities of 1000-9000 tons per
- annum, and in 2019 the total world capacity was ca. 94,000 tons per annum (Steward et al.,

215 2019). There are a number of plants in the planning stage, including a plant with a 20,000-tons 216 per annum shredding circuit, scheduled to open in Germany by 2021 (Neometals, 2020). 217 Several of these were already existing plants for Co and Ni recycling and have been repurposed 218 for LIB recycling. There are numerous recycling plants being developed in China, one of these 219 is by Brunp, who announced the opening of a \$178M plant in the Hunan province in February 220 2020, capable of processing  $> 100\ 000$  tons per annum. Brunp is a subsidiary of the Chinese 221 battery manufacturer CATL and it seems logical that manufacturers and recyclers will be co-222 located.<sup>2</sup>

223 Numerous recycling processes have been reported in the literature and in this study, 10 224 flowsheets (shown in Table 3) have been shortlisted for more detailed analysis. Note that the 225 extensive methodology for this analysis is given in the supplementary information. Each has a 226 series of leaching and precipitation steps and the aim is to use current energy and chemical 227 processes to determine theoretically how close these come to 2-6/kg of battery. Half of the 228 processes use whole cells which have been through shredding, and half have electrodes which 229 have been separated, and the aim of this analysis is to show whether there is an economic 230 advantage to separation prior to leaching.

231 The purpose of this comparative techno-economic assessment (TEA) is to compare the 232 efficiency of various hydrometallurgical recycling processes in the literature, using data 233 including process time, energy requirements such as electricity used for heating and agitation, 234 process cost, net revenue, starting material, leaching efficiency, recovery efficiency, purity of 235 final products, and process complexity. The costs of the leaching and recovery phases are 236 reported separately. Estimates of gross profits are provided as comparators, but do not take into 237 account operating, labour, loans and investments, repayments and insurance, which will be 238 similar irrespective of the process. These CAPEX (Capital Expenditure) and OPEX 239 (Operational Expenditure) costs have previously been modelled (Rohr et al. 2017). Of these, 240 labour is one of the most difficult to estimate since the scale of automotive recycling that will 241 be required by 2035 will necessitate considerable automation. Disassembly from packs into 242 modules (and modules into cells) will most probably use robotic manipulation. The extent of 243 this is difficult to gauge at the moment but will apply equally whether dismantling or shredding 244 are used. The aspects needed to enable disassembly are covered in a recent review (Thompson

 $<sup>^{2}</sup>$  That said, the economics of recycling are more generally more sensitive to the distance between sources of waste and the recycler, than transport onwards to final manufacture.

- et al., 2020). An assessment of the automation potential of electric vehicle battery disassembly 245
- 246 has recently been published (Hellmuth et al. (2021))
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Table 3: Summary of chosen hydrometallurgical processes from literature.										
	Shredding									
#	Pre-treatment	Dissolution process	Recovery process	Literature reference						
1	Crushing, wet scrubbing and drying	Reductive leaching	Precipitation of Mn and Fe, solvent extraction of Cu and Co, Co pptd.	(Dutta et al., 2018)						
2	Provided by e-waste company	Reductive leaching	Flotation and precipitation	(Huang et al., 2016)						
3	Shredding, sieving, drying and calcining	Electrochem leaching	Electrowinning of Mn and Co	(Prabaharan et al., 2017)						
4	Crushing, sieving, grinding, alkaline leaching, reduction roasting	CO <sub>2</sub> /H <sub>2</sub> O leach of Li, acid leach of NMC	Evaporation of Li filtrate, solvent extraction of Ni, Co and Mn	(Hu et al., 2017)						
5	Wet shredding, flotation, sieving, calcining	Leaching	Precipitation of Co and Mn	(Barik et al., 2017)						
		Disassen	nbly							
6	Discharge, disassembly and crushing	Reductive leaching	Co-precipitation after calibration of Co, Mn and Ni	(Kim et al., 2014)						
7	Discharge, disassembly, cathode separation	Reductive leaching	Co-precipitation	(Gao et al., 2017)						
8	Discharge, disassembly, separation, cathode calcining	Reductive leaching	Co-extraction of Mn, Co and Ni, co-precipitation of Li and co-precipitation of NMC	(Yang et al., 2017)						
9	Disassembly, mechanical separation	Delamination	Sieve	(Marshall et al., 2020)						
10	Disassembly, high power mechanical separation	Delamination	Sieve	Unpublished						

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252 Input material for each process is defined as one of the following:

Process 1 (Dutta et al., 2018); Process 2 (Huang et al., 2016); Process 3 (Prabaharan et al., 2017); Process 4 (Hu et al., 2017); Process 5 (Barik et al., 2017); Process 6 (Kim et al., 2014); Process 7 (Gao et al., 2017); Process 8 250 (Yang et al., 2017); Process 9 (Marshall et al., 2020); Process 10 (University of Leicester, unpublished) 251

- 253 • *Cathode* – comprising of the active material, binder, conductive additive and Al current 254 collector;

255 • *Cathode black mass* – comprising of the active material, binder and conductive additive 256 (the current collector has been removed by means of chemical or mechanical 257 delamination);

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• *Cathode calcined black mass* – comprising of the active material only (the black mass 259 has been calcined at high temperatures to remove the binder and conductive additive); in which the black mass and calcined black mass will contain Al impurities. 260

261 These starting materials will undoubtedly affect recycling processes as the valuable active 262 material becomes more dilute in the waste stream. For example, liberation of the black mass 263 from the current collector prior to leaching may be beneficial in that the recovered active 264 material has a higher purity; however, the delamination step could add extra cost and time. Predischarging the cells will add an extra layer of safety, but also result in cost implications 265 266 (Christensen et al., 2021). This TEA will compare the cost and value of comparable literature 267 processes to identify why there is interest in disassembling cell components prior to 268 delamination rather than shredding them.

Table 4 shows the conditions for the 10 processes shown in Table 3 using the methodology 269 270 described in the supplementary information. Whilst most of these processes use common 271 inorganic acids, the costs per kg of battery differ significantly due to the solid: liquid ratio 272 (S/L), the time of extraction and the temperature of the process. From a leaching perspective, 273 it can be seen that all processes except 7 can leach the material for the theoretical \$2-6 / kg cost 274 target. This is due to the high costs of formic acid and redox agent, in combination with a low S/L ratio. This shows that only the cheapest aqueous lixiviants are viable on shredded material. 275 276 Note that none of the subsequent calculations includes labour or on-costs, which will change 277 the viability depending on the scale of the process.

278 Recovery of the material from solution is relatively similar across the 10 processes evaluated 279 here, and most involve precipitation and calcination. The cost of these are approximated using 280 a similar process to that adopted in Table 4 and the details are again shown in the supplementary 281 information.

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*Table 4: Leaching conditions and costs per kg of input electrode material for 10* hydrometallurgical processes.

Shredding									
Process no.	Leach feed	Acid (M)	Redox agent (M)	T (°C)	t (h)	S/L (g/L)	Leaching (\$/kg)		
1	Cathode black mass	H <sub>2</sub> SO <sub>4</sub> (2)	H <sub>2</sub> O <sub>2</sub> (4.26)	30	3	75	2.32-2.96		
2	Cathode black mass	HCl (6.5)	H <sub>2</sub> O <sub>2</sub> (6.4)	60	2	200	1.41-1.86		
3	Calcined black mass	$H_2SO_4(2)$	Electrolysis, 400 A/m <sup>2</sup>	25	3	75	0.93-0.94		
4	Calcined black mass	H <sub>2</sub> SO <sub>4</sub> (3.5)	-	85	3	200	ca. 0.16		
5	Calcined black mass	HCl (1.8)	-	50	1.5	200	0.08-0.10		
			Disassembly						
6	Cathode black mass	H <sub>2</sub> SO <sub>4</sub> (2)	H <sub>2</sub> O <sub>2</sub> (2.13)	60	2	100	0.99-1.24		
7	Cathode	Formic (1)	H <sub>2</sub> O <sub>2</sub> (6)	60	1	50	5.80-7.38		
8	Calcined black mass	H <sub>2</sub> SO <sub>4</sub> (4)	H <sub>2</sub> O <sub>2</sub> (2)	90	2	125	0.88-1.08		
9	Cathode	Oxalic (0.5)	0.004	50	0.08	1000	0.28-0.38		
10	Cathode	NaOH (0.1)	0.73	25	0.008	1000	ca. 0.16		

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The data in Table 5 show that these costs are typically less than 1 / kg of material and so comparable or less than those listed in Table 4. The leaching and recovery steps combine to give costs of approximately 1-2 / kg as shown in the final column, which are mostly below the targeted 2-6 / kg cost.

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Shredding									
Process	Step	T (°C)	t (h)	Cost (\$/kg)	Reusable solvent cost (\$/kg)	Total recovery cost (\$/kg)	Total recycling cost (\$/kg)		
1	Precipitation	25	4	0.13-0.14	-	1.09-1.18	3.41-4.14		
	SX	25	0.3	0.97-1.04	104 - 458*				
2	Precipitation	25	10	0.68-0.71	-	0.96-1.05	2.36-2.92		
	Flotation	25	0.5	0.28-0.35	-				
3	Precipitation	25	1	0.76-0.83	-	1.26-1.35	2.19-2.29		
	Electrowinning	80	1.4	ca. 0.51	-				
4	SX	25	0.4	ca. 0.80	2.36-2.85	ca. 0.80	ca. 0.96		
5	Precipitation	25	2	0.20-0.27		0.20-0.27	0.27-0.37		
			Ι	Disassembly					
6	Precipitation	25	24	0.12-0.14	-	ca. 0.46	1.44-1.70		
	Calcination	450, 900	27	ca. 0.33	-				
7	Precipitation	25	24	0.57-0.71	-	0.90-1.04	6.70-8.43		
	Calcination	450, 900	27	ca. 0.33	-				
8	Precipitation	25	24	0.21-0.25	-	0.66-0.70	1.54-1.78		
	SX	25	0.1	ca. 0.13	5.33-11.37				
	Calcination	450, 900	27	ca. 0.33	-				
9	Calcination	450, 900	27	ca. 0.33	-	ca. 0.33	0.61-0.70		

Table 5: Recovery and regeneration costs per kg of input electrode material for the 10processes shown in Table 3 (See supplementary information for details).

10	Calcination	450, 900	27	ca. 0.33	-	ca. 0.33	ca. 0.49

\* Note that no loss of solvent is assumed. Even a 1% loss will significantly affect process
economics, due to the high price of organic solvents. SX = Solvent extraction

### 292 **2.2 Value of products**

293 Whilst the leaching costs are mostly compatible with the overall total processing cost of 2 -294 6 / kg of battery described above, this does depend on attaining a product purity and activity 295 which is compatible with unused materials listed in Table 2. This clearly depends on the form, 296 purity and yield of the products from the recovery stage. Table 6 lists the final products reported 297 from the literature for the ten processes listed in Table 3 and where available, their recovery 298 and purity. From these, approximate values have been estimated for the products from each 299 stream assuming that the materials recovered are active as battery materials, or if they are 300 recovered as sulfates or other intermediates, that they can be converted to active materials for 301 negligible cost. Using this approach the gross profit of material can be assessed by comparing 302 this to the original material. From this, a percentage saving can be calculated for recycling with 303 respect to using new material. There is a large variation in gross profit values in Table 6. This 304 reflects the average final product costs (i.e. the average recommended retail price of final 305 products from the recycling process), which result from a range of purities and is therefore 306 appropriate for this analysis.

Spangenberger et al. (2018) used a similar approach to that used above to compare pyrometallurgy, hydrometallurgy and direct recycling and found a cost saving of 6, 13 and 27% respectively compared to using uncycled material. The calculated hydrometallurgical value is similar to the values listed in Table 6 for processes 1-5. This shows the commonality with our approach, but also underlines the extra value that could be obtained by electrode separation. Spangenberger et al. (2018) found that it was only with LCO that the cost saving was in the range 38 to 43%.

From the data in Table 6, it can clearly be seen that the economics of the process depends not only on the cost of the leaching and recovery, but crucially on the form of the product and its yield and purity. The cost savings listed in Table 6 are similar to those shown by Steward et al. (2019) and are given a range to account for the different battery chemistries. This shows that the cost of hydrometallurgical recycling can deliver new material which could potentially be used in batteries for a lower cost than using virgin material. The main caveat for this approach is whether the recovered material demonstrates the same activity as uncycled

- material. Whilst this has been demonstrated on a small scale, it has not been applied due to the economies of scale required to achieve costs similar to those shown in Tables 4-6. The cost savings also depend on minimising contamination from other metals, including copper and aluminium from the collector materials.

Table 6: Final products and net profits of 10 hydrometallurgical processes where thebattery cell is shredded or disassembled.								
		Shredding						
Process no.	Final products (purity, %)	Recovery (%)	*Gross Profit (\$/kg battery)	% cost saving of recycling				
1	MnO <sub>2</sub> , <u>Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, CuSO<sub>4</sub>,</u> CoSO <sub>4</sub> (< 98), <u>Li<sub>2</sub>SO<sub>4</sub> (aq)</u>	Co: 98, Cu: 100, Li/Mn/Fe (assum.):80	-0.19-0.94	-2-9				
2	MnO <sub>2</sub> / <b>Mn<sub>2</sub>O<sub>3</sub></b> (99), <u>Li<sub>3</sub>PO<sub>4</sub></u> (99), FeCl <sub>3</sub> (98)	Li/Mn: 81, Fe: 85	0.19-1.35	2-13				
3	<b>Co</b> (99), MnO <sub>2</sub> (96), <u>Li<sub>2</sub>CO<sub>3</sub></u>	Co: 97, Mn: 98, Li (assum.): 80	1.31-1.61	13-16				
4	$\frac{\text{Li}_2\text{CO}_3(100)}{\text{CoSO}_4(100)}, \text{MnSO}_4(100), \text{NiSO}_4(100)$	Li: 85, Ni: 97, Mn: 99, Co: 98	0.58-1.81	6-18				
5	<u>Cu(OH)<sub>2</sub>, Al(OH)<sub>3</sub></u> , <b>CoCO</b> <sub>3</sub> , <u>Li<sub>2</sub>CO<sub>3</sub>, NaCl, </u> MnO <sub>2</sub> /Mn <sub>3</sub> O <sub>4</sub>	Mn: 95, Co: 90, Li/Al/Cu (assum.): 80	0.94-1.87	9-19				
		Disassembly						
6	Li <sub>2</sub> CO <sub>3</sub> , NMC111, <u>Al(OH)</u> 3	Li: 80, Co/Mn/Ni: 100 Al (assum.): 80	3.05-5.37	31-54				
7	<u>Li<sub>2</sub>CO<sub>3</sub> (99.9),</u> NMC111	Li: 98, Co/Mn/Ni: 99.9	2.66-5.27	27-53				
8	NMC111, mixed hydroxides, Li <sub>2</sub> CO <sub>3</sub>	Ni: 85, Mn: 100, Co: 99, Li (assum.): 80	2.06-3.70	21-37				
9	LMO, <b>LNCA</b> , Al	LMO (assum.): 95 LNCA (assum.): 95 Al: 100	3.40-8.04	34-80				

10	NMC, <b>LMO</b> , Al	NMC (assum.): 95	4.80-8.51	48-85
		LMO (assum.): 95		
		Al: 100		

Underlined products are not included in the calculations. Products in bold contributed the most
to the cost savings for that particular process. \*Gross profit = final product value minus cost of
leaching and recovery (Note labour costs and overheads omitted).

329

330 Recovery of lithium is an important consideration and although Figure 1 shows that the 331 conducting salt is a significant cost of the overall cell costs, recovering lithium salts does not 332 necessarily help with the economics of the overall cathode recycling process. Most simple 333 lithium salts cost in the region \$5-7 / kg. The cost of producing lithium hexafluorophosphate 334 from lithium carbonate or lithium chloride was determined to be in the range \$20-30 / kg 335 (Susarla and Ahmed, 2019), so recovery of lithium in the form of a chloride or carbonate does 336 not significantly improve the economics of recycling due to its relatively low mass fraction and 337 the comparatively high cost of converting it to the hexafluorophosphate.

This section has confirmed that hydrometallurgical processing of end-of-life LIB material can be carried out without the need for gate fees, providing that the scale is large enough to reduce the labour costs and overheads and the material recovered and regenerated is approximately as active as the initial, uncycled material. The results for the shredded material correlate with the previous economic estimations by Spangenberger et al. (2018).

343

#### 344 **2.3. Shredding vs. disassembly**

345 The case for shredding is compelling; almost all municipal waste recycling starts with size 346 reduction. It averts difficulties associated with product opening, and all products can be 347 handled by the same method. The only drawbacks are the major ones of having a more complex separation process resulting in less pure product streams; it is mechanically simple, but 348 349 chemically more complicated downstream. The case for disassembly is naturally the opposite: 350 simpler separation, purer products, but it is more complex and potentially more hazardous to 351 open cells. The case against disassembly is further complicated by pack and module design. It 352 has been shown that the disassembly of a battery from an Audi Q5 hybrid vehicle involved 24 353 steps (Wegener et al., 2014). Manual disassembly of a Nissan Leaf pack can take 2 h even for 354 a skilled handler without any module or cell testing, which will increase the disassembly time. 355

356 The controlled dismantling and disassembly of LIBs has many advantages over shredding of 357 the components. Shredding is commonly seen in the processes developed by Toxco, Recupyl 358 and Lithorec that recycle LIBs. Although these processes were not originally designed for 359 recycling LIBs they have been adapted due to increased demand (Or et al., 2020; Marshall et 360 al., 2020). However, these existing processes often focus on the recovery of high value 361 materials within the cells, predominantly cobalt, with other materials being lost, either 362 unrecovered or to mixed waste streams. Valuable metals are collected as molten alloys that 363 require further treatment through hydrometallurgical methods. Lithium is lost in the slag along 364 with other oxides and gases. With the emergence of new cathode cells that move away from 365 LCO (LiCoO<sub>2</sub>) majority cobalt cathodes (largely used in the first-generation of electric vehicle 366 LIBs but still to some extent in China) and advance towards mixed metal cathodes, it is vital 367 that efficient recycling processes for all LIBs is established. Disassembly aims to recover 368 materials and components of all value for reuse or recycle, resulting in streams of high purity 369 materials.

370

371 Studies by Marshall et al. (2020) focus on the manual dismantling of cells, investigating the 372 various processes needed for component and material recovery, keeping in mind health and 373 safety considerations at each stage. It is important to note that, for future success with LIB 374 recycling and recovery, the potential for these processes to become automated is fundamental 375 to reduce operational costs and time. In order to achieve a circular economy, it is crucial that a 376 well-understood and safe process for dismantling LIBs is established and endorsed. In a truly 377 circular economy, it would be beneficial to recover the materials in high purity waste streams 378 for reuse and recycling of components, consequently dismantling can have advantages over 379 shredding.

380 Table 6 shows that the cost savings for disassembled cells are always comparable or larger than 381 cells which have been shredded. This is due principally to the purity and yield of the products 382 but also to the simpler flowsheets. Of the processes studied, the more economically beneficial 383 flowsheets have fast delamination catalysed by ultrasound, with high solid: liquid ratios 384 (Processes 9 and 10). In both of these cases, much shorter times are required to process the 385 material, significantly increasing the space time yield. The shorter process times also enable a 386 semi-continuous process to be operated, with the lixiviant being re-used. Since these 387 parameters were shown to control the costs in Tables 4 and 5 it is not surprising that both of 388 these processes have lower operational costs than those for the shredded material. To achieve

the optimum flow sheet from an economic perspective it is important to have fast delamination, such that a continuous flow process can result and the lixiviant can be recycled. Suppressing the digestion of the current collector is important for product purity, enabling lixiviant reuse and increasing process kinetics.

393 Shredding of the cells rather than disassembly forms a mixture of all the materials encased in 394 the pack. Consequently, many further processes are needed to separate all the different 395 materials. The Lithorec process uses electrical, mechanical, mild thermal and 396 hydrometallurgical treatments to recover the valuable materials from the batteries (Harper et 397 al., 2019). The anode and cathode materials present the biggest challenge as they can consist 398 of complex chemistries with various chemical components, in which the design, even at cell 399 level, is varied between each manufacturer. Contamination of the material pools occurs, and 400 crucially only the valuable materials are recovered, meaning the remaining materials such as 401 plastics and lithium are lost in mixed streams of waste that require further recovery methods to 402 reclaim. Disassembly as described by Marshall et al. (2020) takes a different approach to 403 reclaiming the materials through pouch cell disassembly. The described method is optimised 404 for the cells dismantled (Nissan Leaf) but some of the general principles described can be 405 applied to most cells. Using this method, Marshall reported that on average a total of 80% and 406 77% of material was recovered from the quality control reject cells and EoL cells, respectively.

407

#### 408 **3. Barriers to disassembly**

409 The main problem with disassembling the battery for reuse or recycling is the sheer complexity 410 of the battery. The high power originates from a large surface area of active material and 411 numerous cells in series. The number of individual cells that make up a module and pack vary 412 depending on the manufacturer. The first-generation Nissan Leaf contains 192 pouch cells -4413 cells in each module – whilst the Tesla S model contains 7104 cylindrical cells – 444 cells per 414 module (Harper et al., 2019). This large difference in the number of cells contained in a pack 415 highlights the complexity of the disassembly challenge. Even with the lower figure, the Nissan 416 Leaf model contains 192 individual cells that need to be dismantled to separate the internal 417 components and materials into pure streams of waste. The 48 modules also have to be separated from their associated clips, screws and glues to liberate the cells. The issue of how to safely 418 419 and rapidly dismantle the pack, module and cells is not trivial. The prevalence of fluorine-

- 420 containing compounds in the cells, and the potential presence of hydrogen fluoride (HF) in the
- 421 spent cell causes health and safety issues with handling the materials (Harper et al., 2019).
- 422

423 The shell that encases the pack needs to be robust to ensure it can protect the internal 424 components when the electric vehicle is operational. The outer pack design is not built with 425 recycling at EoL in mind, so the procedures involved in opening them can be long and time-426 consuming due to the many steps needed to dismantle, but also to minimise the risks of short 427 circuiting causing potential damage to the cells, fires, and harm to the dismantler. The number of cells increases the workload of disassembly, as more cells are required to be opened to obtain 428 429 pure waste streams, and this increase in workload could increase the labour costs, and critically 430 drive the cost of processing towards the higher end of the \$2-6 per kg of battery range.

Another issue concerning the way that the cells and modules are assembled into packs that is challenging for disassembly is the glue that holds the cells and modules together. Understandably this glue is an essential component in the building of the pack and ensures rigidity whilst the car is in service. The cells are hermetically sealed to prevent the contents of the cells being exposed to moisture. The glues are commonly dissolved using organic molecular solvents. However, this method does not allow dismantling to be a viable recycling method due to the nature of the solvent and the time required.

438 Removal of the binder – the component that holds the active material to the current collector 439 foil in the electrode – is another challenging aspect in disassembly of batteries. Traditionally, 440 PVDF (polyvinylidene fluoride) is used in LIBs due to its good electrochemical stability, 441 binding capability and the ability to absorb electrolyte for facile transport of Li to the active 442 material (Chou et al., 2014). Despite its advantageous properties, which makes it a first choice 443 when considering the electric vehicle in service, at end-of-life, PVDF removal via pyrolysis or 444 dissolution using NMP (N-methyl-2-pyrrolidone) produces toxic waste. Harper et al. (2019) 445 suggest that some manufacturers are moving away from PVDF binders, nevertheless this does 446 not compensate for the existing batteries that already contain PVDF (Harper et al., 2019). 447 Therefore, a viable route to remove PVDF must be found in order to process the EoL batteries 448 where PVDF is already in the chemistry. As PVDF is insoluble in water, organic molecular 449 solvents need to be used. NMP is used to process and separate PVDF from the black mass. In 450 terms of sustainability and from a green chemistry perspective, it is inadequate, as during the

451 process, the decomposition of the PVDF and solvent delamination (using NMP) produces HF, 452 a gas which falls under current REACH regulations (European Commission, 2006). 453 Additionally, organic solvents are costly and dangerous to handle and require specialist 454 engineering for plants (to prevent side reactions, etc.). This results in greater upfront capital 455 expenditure. When Li and PVDF come into contact, the reaction between the 2 is exothermic 456 and could potentially cause thermal runaway, therefore it is essential that an alternative to the 457 PVDF binder is found (Nirmale et al., 2017). Suggestions by Marshall et al. (2020) emphasise 458 the potential of using a solvent stripping process to remove PVDF from anode and cathode 459 materials using a green solvent. It is vital that heat treatment is avoided when attempting to 460 reclaim PVDF to prevent the formation of HF. Alternatively, other methods may need to be 461 investigated to recover the binder. It is important to note that the process of binder removal and 462 recovery will be unique to every battery as various binder materials are used. As manufacturers 463 move away from PVDF to alternative binders, the binder removal process could simplify if 464 less harmful chemicals are needed to remove and recover the material, meaning that 465 considerations of secondary waste products, such as the liberation of HF, are not necessary.

The wires that connect the cells together add another level of complexity to manual 466 disassembly, as seen in Figure 2. The wired network runs throughout the pack at every level 467 and is essential to the function of the battery. It is clear from the diagram in Figure 2 that this 468 469 network is highly complex, and disassembly is a time-consuming laborious process to ensure 470 the correct wires are disconnected at the appropriate time. The wired network within the battery 471 pack will differ depending on the manufacturer. Automation of this part of the battery 472 disassembly would be extremely challenging as robots work from automated programme functions. The condition of the wires could also differ in the same model of battery depending 473 474 on the life the battery has had; some wires may be broken in some cells, whilst others appear 475 fairly new and unused, again depending on the life the battery had prior to the need to 476 disassemble.



478 Figure 2: A schematic diagram showing the structure of a battery pack, module and cell and
479 the challenges presented at each stage for battery disassembly (Expanded from Harper et.al.,
480 2019)

- 482 Underlying all of these design features that make disassembly challenging are the safety issues483 and considerations that need to be made when disassembling the pack. This is ultimately the

484 biggest barrier to making disassembly a viable recycling route, especially when considering 485 scaling the laboratory process up for industrial application. The packs are designed to ensure 486 safety in the battery when the vehicle is in service, but this is at the cost of recycling efficiency. 487 Currently dismantling has to be done in an inert atmosphere, usually a glove box filled with 488 argon, to ensure the cell contents does not come into contact with moisture. The potential that 489 the lithium salt (LiPF<sub>6</sub>) could come into contact with water is significant, potentially resulting 490 in catastrophic effects if it does occur, including the generation of HF - a known product of 491 electrolyte degradation on exposure to humid air (Lebedeva and Boon-Brett, 2016). Chemical 492 additives employed to improve the performance, longevity or safety of LIBs can also form up 493 to 5% of the electrolyte and are generally regarded as commercial secrets (Zhang, 2006; 494 Haregewoin et al., 2016), preventing detailed knowledge of their toxicity by recyclers or 495 dismantlers. Disassembly also requires specialist insulated tools and specialist training by the 496 dismantler due to the high voltage of automotive batteries. The risk of short circuiting the pack 497 is prominent when disassembling, and this could lead to thermal runaway, potentially 498 triggering the production of HF gas. When the gas accumulates and is trapped in the cell this 499 leads to expansion and ultimately cell explosion (Harper et al., 2019). Thermal runaway in a 500 single cell could also cause a chain reaction and result in pack level battery failure (Wu et al., 501 2019). [For a review of Lithium Ion Battery Safety, which outlines some recent LIB battery 502 fires and fires in recycling facilities, consult Christensen et al. (2021).] It is clear that a shift 503 towards safer materials would improve the safety of dismantling packs. Additionally, 504 automation of the process would also improve safety, as the risk to human operators is 505 decreased.

506

507 All of the above issues in the battery design make disassembly a challenging way to process 508 end-of-life EV batteries. The process could be improved if recycling is kept in mind when 509 designing the battery and how it is assembled. Recycling is often seen as an end of the line 510 process; however, this attitude needs to change as recycling becomes ever more important due 511 to the decarbonisation of the world's energy production and consumption. Simple, small 512 adjustments to the design could vastly improve the disassembly process, making it the primary 513 choice in recycling batteries, not only in terms of purity of the products, but in terms of 514 processing costs and time. By improving the design and using greener materials within the cell

515 chemistries, this would in turn improve the safety aspects of the process, arguably the largest 516 barrier to disassembly, and why shredding is currently the preferred cell decommissioning and 517 passivation method.

518

519 At present, many dismantling facilities rely on manual disassembly (Garg et al., 2020). In the 520 future, robotic disassembly shows promise as an economical way to disassemble EV battery 521 packs. Given the time taken for manual disassembly, automated methods of disassembly 522 employing robotics and AI are seen as a promising avenue for enhancing the economics of LIB 523 recycling. Li et al. (2019) developed a vacuum conveyor belt that utilises the polymer separator 524 between electrodes as a means of separating anode and cathode. Alfaro-Algaba and Ramirez 525 (2020) conducted a techno-economic evaluation of the automated disassembly of Audi A3 526 Lithium e-Tron batteries, modelling partial and complete disassembly. They construct a range 527 of scenarios and analyse the profitability of variants of remanufacture, reuse and recycling on 528 a range of hypothetical cell conditions, although recycling is not modelled in detail. The 529 economics of the processes depend greatly on the level of disassembly and destinations of the 530 pack / modules extracted. However, developing a system that can deal with batteries from a 531 single-manufacturer is one challenge, but developing a universal system for disassembly, given 532 the wide variety of batteries on the market, represents a different magnitude of challenge (Garg 533 et al., 2020).

534

### 535 **4. Design for disassembly**

536 As described in the previous section, if disassembly is the main route for reprocessing, then the 537 packs, modules and cells need to be designed for automated opening. Figure 2 shows the fixing 538 mechanism used to seal and attach cells into modules and packs. There are numerous aspects 539 which could in principle be simple to solve but require standardisation. Even prior to 540 disassembly, assessment of the battery to determine whether second life is permissible is 541 hampered by interfacing the battery with the analysis hardware. A recent survey determined 542 that there were more than 20 types of connectors in use, with a variety of male, female and 543 screw connections (Thompson et al., 2020). It would also benefit from labelling, such that 544 different battery chemistries could be automatically separated before disassembly, resulting in 545 purer product streams. Some of the regulatory issues around battery labelling, and the impact 546 of the new EU battery regulations, are discussed in Ahuja et al. (2021) in this special issue.

547 From a disassembly perspective, the recycling plant of the future would prefer a standard cell 548 geometry to simplify disassembly, with a high active mass content per cell. We have recently 549 shown that the proportion of active mass can vary significantly depending on battery design, 550 varying from 60% for pouch cells, to 82% for prismatic cells (Thompson et al., 2020). Recent 551 developments from the Chinese battery manufacturer BYD use long cells which fit into a rigid 552 busbar. The so-called Blade Battery has a cell of 96 x 9 x 1.35 cm. It has a cell-to-pack (CTP) 553 system which negates the need to build modules. The makers claim to have significantly 554 increased the volumetric energy density (by up to 50%) compared to conventional LFP battery 555 packs. The CTP system makes pack disassembly easy as it simplifies connectors and 556 significantly reduces adhesives and wires in the pack structure. Being able to easily remove the 557 cells from the pack aids not only end-of-life processing, but also enables faulty cells to be 558 swapped if diagnostic tests indicate a drop in pack performance, extending pack life. Such 559 blade cells have withstood more than 3,000 charging/discharging cycles, which is equivalent 560 to 1.2 million km in an automotive application. If the cells themselves can be opened at EoL 561 then electrode separation should be easy as electrode connectors are at opposite ends of the 562 blade. This cell design fulfils many of the criteria necessary for simple disassembly.

563 The final aspect of cell design needed to enable rapid and efficient separation and recycling is 564 the use of water soluble binders. Carboxymethyl cellulose (CMC)/ styrene butadiene rubber is 565 now commonly used as a binder for anode materials and can be cast using water but it is less 566 stable with composite cathodes. Polyvinylidine fluoride (PVDF) is standardly used as the 567 binder but it needs to be cast using N-methyl-2-pyrrolidone which is toxic, expensive and 568 flammable. Li et al. (2020) recently showed that CMC/PVDF could be used as a binder for the 569 cathode and this could be cast using aqueous solutions. It was also shown that the electrode 570 after service could be recycled using water and the NCM523 recovered retained most of its 571 initial performance once it had been re-lithiated.

572

### 573 **5. Conclusions**

This study has shown that, based around a target price of \$100 / kWh for automotive battery systems, hydrometallurgical processing can potentially yield product streams that can be reused in new cells. Using retro-economic analysis it is shown that processing costs for batteries should be in the range \$2-6 / kg of battery to enable processing that is free from gate fees. Many of the processes run at pilot scale are in this cost range, potentially resulting in a cost saving compared with using virgin material. It is, however, clear that disassembly and 580 separation could yield processes which are less costly and produce materials with higher value than shredding and leaching. These enhanced processes have the potential to conserve more 581 582 value within a circular LIB supply chain than some existing methods where value is destroyed. 583 In general, it was shown that the cost saving using shredded material was generally < 20%, 584 whereas disassembly could potentially result in cost savings in the range 40 to 80% depending 585 on purity (not accounting for the actual step of disassembling the cell). At present, materials containing higher cobalt and nickel content are the only materials which are economically 586 587 viable to recycle. As chemistries transition to less costly lower cobalt formulations, battery recovery value decreases (Richa et.al. 2017; Wang et.al. 2014<sup>1</sup>; Wang et.al. 2014<sup>2</sup>; Yang et.al. 588 589 2021) As these contents are reduced, more efficient recycling techniques are required so that 590 all battery materials remain in a circular economy. While lithium iron phosphate is a lower cost 591 material, it is still essential to recycle phosphorus. This study suggests that the key step to 592 retaining value in the recycling process is product purity and this is likely to be higher in a 593 process which does not shred the material as the first step.

594

595 The barriers to disassembly and separation arise from a complex cell design with numerous 596 cells and modules constituting the overall pack. Simplifying the arrangement of cells and 597 designing cells such that they can be opened would facilitate disassembly. Many of the design 598 issues require standardisation in aspects such as labelling and connector design. Ahuja et al. 599 (2021) in this special issue discuss some of the challenges around product regulation and 600 labelling and information requirements to comply with the new EU Battery Regulations which 601 start to address some of these issues. Phasing out solvent-soluble glues would simplify 602 liberation and separation. Packs should also be designed with automated, intelligent 603 disassembly in mind, as it is quite clear that with the size of the projected market, this is the 604 only approach that will be viable for treating such a volume of material.

605

This requires coordination throughout the LIB supply chain, with manufacturers and component makers earlier in the chain having a greater appreciation of the need for improved circularity and the challenges faced by waste processors later in the supply chain.

609

610 It is quite clear that a mixed approach to recycling LIBs will be necessary. With small, difficult 611 to open cells containing high Co content, pyrometallurgical approaches will dominate. These 612 will have higher process costs and result in lower overall elemental recovery rates. As safe

- 613 shredding methods are developed, hydrometallurgical processing will dominate as it will be
- 614 lower cost and lead to larger proportions of material being recovered. Ultimately as cell design
- 615 progresses, separation and delamination will predominate as it enables purer products with
- 616 higher yields at lower costs. Recent developments such as blade cells enable easier disassembly
- 617 with modular to cell-to-pack assemblies.
- 618

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- Removal of wiring looms tricky
- Manipulation of connectors (especially where locking tabs fitted)
- High voltages until wiring loom / module links removed
- Lack of data on module condition in many present EV batteries
- Lack of labelling and identifying marks
- Potential fire hazards
- Potential HF off-gassing
- Sealants may be used in module manufacture (difficult to remove)
- Cells stuck together in modules with adhesives (difficult to separate)
- Components may be soldered together (difficult to separate)
- Module state of charge may not be known
- Clean separation of anodes and cathode for direct recycling difficult.
- Very finely powdered materials present risks (nanoparticles)
- Potential for HF compounds formed from electrolyte
- Potential for thermal effects if cells shorted during disassembly
- Chemistries not always known / proprietary
- Additional challenges with cylindrical cells (unwinding spiral)
- Disassembly of stacked structure with encapsulated anodes.

