



10 **ABSTRACT**

11 The demand for lithium-ion batteries (LIBs) has increased substantially over the last few decades due to  
12 their longer lifetime, greater resistance to self-discharge, and higher output voltage compared to other  
13 battery types. With the global trend of electrifying vehicle fleets, the number of LIBs reaching their end-  
14 of-life (EOL) is expected to grow substantially in the next decade. These EOL LIBs represent a significant  
15 secondary source of materials (e.g., Li, Co, Ni, Mn) that can be recovered and reused in LIBs or other  
16 products. In this study, we developed a bioleaching process that could recover critical materials from EOL  
17 LIBs in an economical and environmentally sustainable manner under industrially relevant conditions.  
18 Black mass, i.e., cathode-containing powder, prepared from EOL LIBs was leached using a biolixiviant  
19 produced from corn stover by *Gluconobacter oxydans* bacteria. Iron(II) was used as a reducing agent to  
20 promote metal dissolution. Techno-economic analysis (TEA) estimated a potential average profit margin  
21 of 21% for processing 10,000 t of black mass per year, which represents approximately 30% of the available  
22 black mass in the US in 2020. Life cycle assessment (LCA) demonstrated that bioleaching of spent LIBs  
23 could be more environmentally sustainable than alternative hydrometallurgical recovery methods such as  
24 hydrochloric acid leaching (16–19 kg vs. 43–91 kg CO<sub>2</sub> equivalent global warming potential per kg of  
25 recovered cobalt). The TEA results are highly dependent on the cost of black mass production, which varies  
26 by EOL LIB collection and transportation costs. Emerging technologies for deactivating used LIBs for fire  
27 safety at collection centers will allow the transport of EOL LIBs as non-hazardous materials, lower the cost  
28 of preparing black mass and thereby increase economic prospects for EOL LIBs recycling using this  
29 approach.

30 **KEYWORDS:** Lithium-ion batteries, Biohydrometallurgy, Organic acids, *Gluconobacter oxydans*,  
31 Recycling

## 32 1. INTRODUCTION

33 The massive loss of non-renewable materials in the linear economy (i.e., make-use-dispose) has heightened  
34 interest in novel approaches to recover materials from end-of-life (EOL) products (Deng et al., 2020).  
35 Lithium-ion batteries (LIBs) are one of the most critical EOL products to recycle due to their importance  
36 to clean energy and anticipated material shortages. Around 70% of the global cobalt reserve is projected to  
37 be consumed for batteries by 2040 (Roskill, 2020), and LIB demand is expected to outpace the raw materials  
38 supply by 2030 (Benchmark Mineral Intelligence, 2021). This growing demand is primarily due to LIBs'  
39 light weight, high-energy density, long cycle life, and low self-discharge rate, making them attractive for  
40 use in many applications, including consumer electronics and electric vehicles (EVs). Increased production  
41 of LIBs will place a greater strain on the geological reserves of metals such as cobalt, lithium, manganese,  
42 and nickel that are commonly used in LIB cathodes (Roskill, 2020). Recovering these metals from spent  
43 LIBs will be critical for mitigating future supply risks and alleviating the pressure on geological reserves  
44 (Fan et al., 2020).

45 Cathode materials constitute the largest value from LIB recycling, and LIB recycling has focused on  
46 cathode recycling by three major methods (parts of which can be combined or integrated):

- 47 1) Direct recycling recovers LIB cathode materials with minimal processing while maintaining the  
48 original cathode crystal structure. Preserving cathode morphology is the main advantage of this  
49 method that can significantly reduce recycling costs such as energy and chemical consumption  
50 (Gaines, 2018). Direct recycling is still in the early stage of development, and its industrial adoption  
51 is challenged by the need for pre-sorting of cathode types in order to apply the appropriate  
52 rejuvenation technology and the rapid development of new cathode chemistries that can render  
53 recovered cathode compositions obsolete.
- 54 2) Pyrometallurgy utilizes high-temperature thermal treatment to separate cathode-containing metals  
55 from other constituents by burning off electrolytes, binders, and plastics. The valuable metals are  
56 reduced to an alloy of copper, cobalt, nickel, and iron. Meanwhile, lithium and aluminum are  
57 typically lost in the slag (Lv et al., 2018). Pyrometallurgy is a simple recycling method that could  
58 be applied to any cathode type, but it is an energy-intensive process with high toxic gas emissions  
59 (Alipanah et al., 2021). It is also not sufficient for the final recovery of the components for LIB  
60 remanufacture; hydrometallurgical operations are applied to the alloy and slag (Makuza et al.,  
61 2021).
- 62 3) Hydrometallurgy involves the use of acids and reductants to leach the metals from the cathode  
63 materials. This method can be applied to any cathode chemistry and has the flexibility to target  
64 multiple metals to be recovered into metal salts. Although hydrometallurgical processes show  
65 higher energy efficiency than pyrometallurgical processes, in general they entail high chemical and  
66 water consumption, and wastewaters must be treated (Alipanah et al., 2021).

67 Biohydrometallurgy has attracted the attention of numerous researchers due to its potential environmental  
68 benefits in metal recovery, such as minimizing the waste generated and thereby lowering treatment costs  
69 (Yu et al., 2020). Bioleaching is a well-developed multidisciplinary process involving chemistry, biology,  
70 and metallurgy; it is one of several applications within hydrometallurgy for metal recovery from ores and  
71 wastes (Brierley, 2016). For example, copper is recovered at the Morenci Mine, USA, (230,000 t/y) at an  
72 industrial scale through bioleaching (Watling, 2015). For iron containing sulfidic ores (e.g., copper),  
73 acidophilic sulfur and iron oxidizing microorganisms are employed for promoting solubilization of target

74 minerals (Kaksonen et al., 2018). For non-sulfidic materials such as EOL LIBs carbon oxidizing  
75 heterotrophic microorganisms can be used for bioleaching (Bahaloo Horeh et al., 2016). These  
76 microorganisms produce organic acid that can help solubilize the target metals from the waste stream  
77 (Golmohammadzadeh et al., 2018).

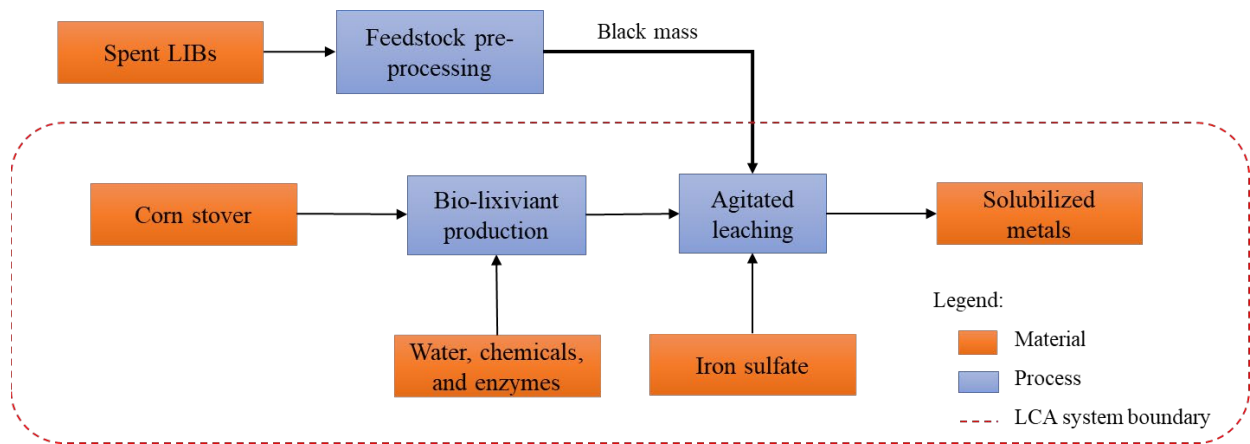
78 While bioleaching approaches have a number of potential benefits, two primary factors give pause when  
79 considering the actual application of bioleaching for metal recovery from industrial wastes such as EOL  
80 LIBs. First, lab studies of bioleaching are often carried out at low pulp densities (i.e., solid to liquid ratios)  
81 in order to achieve high leaching efficiencies (yields), although actual deployment of such low pulp  
82 densities would likely engender high processing costs and environmental impacts. For example, Biswal et  
83 al., (2018) employed *Aspergillus niger* to bioleach EOL LIBs with 0.25% pulp density and achieved  
84 leaching efficiencies of 100% for lithium and 82% for cobalt. Bahaloo-Horeh et al., (2018) applied  
85 bioleaching with *Aspergillus niger* at pulp densities <1% to leach 100% of lithium, 38% of cobalt, 45% of  
86 nickel, and 72% of manganese from spent LIBs of smart phones. Xin et al., (2016) investigated bioleaching  
87 of spent LIBs using *Acidithiobacillus thiooxidans* and *Leptospirillum ferriphilum* with 1% pulp density to  
88 leach more than 95% of lithium, cobalt, nickel, and manganese. Ghassa et al., (2021) utilized a mixed  
89 culture of thermophilic chemolithotrophic bacteria (*Acidithiobacillus caldus*, *Leptospirillum ferriphilum*,  
90 *Sulfobacillus spp.* and *Ferroplasma spp.*) with 1% pulp density to leach 99% of cobalt and nickel and 84%  
91 of lithium. Metal extraction efficiency can decrease at higher pulp densities due to the presence of alkaline  
92 wastes or metal toxicity to *in situ* leaching microorganisms (Pathak et al., 2017). Several research groups  
93 have attempted to increase the pulp density, but extraction efficiencies suffered. For example, Niu et al.,  
94 (2014) investigated bioleaching of spent LIBs with pulp densities between 1% and 4%. The leaching  
95 efficiency decreased from 52% to 10% for cobalt and from 80% to 37% for lithium, when pulp density  
96 increased from 1% to 4%. Similar trends were observed in another bioleaching study of spent LIBs by  
97 Naseri et al., (2019) in which leaching efficiencies dropped from 100% for lithium and cobalt to < 70% for  
98 lithium and <20% for cobalt, when pulp density increased from 1% to 10%. As higher pulp density could  
99 significantly reduce bioleaching cost and environmental footprint by reducing the required quantities of  
100 microorganisms and/or biologically produced acids, finding the optimal balance between pulp density and  
101 leaching efficiency is crucial to ensure the maximum economic and environmental benefits.

102 Second, the bioleaching process kinetics can be slow, which adversely affects economic and environmental  
103 sustainability. For instance, Bahaloo Horeh et al., (2016) applied fungal leaching (*Aspergillus niger*) for 14  
104 days and achieved leaching efficiencies of 95% for lithium, 45% for cobalt, and 38% for nickel. Naseri et  
105 al., (2019) used *Acidithiobacillus ferrooxidans* bacteria and the highest leaching efficiencies (100% for Li,  
106 88% for Co, and 20% for Mn) were achieved after 12 days. Niu et al., (2014) utilized *Alicyclobacillus* and  
107 *Sulfobacillus* bacteria and leached 89% of Li and 72% of Co after 11 days. The cost and environmental  
108 footprint of bioleaching increase with longer leaching time due to (1) higher energy consumption for  
109 temperature control and leachate agitation as well as (2) larger or a greater number of bioreactors required  
110 to bioleach the same amount of feedstock.

111 The existing literature has focused on maximizing metal extraction efficiency without considering costs  
112 and environmental footprint. None of the studies cited above have analyzed the economic and  
113 environmental life cycle consequences of bioleaching LIBs, which would be necessary considerations for  
114 commercialization of any technology for LIB recycling. Performing techno-economic analysis (TEA) and  
115 life cycle assessment (LCA) in the early stages of technology development is essential to identify, quantify,  
116 and ideally ultimately to surmount the technical, economic, and environmental barriers to technology

117 maturation and improve technology competitiveness for industry adoption. Here we report a new  
118 bioleaching approach for EOL LIBs, based on production of organic acids by the bacterium *Gluconobacter*  
119 *oxydans*, together with the results of TEA and LCA for the process.

120 The overall bioleaching process and the LCA system boundary to recover valuable metals from spent LIBs  
121 are shown in Figure 1. The EOL LIBs go through pre-processing steps in which cathode-containing powder,  
122 called black mass, is produced through a sequence of processes including discharging, dismantling, and  
123 shredding. The black mass is then bioleached using a lixiviant containing organic acids (predominantly  
124 gluconic acid) derived from the agricultural waste corn stover (Jin et al., 2019). The leaching process is  
125 conducted in the presence of the reducing agent  $Fe^{2+}$  (provided by addition of ferrous sulfate). The process  
126 outputs are solubilized cobalt, lithium, nickel, and manganese.



127

128 **Figure 1.** The overall bioleaching process for metal recovery from spent lithium-ion batteries.

129

## 130 2. METHODOLOGY

131

132

### a. Black mass source and characterization

133 The black mass used in this study was provided by Retrie Technologies (Trail, British Columbia, Canada)  
134 for laboratory-scale experiments. The black mass was obtained from the Retrie process flow, wherein  
135 spent LIBs of various cathode types undergo a series of mechanical treatments, including multistage  
136 crushing and sieving, magnetic separation to remove steel casing, and fine crushing to remove small pieces  
137 of current collectors (Ekberg and Petranikova, 2015). The black mass used in the studies reported here  
138 contained 2.3% lithium, 19.3% cobalt, 4.5% manganese, and 5.6% nickel, by mass.

139

### b. Bioleaching process

140 *Biolixiviant production:* Biolixiviant was prepared using *Gluconobacter oxydans* strain B58 grown on corn  
141 stover hydrolysate (the corn stover was hot water pretreated, and enzymatically hydrolyzed) in Pikovskaya  
142 phosphate modified (Pkm) medium at 30°C, with shaking at 150 rpm for 42 hr, similar to methods described  
143 previously (Jin et al., 2019). The corn stover hydrolysate containing 28 g/L glucose was converted by the  
144 *G. oxydans* into an organic acid-rich mixture including citric, gluconic, xylonic, acetic, formic, and succinic

145 acid. Gluconic acid was the dominant organic acid at 89 mM, accounting for ~60% (molar basis) of the  
146 identified organic acids.

147 *Metals leaching:* Bioleaching was conducted with filtered (0.22 μm) biolixiviant diluted with nanopure  
148 water to 75 mM gluconic acid and applied to the black mass (Retriev Technologies) at 2.5% solid to liquid  
149 ratio (w/w). Ferrous sulfate was used as a reducing agent to help solubilize metals in the leaching process.  
150 The iron(II) requirement was estimated using the stoichiometry shown in Eq. (1) for an NMC cathode  
151 example (Diaz et al., 2020).



153 where  $x + y + z = 1$

154 Samples were incubated at 55°C for 30 hrs in sealed flasks on an orbital shaker at 150 rpm. These conditions  
155 were selected from an optimization process that explored a variety of bioleaching conditions (Alipanah et  
156 al).

157 *Analytical methods:* Sugar and organic acid content were determined by high performance liquid  
158 chromatography (HPLC) as described previously (Reed et al., 2016). Metal concentrations were determined  
159 with atomic adsorption spectroscopy (AAS; Agilent 240FS 200 Series) using acetylene and nitrous oxide  
160 and the data was analyzed using SpectraAA software (Agilent). Calibration standards were prepared with  
161 Zn (1 mg/L), Al (40 mg/L) and 5 mg/L for all other metals of interest. HCl (3%) was used to prepare sample  
162 dilutions (Strauss et al., 2021).

### 163 **c. Materials and energy**

164 This study evaluated the following factors that were estimated to have a significant influence on the  
165 economic performance of bioleaching: pulp density, organic acid concentration, leaching temperature and  
166 time. These factors had direct effects on the material and energy consumptions of bioleaching, and therefore  
167 their effects were modeled as follows.

168 *Pulp density:* Eq. (2) shows the pulp density calculation formula in which slurry weight is the summation  
169 of black mass and biolixiviant weights.

$$170 \text{Pulp density} = \frac{\text{Black mass weight}}{\text{Slurry weight}} * 100 \quad \text{Eq. (2)}$$

171 *Gluconic acid concentration:* For this study, corn stover was utilized as the main carbon source for *G.*  
172 *oxydans* biolixiviant production, of which gluconic acid is the primary organic acid. The gluconic acid  
173 concentration is determined by the number of sodium gluconate moles per liter of the acid.

174 *Leaching temperature and time:* The energy consumption and volume of the leaching tank were determined  
175 by the temperature and duration of the leaching process. The leaching was performed at 55°C for 30 h. To  
176 scale-up laboratory experiments to an industrial scale, the electricity consumption for an agitated leaching  
177 tank was calculated based on the methodology from Piccinno *et al.* (Piccinno et al., 2016). The heating  
178 energy required to raise leachate temperature from room temperature (assumed as 20°C) to 55°C was  
179 designated  $Q_{raise}$  (unit: Joule), and loss of energy during 30 h of leaching to maintain the leaching  
180 temperature was designated  $Q_{loss}$  (unit: Joule). The energy requirement for agitation of leachate was  
181 estimated at 2 kW/m<sup>3</sup> during the leaching time (Efe et al., 2013). The total energy requirement is the

182 summation of energy for raising the temperature, maintaining temperature, and agitation of leachate for the  
183 leaching duration. Eqs. (3-5) show the actual formulas used for the energy requirement calculations.

$$184 \quad Q_{raise} = C_p * m_{mix} * (T_r - T_0) \quad \text{Eq. (3)}$$

$$185 \quad Q_{loss} = A * \frac{K_a}{s} * (T_r - T_0) * t \quad \text{Eq. (4)}$$

$$186 \quad Q_{TOTAL} = \frac{Q_{raise} + Q_{loss}}{\eta_{heat}} + \frac{2 * V * t * 1000}{Q_{agitation}} \quad \text{Eq. (5)}$$

187  $C_p$ : specific heat capacity (J/kg/K)

188  $m_{mix}$ : mass of leachate (kg)

189  $T_r$ : target leaching temperature (°K)

190  $T_0$ : starting leaching temperature (°K)

191  $A$ : surface area of the leaching tank (m<sup>2</sup>)

192  $K_a$ : thermal conductivity of the insulation material (W/(m.K))

193  $s$ : insulation thickness (m)

194  $t$ : leaching time duration (s)

195  $\eta_{heat}$ : efficiency of the heating element (%)

196  $V$ : volume of leachate (m<sup>3</sup>)

197

198 To evaluate the economic and environmental impacts of these factors, TEA and LCA were conducted,  
199 followed by sensitivity analysis.

200

#### 201 **d. Techno-economic analysis**

202 This study assumed an industrial-scale bioleaching plant capable of processing 10,000 t of black mass a  
203 year, representing ~30% of the available black mass in the US in 2020 (Melin, 2018), over the next 30 years  
204 in the United States. Direct costs of the process include: (1) material costs which consist of purchasing  
205 black mass, iron sulfate, and gluconic acid; (2) water and electricity as the utilities consumed for leaching;  
206 (3) other direct costs including transport of black mass to the bioleaching plant, waste management, labor  
207 for operation and maintenance, and material/supplies for the required maintenance.

208 *Material cost*: This study used the EverBatt tool (Dai et al., 2019) to estimate the cost of black mass. EOL  
209 LIB prices are usually based on the cobalt content of their cathodes (Dai et al., 2019), as cobalt is the most  
210 valuable material in LIB cathodes. The black mass we analyzed contains around 19% cobalt by weight.  
211 Therefore, the cost of the EOL LIBs from which the black mass was produced was assumed to be \$0.38/kg,  
212 proportional to the EverBatt assumption for NMC111, which contains 10% cobalt by weight. The cost for  
213 collection of EOL LIBs from end-users to the recycler producing black mass was estimated as \$0.25/kg  
214 using the assumptions of 900-mile transport distance and \$0.14/ton-mile transportation fee (Austin, 2015).  
215 For the black mass production cost at the LIB recycling facility, the model built on EverBatt estimated a  
216 cost of \$0.5/kg. The black mass purchasing cost for the bioleaching operation was considered as the  
217 summation of the above-mentioned costs plus a 15% profit margin for the recycler producing black mass  
218 from EOL LIBs (i.e., \$1.29/kg). We relied on a previous study for the cost of microbially produced gluconic  
219 acid from corn stover (Zhang et al., 2016). Zhang *et al.* estimated that each kg of sodium gluconate could  
220 be produced for a total cost of \$0.45.

221 *Other costs:* The salary for operating labor was estimated at \$26.48/h (US Department of Labor, 2021), and  
222 the needed personnel was calculated based on industry standards (Alkhatat and Gerrard, 1984), assuming  
223 8,000 h/y of operation. This study utilized factors provided by Silla *et al.* (Silla, 2003) to estimate other  
224 costs such as quality control, supervision, and maintenance associated labor, material, and supplies. Capital  
225 costs included major equipment (i.e., leaching tank (“Matches’ Process Equipment Cost Estimates,” 2021))  
226 and required infrastructure (e.g., land). Factors established by Silla *et al.* (Silla, 2003) were also used to  
227 estimate indirect (e.g., property taxes) and general (e.g., marketing) costs of the process. To mitigate the  
228 effects of uncertainties in the implementation of proposed process and future market condition, capital cost  
229 was increased by 15% as a contingency cost.

230 *Revenue:* The selling price per kg of lithium (as hydroxide), cobalt, manganese, and nickel were assumed  
231 as \$16, \$36, \$2.8, and \$14, respectively (London metal exchange, 2021; USGS, 2021). It should be noted  
232 that metal prices (USGS, 2021) are currently available only for 99+% pure individual metals, whereas the  
233 proposed bioleaching process delivers mixed solubilized metals. The EverBatt tool (Dai *et al.*, 2019)  
234 estimated that the costs for post-leaching processes to obtain 99+% pure individual metals, including  
235 separation and purification, account for 30% of the total cost of EOL LIB recycling with the same  
236 processing capacity that uses a generic hydrometallurgical process. Therefore, the selling prices of the  
237 bioleaching output metals were discounted by 30% from their respective 99+% pure individual metal prices  
238 to approximate the revenue from bioleaching in this study.

239 To evaluate the economic viability of the proposed bioleaching process, net present value (NPV) was  
240 utilized. Eq. (6) shows the formulation used for calculating of NPV where (*NCF<sub>t</sub>*) is net cash flow over time  
241 *t*, and *i* is the discounting rate (8%) over *n* periods (30 years). More TEA assumptions are available in  
242 Supporting Information Table S1 and Table S2.

$$243 \quad NPV = \sum_{t=0}^n \frac{NCF_t}{(1+i)^t} \quad \text{Eq. (6)}$$

244

#### 245 **e. Life cycle assessment**

246 The goal of LCA was to quantify the environmental impacts of bioleaching for value recovery from  
247 LIB black mass. The LCA system boundary includes transportation of black mass to the bioleaching  
248 facility, gluconic acid preparation (Jin *et al.*, 2019; Zhang *et al.*, 2016), and bioleaching. The functional  
249 unit is 1 kg of cobalt recovered from the black mass as the main product.

250 As the output from the bioleaching process includes lithium, cobalt, manganese, and nickel, which have  
251 high disparities in value (e.g., \$36/kg cobalt vs. \$3/kg manganese), economic allocation was applied for  
252 estimation of impacts associated with recovering cobalt (see Supporting Information Table S3 for the  
253 allocation factors). Cobalt was chosen because it is the most valuable metal in the black mass composition  
254 and has a critical effect on the profitability of the process. Economic allocation was the preferred approach  
255 when (1) the values of co-products were substantially different and (2) revenue was the main driver of the  
256 value recovery; Without economic incentive, the proposed process might not occur to have any  
257 environmental impacts (Santero and Hendry, 2016). Table 1 shows the unit processes for the functional  
258 unit after economic allocation.

259 **Table 1.** Unit processes for recovering 1 kg cobalt from the black mass after economic allocation.

Unit process	Process	Value			Unit
		Low	Baseline	High	
Transport, freight, lorry, unspecified {GLO}  market for transport, freight, lorry, unspecified   APOS, U	Feedstock collection	2.690E+01	2.93E+01	3.17E+01	tkm
Corn stover, production, average, US, 2022/kg/RNA	Gluconic acid production	6.37E+00	6.93E+00	7.49E+00	kg
Sulfuric acid {GLO}  market for sulfuric acid   APOS, U	Gluconic acid production	1.36E-01	1.47E-01	1.59E-01	kg
Steam, in chemical industry {GLO}  market for   APOS, U	Gluconic acid production	1.09E+00	1.19E+00	1.28E+00	kg
Lime {GLO}  market for lime   APOS, U	Gluconic acid production	1.04E-01	1.13E-01	1.22E-01	
Oxygen, liquid {RoW}  market for   APOS, U	Gluconic acid production	1.01E-01	1.10E-01	1.19E-01	kg
Protein pea {GLO}  market for   APOS, U	Gluconic acid production	5.27E-02	5.74E-02	6.20E-02	kg
Phosphate fertiliser, as P2O5 {RER}  diammonium phosphate production   APOS, U	Gluconic acid production	3.91E-03	4.25E-03	4.59E-03	kg
Corn steep liquor/kg/RNA	Gluconic acid production	8.98E-03	9.77E-03	1.06E-02	kg
Sodium hydroxide, without water, in 50% solution state {GLO}  market for   APOS, U	Gluconic acid production	5.28E-01	5.75E-01	6.22E-01	kg
Compressed air, 800 kPa gauge {GLO}  market for   APOS, U	Gluconic acid production	1.46E+00	1.59E+00	1.72E+00	kg
Activated carbon, granular {GLO}  activated carbon production, granular from hard coal   APOS, U	Gluconic acid production	7.23E-02	7.86E-02	8.50E-02	kg
Electricity, biomass, at power plant NREL/US U (No transportation)	Gluconic acid production	5.53E+00	6.01E+00	6.50E+00	kWh
Water, completely softened, from decarbonised water, at user {GLO}  market for   APOS, U	Gluconic acid production and bioleaching	1.83E+02	2.00E+02	2.16E+02	kg
**Electricity, medium voltage {US}  market group for   APOS, U	Bioleaching	1.92E+01	2.09E+01	2.26E+01	kWh
Iron sulfate {RoW}  market for iron sulfate   APOS, U	Bioleaching	3.75E+00	4.13E+00	4.51E+00	kg
<b>Emission to water</b>					
Wastewater	Bioleaching and biolixiviant production	1.83E+02	2.00E+02	2.16E+02	kg
Glucose	Biolixiviant production	3.87E-01	4.22E-01	4.56E-01	kg
Calcium compounds	Biolixiviant production	1.87E-01	2.04E-01	2.20E-01	kg
<b>Emission to soil</b>					
Sodium salt	Biolixiviant production	8.49E-01	9.24E-01	9.99E-01	kg
<b>Emission to air</b>					

Carbon dioxide	Biolixiviant production	1.75E-01	1.90E-01	2.05E-01	kg
Oxygen	Biolixiviant production	3.90E-01	4.24E-01	4.58E-01	kg
Nitrogen	Biolixiviant production	1.29E+00	1.41E+00	1.52E+00	kg

### Output

Cobalt		0.92	1	1.11	kg
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\*\*Electricity generated from burning biomass (lignin residue from corn stover) is assumed to be used for leaching operation

As the study aimed to evaluate the value recovery from black mass in the US, the U.S. Environmental Protection Agency's TRACI (Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts) was utilized to classify and characterize the environmental impacts. ReCipe 2016 midpoint (H) was also used to augment TRACI for estimation of the water consumption impact.

## 3. RESULTS AND DISCUSSION

### a. Bioleaching results

The bioleaching approach developed for this study used biolixiviant that consisted of a relatively dilute solution of acid in comparison to other leaching methods (0.075M vs.  $\geq 1$ M), but it still yielded comparable metal extraction efficiencies (Table 2, references included in the table). The bioleaching processes (i.e., the first two rows in Table 2) needed longer leaching time, but the required temperature was lower (an industrial consideration for lower costs). TEA and LCA implications of the proposed bioleaching experimental conditions will be further discussed in the following subsections.

**Table 2.** Comparison of bioleaching conditions and the resulting metal extraction performance with competing leaching technologies for LIBs.

Leaching technology	Recycled battery chemistry	Leaching acid	Leaching condition*	Co recovery %	Li recovery %	Mn recovery %	Ni recovery %
This study	Co=19% Li=2% Mn=5% Ni=6%	Gluconic acid	0.075 M, 25 g L <sup>-1</sup> , 55°C, 30 h	71–86	100	100	57–84
(Xin et al., 2016)	Co=16% Li=4% Mn=21% Ni=15%	Biogenic H <sub>2</sub> SO <sub>4</sub>	Unknown, 10 g L <sup>-1</sup> , 30°C, 216 h	96	95	95	97
(Wang et al., 2009)	LiCoO <sub>2</sub> (Co=57% Li=7% assumed)	Hydrochloric acid	4 M, 20 g L <sup>-1</sup> , 80 °C, 1 h	97	97	98	97
(Guzolu et al., 2017)	Co=20.56% Li=3.43% Mn=12.3% Ni=0.72%	Hydrochloric acid	5 M, 10 g L <sup>-1</sup> , 95 °C, 70 min	99	98	-	-
(Meshram et al., 2015)	Co=35.8% Li=6.5% Mn= 11.6% Ni=10.06%	Sulfuric acid	1 M, 50 g L <sup>-1</sup> , 95 °C, 4 h	66	93	50.02	96.3
(Latif and Ahmed, 2017)	Co=6.43% Li=4.37%	Sulfuric acid	1.5 M, 15 wt.% H <sub>2</sub> O <sub>2</sub> , 40 g L <sup>-1</sup> , 60 °C, 1 h	94.07	98.1	-	-
(Chen and Zhou, 2014)	Co=53.8% Li=4.37%	Citric acid	2 M, 2% H <sub>2</sub> O <sub>2</sub> , 50 g L <sup>-1</sup> , 70 °C,	99	93		

			80 min				
(Musariri et al., 2019)	Co=28.82% Li=9.73% Mn=24.39% Ni=33.91%	Citric acid	1.5 M, 10 g L <sup>-1</sup> , 95 °C, 30 min	95	97	-	99
(Li et al., 2012)	LiCoO <sub>2</sub> (Co=57% Li=7% assumed)	Ascorbic acid	1.25 M, 25 g L <sup>-1</sup> , 70 °C, 20 min	94.8	98.5	-	-
(Li et al., 2015)	Co=57.94% Li=6.76% Mn=0.91% Ni=0.76%	Succinic acid	1.5 M, 25 g L <sup>-1</sup> , 4 wt.% H <sub>2</sub> O <sub>2</sub> , 70 °C, 20 min	96	96	-	-
(Li et al., 2017)	Co=17.68% Li=6.79% Mn=16.46% Ni=17.58%	Lactic acid	1.5 M, 20 g L <sup>-1</sup> , 0.5 wt.% H <sub>2</sub> O <sub>2</sub> , 70 °C, 20 min	98.9	97.7	98.4	98.2

278 \*Leaching condition refers to acid concentration (M), pulp density (g/L), hydrogen peroxide (wt.%, if any), leaching temperature  
279 (°C), and leaching time (minutes or hours).

## 280 b. Life Cycle Assessment

281 The environmental impacts associated with recovering 1 kg of cobalt are shown in Table 3, comparing the  
282 proposed bioleaching process with other leaching technologies discussed in Table 2. It shows that  
283 bioleaching is an environmentally competitive technology for recycling LIBs in all the impact categories  
284 under investigation. The environmental advantage is mainly due to its lower acid consumption compared  
285 to other chemical leaching methods (Chen and Zhou, 2014; Guzolu et al., 2017; Latif and Ahmed, 2017; Li  
286 et al., 2017, 2015, 2012; Meshram et al., 2015; Musariri et al., 2019; Wang et al., 2009) and shorter leaching  
287 time compared to other bioleaching method (Xin et al., 2016) which lowered electricity consumption.  
288 Sulfuric acid leaching developed by Meshram et al. (2015) and citric acid leaching developed by Chen and  
289 Zhou (2014) showed comparable environmental performances with our proposed bioleaching. However,  
290 their feedstocks contained higher concentrations of Co than our study (i.e., 35.8% and 53.8% vs. 19%). If  
291 the same black mass material was used, the results may be less favorable for these two processes.

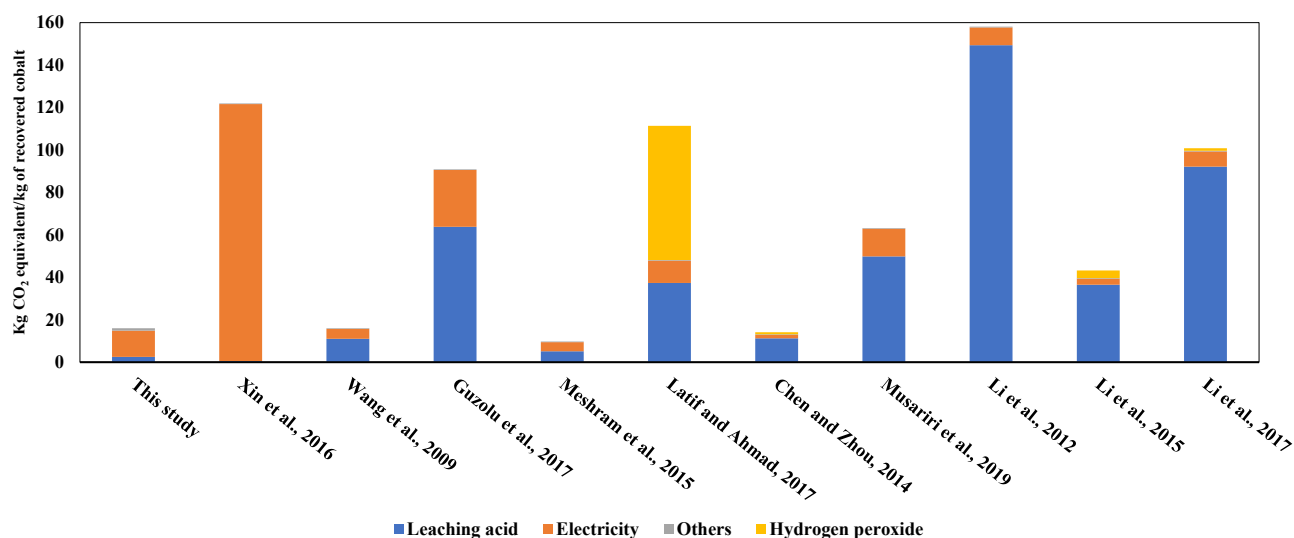
292 **Table 3.** LCA comparison between the current study and other leaching methods in the literature for  
293 recovering 1 kg of cobalt from LIB materials.

Impact category	This study	Xin et al., 2016	Wang et al., 2009	Guzolu et al., 2017	Meshram et al., 2015	Latif and Ahmad, 2017	Chen and Zhou, 2014	Musariri et al., 2019	Li et al., 2012	Li et al., 2015	Li et al., 2017	Individual impact category specific value	
												Low	High
Ozone depletion	1.8E-06	1.0E-05	1.5E-05	3.1E-05	2.8E-06	2.4E-05	5.4E-06	2.4E-05	1.5E-05	8.3E-06	1.1E-05		
Global warming	1.7E+01	1.2E+02	4.3E+01	9.1E+01	9.8E+00	1.1E+02	1.4E+01	6.3E+01	1.6E+02	4.3E+01	1.0E+02		
Smog	2.0E+00	3.1E+00	2.8E+00	6.0E+00	5.8E-01	6.0E+00	1.1E+00	4.5E+00	8.4E+00	1.5E+00	4.4E+00		
Acidification	7.1E-02	3.6E-01	2.1E-01	4.4E-01	4.3E-02	4.7E-01	7.4E-02	3.2E-01	8.7E-01	1.4E-01	3.7E-01		
Eutrophication	1.2E-01	9.9E-01	2.1E-01	4.6E-01	5.5E-02	3.8E-01	5.7E-02	2.9E-01	1.1E+00	1.1E-01	3.1E-01		
Carcinogenics	1.2E-06	1.0E-05	2.4E-06	5.2E-06	6.0E-07	1.8E-05	9.0E-07	3.4E-06	7.0E-06	2.0E-06	4.6E-06		
Non carcinogenics	4.3E-06	3.5E-05	1.2E-05	2.6E-05	2.8E-06	2.7E-05	4.0E-06	1.8E-05	9.0E-05	4.8E-06	1.2E-05		

Respiratory effects	4.5E-02	3.7E-01	8.3E-02	1.8E-01	2.1E-02	1.4E-01	2.3E-02	1.2E-01	1.9E-01	3.7E-02	7.9E-02
Ecotoxicity	1.2E+02	9.5E+02	2.6E+02	5.6E+02	6.2E+01	1.0E+03	8.6E+01	3.7E+02	8.3E+02	1.5E+02	3.4E+02
Fossil fuel depletion	1.3E+01	9.2E+01	4.1E+01	8.8E+01	9.3E+00	1.5E+02	1.5E+01	6.3E+01	1.9E+02	1.0E+02	2.4E+02
Water consumption**	2.9E-01	9.7E-01	1.2E+00	2.6E+00	2.5E-01	5.0E+00	4.6E-01	1.9E+00	-1.6E+01	5.1E-01	1.7E+00

294 \*\* Water consumption was assessed by ReCipe 2016 midpoint (H).

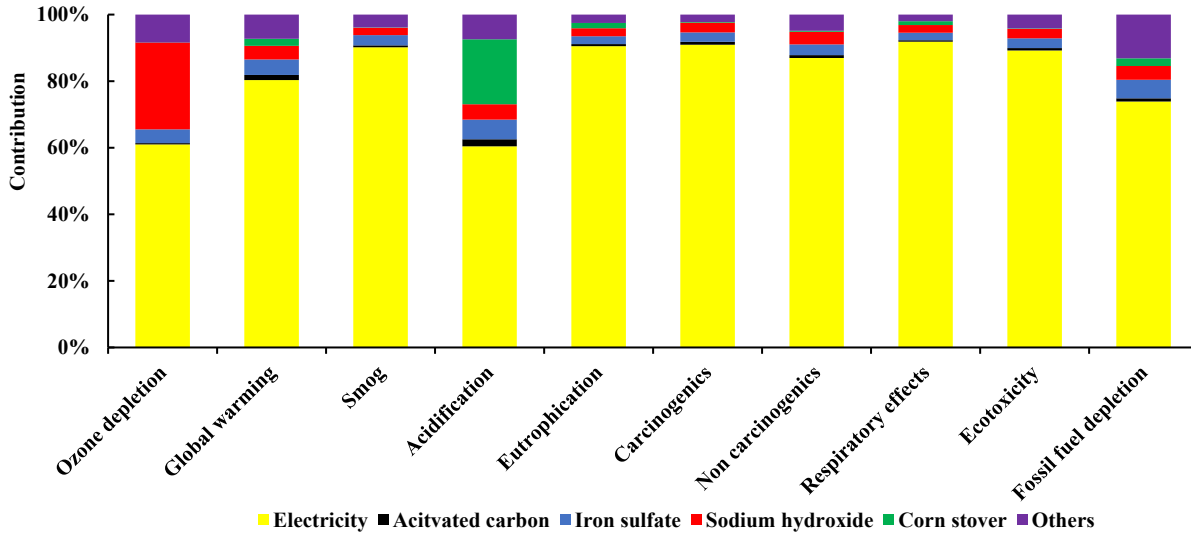
295 Figure 2 shows the result of comparison in global warming potential (GWP) category. The proposed  
 296 biotechnology showed 16–19 kg CO<sub>2</sub> equivalent GWP per kg of recovered cobalt after economic allocation,  
 297 whereas the other leaching technologies were estimated to generate 10–158 kg CO<sub>2</sub> equivalent GWP per  
 298 kg of recovered cobalt after economic allocation.



299  
 300 **Figure 2.** Global warming potential comparison between the proposed bioleaching technology, using  
 301 biolixiviant produced from corn stover and containing 75 mM gluconic acid, and other leaching methods  
 302 for recovering 1 kg of cobalt from lithium-ion battery materials (see Table 2 for the specific references).

303 The environmental hotspot (i.e., the main contributor to the associated impacts) of the proposed bioleaching  
 304 process was identified as electricity (e.g., 80% of total GWP), which was required for the agitated leaching  
 305 (Figure 3). The other major contributors were iron sulfate and sodium hydroxide (used for sodium gluconate  
 306 production (Zhang et al., 2016)), accounting for 5% and 4% of total GWP, respectively.

307 Combustion of lignin residue from the corn stover could supply enough heat and electricity for the  
 308 biolixiviant production and provide a surplus of 251 kWh electricity per ton of produced sodium gluconate  
 309 (Zhang et al., 2016). The excess electricity generated from biolixiviant production could be utilized for the  
 310 leaching process. This reduced 4% of the total electricity consumed for the leaching process which equals  
 311 more than 1,000 t of CO<sub>2</sub> equivalent footprint reduction per year.

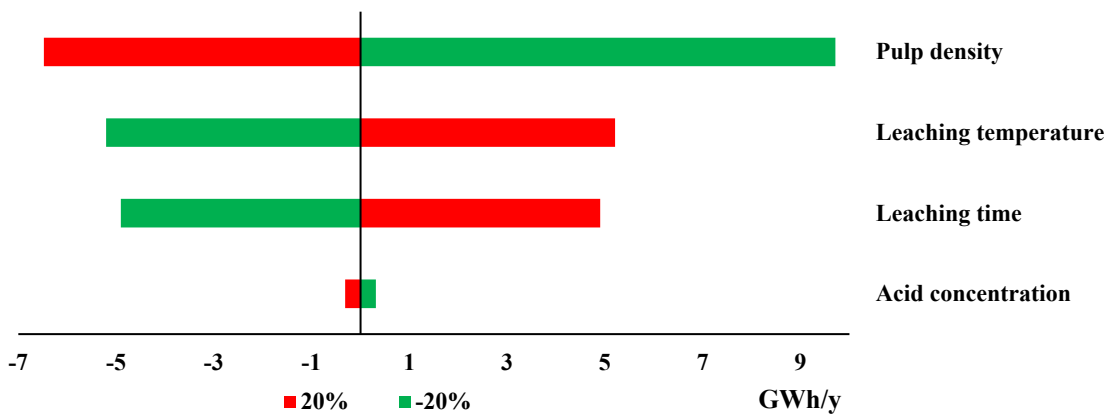


312

313

314 **Figure 3.** Life cycle impact of recovering 1 kg of cobalt from black mass through the bioleaching  
 315 process.

316 The sensitivity analysis represented in Figure 4 shows the major influencing factors on electricity  
 317 consumption and, consequently, on the bioleaching environmental impact. Pulp density significantly affects  
 318 energy usage because it has the greatest effect on the amount of leachate that must be processed (Eq.2).  
 319 With increasing pulp density (i.e., higher solid to liquid ratio), less biolixiviant is required to leach the same  
 320 target processing volume of black mass (i.e., 10,000 t/y), so the energy consumption is reduced. A 20%  
 321 increase in pulp density could result in an annual energy savings of more than 6 GWh. On the other hand,  
 322 a 20% reduction in pulp density has a greater impact on electricity consumption, increasing it by  
 323 approximately 10 GWh/y. Following that, leaching temperature and time have a similar effect, affecting  
 324 energy consumption by approximately 5 GWh/y for a 20% variation. Change in acid concentration has a  
 325 negligible influence on the energy consumption of the proposed bioleaching process.



326

327

**Figure 4.** Sensitivity of the electricity consumption to 20% change in main affecting factors.

328

329 **c. Techno-economic analysis**

330 Overall, the proposed bioleaching process was estimated to be potentially profitable with more than \$110M  
331 NPV over the next 30 years on average. The process could generate around \$42M–\$51M revenue per year  
332 (17%–26% profit margin) with cobalt as the primary source of revenue (>80% of total revenue), and the  
333 profitability of the process depends heavily on the price and recovery efficiency of cobalt (Figure S1).

334 Table 4 shows the total cost breakdown. The total cost of the process falls between \$20 and \$24 per kg of  
335 recovered cobalt. The material cost constituted the largest portion of the total cost. The main cost  
336 contributors to the material expense were identified as the purchasing cost of LIB black mass (36% of the  
337 total cost) and iron sulfate (10% of the total cost).

338 **Table 4.** Total cost breakdown for bioleaching 10,000 t/y of black mass.

<i>Categories</i>	<i>Cost (\$M/y)</i>	<i>% of Total Cost</i>
<b>Material</b>	19.44–19.53	54%
<i>Sodium gluconate</i>	2.74–2.83	8%
<i>Iron sulfate</i>	3.76	10%
<i>Black mass</i>	12.94	36%
<b>Electricity</b>	2.80–2.89	8%
<b>Utility</b>	0.33–0.34	1%
<b>Waste management</b>	1.08–1.10	3%
<b>Labor</b>	0.58	2%
<b>Other</b>	0.25–0.26	1%
<b>Capital</b>	0.76–0.80	2%
<b>Indirect</b>	1.19–1.21	3%
<b>General</b>	6.10–6.15	17%
<b>Income tax</b>	2.64–4.93	10%
<b>Total cost</b>	35.18–37.66	100%

339

340 The positive net profit indicates the economic feasibility of deploying the proposed bioleaching technology  
341 at scale. It would help increase LIB recycling rate, which is currently estimated as 5% in the US, by  
342 improving the economics, a key barrier to a higher recycling rate (Toba et al. (2021)). For example, Li-  
343 Cycle holdings, an emerging LIB recycling company in the North America, reported a net profit margin of  
344 approximately -1000% in the last two years (Macrotrends, 2022). Such a low economic return is not  
345 sustainable, so it is critical to evaluate and improve the economic performance of LIB recycling  
346 technologies, and our TEA results support this goal.

347 **d. Challenges, limitations, and opportunities**

348 Bioleaching has not yet been adopted by industry for large-scale value recovery from spent LIBs, and this  
349 study identified the major challenges that hinder commercialization of the proposed bioleaching  
350 technology. They include uncertain external factors such as black mass purchasing cost, high iron(II)  
351 consumption, changing cathode chemistry towards less cobalt, and economic value of bioleached metals.  
352 Therefore, different scenarios were examined for these factors.

353 *Black mass purchasing cost:* The primary cost driver in TEA is the black mass purchasing cost, which  
354 accounts for 36% of the total costs. The breakeven price for the black mass is ~\$2.19/kg (~69% higher than  
355 the assumed price of \$1.29/kg), above which bioleaching of LIB black mass is not economically viable.  
356 Wang and Yu estimated that the cost of black mass preparation was ~\$5/kg, implying that bioleaching  
357 would be unprofitable under their assumption (Wang and Yu, 2021). With a higher black mass cost, it is  
358 critical to efficiently utilize black mass during the bioleaching process. This does not necessarily mean that  
359 the goal should be recovery of 100% of the metal content. For example, decreasing the pulp density could  
360 result in higher metal recovery rates and higher revenue generation, but it would have to be balanced with  
361 increasing acid consumption and associated costs.

362 It should be mentioned that the transportation of EOL LIBs has a significant impact on black mass  
363 purchasing cost. Due to the flammability of the electrolyte and lithium, spent LIBs are classified as class 9  
364 hazardous materials, and the transportation cost ranges between \$6.28/ton-mile and \$9.4/ton-mile for  
365 medium and heavy heavy-duty trucks (vs. \$0.14–\$0.15 per ton-mile for non-hazardous materials),  
366 significantly affecting the economic viability of recycling processes (Dai et al., 2019). Recent  
367 advancements in deactivating EOL LIBs at the point of collection can improve the economic viability of  
368 the recycling process. For example, OnTo Technology has been developing a new method for preventing  
369 thermal runaway, which can render batteries, including LIBs, inert (Sloop, 2020). Additionally, optimizing  
370 the reverse logistics network for the collection of EOL products from end-users and transporting them to  
371 downstream processors for recycling has enormous potential for lowering transportation costs and  
372 minimizing environmental impacts.

373 *Iron sulfate:* Ferrous iron was used as the reducing agent, and its high consumption in the process made it  
374 the second-largest cost (10% of the total cost). The breakeven price for iron sulfate is ~\$1.7/kg (~3.4 times  
375 higher than the assumed price), above which bioleaching of LIB black mass is not economically viable. In  
376 a recent study by Ghassa et al. (Ghassa et al., 2021), the possibility of reducing iron sulfate consumption  
377 by utilizing iron scraps present in spent LIBs was considered. EOL LIBs have a considerable amount of  
378 iron in the shells and connectors that could be used in the bioleaching process to decrease the recycling  
379 cost. However, current methods for producing black mass from spent LIBs incorporate treatments such as  
380 magnetic separation to remove iron and steel components (Ekberg and Petranikova, 2015).

381 *Changing cathode chemistry:* Several studies have demonstrated that the cost of manufacturing LIBs has  
382 been decreasing steadily due to a variety of factors, including modification of the battery cathode chemistry  
383 to utilize less expensive materials (e.g., the substitution of cobalt with cheaper nickel or manganese,  
384 adoption of LFP in more applications such as electric buses and power tools) (Ding et al., 2019; Televisory,  
385 2017; Wang and Yu, 2021). The transition to less expensive cathode materials reduces recycling revenue  
386 and challenges the economic viability of emerging recycling technologies. As a result, recycling  
387 technologies must be robust enough to handle a greater variety of spent LIBs and continuously reduce costs  
388 in order to compete with virgin material prices for the evolving LIB industry (Alipanah et al., 2021).

389 *Economic value of bioleached metals:* Profitability is not just determined by the cost factors mentioned  
390 above but also by the revenue generated from bioleaching. The price of cobalt, which is the primary source  
391 of revenue for LIB bioleaching, has fluctuated significantly in recent years (USGS, 2011–2021)(  
392 Supporting Information Figure S1). For instance, oversupply of refined cobalt in the US market pushed its  
393 price down to \$29/kg in 2013. Increasing demand from the rechargeable battery and aerospace industries  
394 increased the price of cobalt dramatically to over \$83/kg in 2018. In 2019, increased supply resulted in a

395 halving of the price. For bioleaching to be economically feasible, the price of cobalt should be \$25/kg or  
396 higher; it was assumed to be \$36/kg in this study based on the 2020 value. Another important factor that  
397 affects the estimated revenue from LIB bioleaching is the uncertainty associated with the 30% discount rate  
398 applied in this study to the recovered metal prices to account for the downstream concentration and  
399 separation process costs. TEA showed that LIB bioleaching could tolerate a discount rate up to 52% to stay  
400 profitable in the best case (i.e., highest observed metal recovery rates), and a discount rate up to 42% in the  
401 worst case (i.e., lowest metal recovery rates).

#### 402 **4. CONCLUSION**

403 The sustainable development scenario developed by the International Energy Agency requires increasing  
404 the supplies of lithium by 18–20 times, nickel by 28–31 times, and cobalt by 17–19 times in 2050 compared  
405 to their production levels in 2020 (Xu et al., 2020), raising the pressure on geological reserves. Recycling  
406 of spent LIBs is a promising approach to reduce the supply-demand gap for LIB materials. The existing  
407 literature focused on maximizing metal recovery efficiency from LIB recycling which could jeopardize the  
408 economic and environmental sustainability. Finding the optimal balance among metal recovery efficiency,  
409 processing cost, and environmental footprint is fundamental for sustainable development of bioleaching  
410 technologies.

411 This study identified an economically feasible and environmentally competitive processing condition for a  
412 novel bioleaching technology, which will help the technology advance to a higher technology readiness  
413 level and industry adoption. At the optimal pulp density of 2.5%, gluconic acid concentration of 75 mM,  
414 leaching temperature of 55°C, and leaching time of 30 h, we recovered up to 86% of cobalt, 100% of  
415 lithium, 100% of manganese, and 84% of nickel from LIB black mass. Upon scaling up the process to  
416 bioleaching 10,000 t/y of black mass, we envision a \$110M NPV for a 30-year operation and a reduction  
417 in global warming potential by up to 8 times compared to other reported leaching technologies.

418 The economic feasibility of this bioleaching approach is highly dependent on the black mass purchasing  
419 price, which is affected by the costs of collection and transportation of EOL LIBs for black mass production.  
420 The price of iron sulfate and its consumption also have significant impacts on the economics of the process.  
421 Modifying the existing black mass production processes to increase ferrous iron content in black mass could  
422 decrease iron sulfate consumption during bioleaching and improve the economic and environmental  
423 feasibility.

424 Future efforts to improve and extend the analyses described here could include incorporating the changing  
425 market dynamics for black mass availability, demand for recovered materials, and the price of recovered  
426 materials into TEA and LCA. Future chemistry changes for cathode materials and integration of  
427 downstream purification and separation processes such as solvent extraction should be considered for more  
428 accurate prognostication of economic and environmental prospects for LIB bioleaching. Sustainable  
429 implementation of bioleaching processes would benefit from optimal design of the reverse logistics supply  
430 chain. A LIB bioleaching company should consider all the stakeholders in the supply chain (e.g., black  
431 mass producers and metal refining companies) to optimize the facility location, material and transportation  
432 flows, and business operation (e.g., inventory).

#### 433 **5. ACKNOWLEDGMENT**

434 We thank Idaho National Laboratory scientists C. McNamara for laboratory assistance, M. Shi for AAS  
435 analysis, K. Schaller for HPLC analysis, Dallas M. McCary for designing graphic abstract, and L. Aldana  
436 Diaz for sharing black mass, a gift from Retriev Technologies (Diaz et al., 2020). This research was

437 supported by the Critical Materials Institute, a Department of Energy Innovation Hub led by the U.S.  
438 Department of Energy's Ames Laboratory and supported by DOE's Office of Energy Efficiency and  
439 Renewable Energy's Advanced Manufacturing Office, which supports early-stage research to advance  
440 innovation in U.S. manufacturing and promote American economic growth and energy security. CMI seeks  
441 ways to reduce supply risks on rare-earth metals and other materials critical to the success of clean energy  
442 technologies. Work is conducted under DOE Idaho Operations Office Contract DE-AC07-05ID14517 and  
443 University of Arizona contract DE-AC02-07CH11358; Agreement No. 26110-AMES-CMI. Accordingly,  
444 the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form  
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