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## Conventional vs. direct vs. electrochemical lithium extraction: a holistic TEA–LCA of lithium carbonate production from spodumene

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Meeting surging lithium-carbonate demand requires routes that are efficient, low-impact, and economically viable. We compare the conventional sulfuric-acid process from  $\beta$ -spodumene with a patented direct lithium extraction (DLE) process from  $\alpha$ -spodumene based on low-temperature NaOH roasting, room-temperature water leaching, and CO<sub>2</sub> precipitation, and a newly developed electrochemical direct leaching (EDL) method, which bypasses thermal conversion by operating at room temperature in dilute acid under applied potential. A cradle-to-gate life cycle and technoeconomic analysis shows that DLE reduces global warming potential by 59% ( $2.76 \times 10^3$  vs.  $6.72 \times 10^3$  kg CO<sub>2</sub>-eq) and lowers acidification and smog through reduced heat demand (325 °C vs. 1100 °C) and elimination of sulfuric acid. EDL shows further reductions in fossil fuel depletion, global warming, and respiratory effects. Economically, at 1 t per day, DLE delivers 41% ROROI and \$18.9M NPV, outperforming the conventional route (35% and \$16.0M), while EDL remains profitable (24% ROROI; \$12.5M NPV) though burdened by higher CAPEX. Annual OPEX is lowest for DLE (\$1.6M vs. \$1.9M conventional; \$2.0M EDL). Monte Carlo simulations confirm DLE's superior profitability (+\$652k per year mean profit vs. −\$120k per year conventional) at a small-scale process, while EDL provides intermediate returns (+\$416k per year) and the lowest downside risk. Together, these results show that integrated TEA–LCA assessments capture trade-offs among emerging processes and support responsible innovation toward greener, more resilient critical-mineral extraction.

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### Green foundation

1. This study integrates technoeconomic and life cycle assessments with stochastic modeling to compare conventional, direct lithium extraction (DLE), and electrochemical direct leaching (EDL) of spodumene. It provides a holistic evaluation of profitability, risk, and environmental trade-offs across emerging lithium production technologies.
2. DLE reduces global warming potential by 59%, lowers acidification and smog impacts, and shortens payback periods due to reduced thermal energy and acid use. EDL eliminates high-temperature calcination entirely, which decreases fossil fuel depletion and respiratory effects, while maintaining competitive profitability. Although EDL introduces new burdens from electricity and electrode materials, both advanced methods deliver substantially improved environmental and economic sustainability compared with the conventional sulfuric-acid bake pathway.
3. Future improvements could involve replacing NaOH with greener reagents, using low-carbon electricity for electrochemical operations, and developing recyclable electrodes to reduce ecotoxicity impacts in the EDL method.

## 1 Introduction

With the implementation of the United Nations Sustainable Development Goals (SDGs),<sup>1</sup> lithium (Li) has taken on a

central role in the coal phase-out and greener energy phase-in. This is due to its exceptional properties such as low density ( $0.534 \text{ g cm}^{-3}$ ), high specific capacity ( $3.8 \text{ Ah g}^{-1}$ ), and high electrochemical potential ( $3 \text{ V}$ )<sup>2</sup> and widespread use in lithium-ion batteries (LIBs), the leading technology for electric vehicles (EVs) and grid-scale energy storage.<sup>3</sup> The share of mined Li used in battery manufacturing has increased from approximately 40% in 2016 to over 75% by 2023 and is projected to exceed 90% as EV adoption continues to accelerate.<sup>4</sup> For example, between 2018 and 2024, Li consumption nearly doubled, reaching about 1.1 million tons of lithium carbonate

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equivalents (LCE) in 2024, roughly equal to all demand in 2018.<sup>5</sup> This spike in demand led to extreme market volatility, with lithium carbonate ( $\text{Li}_2\text{CO}_3$ ) prices reaching record highs of over \$80 000 per ton in late 2022, before plunging by more than 70% as new supply outpaced demand growth. By mid-2025, prices had stabilized at around \$10 000 to \$12 000 per ton of LCE. Such market volatility reinforces the need for cost-effective and sustainable Li production expansion in the next decade. Projections indicate global Li demand could reach 3.3 million tons of LCE by 2030,<sup>6</sup> roughly triple current levels. Although Li resources are not considered scarce and are unlikely to pose a limiting constraint this century,<sup>7</sup> ensuring a stable supply at an affordable cost and with minimal environmental impact has become a strategic priority to support the clean energy industry.<sup>8</sup>

Lithium is mainly sourced from brine deposits (e.g., Li-rich brine in Argentina, Bolivia, and Chile) and hard-rock minerals (e.g., spodumene and lepidolite). While Li extraction from brine is generally less energy-intensive, hard-rock mining (e.g., in Australia and China) has gained prominence due to high Li content and faster production timelines.<sup>9,10</sup> By 2024, spodumene accounted for most of the global Li production, with Australia and China together responsible for roughly 54% of supply, while brine operations in Argentina and Chile contributed around 28%.<sup>10</sup> Given spodumene's dominant role in global supply, considerable research efforts have focused on improving extraction methods. Chief among them is the conventional high-temperature calcination and sulfuric acid ( $\text{H}_2\text{SO}_4$ ) baking based Li extraction method from spodumene,<sup>11</sup> which involves a series of complex and energy-intensive steps detailed in section 2.1 (hereinafter referred to as the conventional method). The high calcination temperatures above 1000 °C utilized in the conventional method require substantial energy consumption and operating costs. Maintaining a uniform temperature profile in large rotary kilns is also challenging (i.e., due to temperature gradient and variations in particle size), resulting in inefficiencies and reduced conversion yields.<sup>12–14</sup> Kiln operations also pose concerns, such as the formation of carcinogenic arsenic trioxide and the volatilization of other trace elements at high temperatures.<sup>12,15,16</sup> In addition to conventional heating, hybrid approaches such as microwave-assisted calcination followed by sulfuric acid leaching have been explored to reduce energy and enhance the kinetics of  $\alpha \rightarrow \beta$  spodumene phase transition.<sup>13,17</sup> While promising, large-scale microwave processing is yet to be developed for industrial applications. Beyond the calcination stage, the sulfuric acid baking step presents further limitations. It consumes vast quantities of concentrated  $\text{H}_2\text{SO}_4$ , raising both cost and sustainability issues.<sup>18</sup> This also creates severe corrosion problems and requires significant chemical inputs during neutralization and purification stages further inflating reagent consumption and waste generation.

The core challenge in Li extraction from spodumene is breaking the aluminosilicate matrix and liberating Li without exorbitant energy or acid use. To address this, several studies

have explored the direct extraction of Li from  $\alpha$ -spodumene using alternative techniques such as dry chlorination, hydrofluoric (HF) decomposition, microbial digestion, mechanical activation, and alkaline digestion with the goal of eliminating the need for high-temperature calcination and concentrated acid usage.<sup>13,19–21</sup> However, each of these approaches comes with its own set of limitations that hinder practical implementation. Dry chlorination requires high-temperatures (~1000 °C), suffers from Li loss as volatile  $\text{LiCl}$ , and requires chlorine-handling infrastructure and corrosion-resistant equipment.<sup>22,23</sup> The use of HF is limited by its toxicity,<sup>24</sup> while microbial digestion is hindered by slow kinetics.<sup>12</sup> The use of NaOH-induced mechanical activation followed by water leaching is still in the emerging stage.<sup>19</sup> Although it operates at low temperatures, challenges such as low Li recovery and high NaOH consumption remain unresolved.

Alkaline digestion with sodium hydroxide (NaOH) or potassium hydroxide (KOH) at 250 °C followed by acid leaching shows promise, though eliminating the acid is preferable.<sup>20,25</sup> In addition, Xing *et al.* demonstrated that mixing  $\alpha$ -spodumene with a concentrated NaOH solution and heating to 250 °C in an autoclave achieved 95–96% Li extraction within 2 hours.<sup>21</sup> This shows that alkaline digestion in an autoclave can directly extract Li from  $\alpha$ -spodumene at moderate temperatures (200–250 °C) under pressure. The process converts spodumene into soluble Li while the aluminosilicate structure transforms into an inert zeolite (hydroxysodalite). By avoiding the 1100 °C calcination, this direct alkaline leaching route significantly reduces the peak thermal requirement. However, the use of autoclaves in alkaline digestion presents practical limitations, including high base consumption (much of the reagent is consumed in forming silicate byproducts), high capital and operational costs, heightened safety measures, and challenges in scaling up to industrial production.<sup>18</sup> These factors further highlight the need for low-temperature, acid-free methods for efficient Li recovery from  $\alpha$ -spodumene. In this regard, the authors have developed a patented two-staged process (detailed in section 2.1) involving NaOH roasting at 325 °C followed by room-temperature water leaching (herein after referring to as the direct lithium extraction (DLE) method), achieving over 99% Li extraction.<sup>12,13</sup> This method offers several key advantages, including the relatively low temperature operation compared to the conventional method, the elimination of acid, the conversion of  $\alpha$ -spodumene to water-soluble Li-bearing phases, and the enabling of rapid Li leaching kinetics at ambient conditions.

Beyond demonstrating technical improvements, it is equally important to evaluate the economic viability and environmental sustainability of emerging lithium extraction processes.<sup>6,26</sup> In this regard, techno-economic and environmental assessments are used for this critical evaluation. To be considered sustainable, any alternative route must not only improve technical performance but also reduce costs and environmental burdens compared to conventional methods.<sup>27</sup> Techno-economic analysis (TEA) estimates capital and operating costs, as well as economic metrics such as Net Present



Value (NPV). In contrast, Life Cycle Assessment (LCA) quantifies environmental impacts, including emissions and resource use, across the entire production chain.

Huang *et al.* performed an LCA of lithium recovery from geothermal brines, revealing the carbon footprint advantage of brine-based routes but providing limited insight into cost competitiveness.<sup>6</sup> Similarly, Lappalainen *et al.* (2024<sup>28</sup>) simulated soda-based processing of spodumene and reported 16–72% lower environmental impacts compared to conventional acid baking due to avoided sulfuric acid use, yet no economic evaluation was included. In another study, authors assessed decarbonized lithium production from sedimentary rocks with emphasis on carbon reduction but did not integrate life-cycle trade-offs.<sup>27</sup> TEA and LCA are often applied separately in Li studies, recent research emphasizes the value of combining both. Results of recent LCA studies show that hard-rock Li has a higher carbon footprint due to energy-intensive calcination and reagent use, while brine-based Li poses challenges related to water consumption and land use.<sup>8</sup> A detailed LCA by Lappalainen *et al.* found that a soda-based process had 16–72% lower impacts across multiple categories compared to the conventional method of Li extraction from hard-rock, particularly due to avoided acid use and reduced reagent.<sup>28</sup> Electrochemical leaching is another solution that can address the challenges associated with Li extraction from ores.<sup>29</sup>

Compared to chemical leaching methods, it surpasses the dependence on high temperature and concentration of the leaching agent to activate the reaction. However, the electrochemical direct leaching (EDL) is limited by its heterogeneous nature, which typically leads to large overpotentials, side reactions, and poor faradaic efficiency.<sup>30</sup> Soluble leaching promoters such as H<sub>2</sub>O<sub>2</sub>,<sup>31</sup> and SO<sub>2</sub><sup>32</sup> are added as the homogeneous agent to promote electron transfer kinetics. In contrast to environmental assessment, economic data remain limited; however, primarily TEA studies indicate that while new methods may reduce operational costs, they require higher capital investment, especially in systems such as calcination followed by CO<sub>2</sub> pressure leach, precipitation, and Li<sub>2</sub>CO<sub>3</sub> purification.<sup>33</sup> To put this into perspective, the costs of current Li production from geothermal brine vary widely, ranging from \$2 to \$9 per kg LCE, depending on the method and geographical location.<sup>6</sup> In another study, conventional lithium extraction from hard rock deposits was economically compared with extraction from brine and sedimentary sources to evaluate options for strengthening the U.S. domestic supply chain.<sup>34</sup> Furthermore, Xu *et al.* proposed a cost-effective, rapid, and acid-free method for extracting lithium from spodumene ore. This process achieves high-purity lithium chloride and excellent yield within seconds, significantly reducing energy consumption, waste generation, and processing costs compared to conventional method.<sup>35</sup>

Despite growing attention to the chemistry and technical feasibility of DLE and EDL, the literature lacks an integrated and uncertainty-aware framework that simultaneously captures both environmental and economic dimensions of these emerging technologies. Previous studies typically apply LCA and

TEA alone, providing a fragmented view that limits understanding of trade-offs between resource use, emissions, and profitability. In addition, most assessments are deterministic, failing to address how variations in energy price, lithium market value, or process efficiency affect sustainability outcomes. The central contribution of this study is therefore not only the comparison of three lithium extraction technologies but the implementation of an integrated TEA–LCA framework under uncertainty through Monte Carlo simulations. This approach enables the quantification of both cost and environmental risks, offering a more realistic and decision-relevant evaluation of emerging lithium extraction methods. By linking process performance, economic robustness, and life-cycle impacts, this work fills a key gap and supports the development of responsible and sustainable lithium production routes. Given the challenges that greener, but more costly, methods face in competitive markets, this study identifies key cost drivers and environmental hotspots to inform the future scaling up of cleaner Li production technologies.

## 2 Methodology

### 2.1 LCE production approaches

The production of LCE from spodumene ore through the conventional method is a well-established sequence of physical, thermal, and hydrometallurgical steps, as illustrated in Fig. 1. This include high-temperature (~1100 °C) calcination of  $\alpha$ -spodumene concentrate (~7.5 tons) for about 2 hours to convert into more porous and reactive  $\beta$ -spodumene phase, followed by concentrated H<sub>2</sub>SO<sub>4</sub> (93%) baking for 30 minutes using 145% of the stoichiometric requirement at 250 °C to form lithium sulfate (Li<sub>2</sub>SO<sub>4</sub>) *via* ion exchange with hydrogen.<sup>36</sup> Lithium is then recovered from Li<sub>2</sub>SO<sub>4</sub> by water leaching using a liquid-to-solid ratio of 1.85 at 90 °C for 1 hour. Calcium carbonate is added to neutralize excess acid (*i.e.*, to a pH around 6–7) and remove impurities (*e.g.*, Al and Fe). Finally, Na<sub>2</sub>CO<sub>3</sub> is used to precipitate Li<sub>2</sub>CO<sub>3</sub> (*i.e.*, precursor to the production of LiOH).<sup>37</sup> Stirring is applied at approximately 200 rpm, and under these conditions, Li recovery at this stage reaches around 96%.<sup>36</sup>

Leaching is followed by solid–liquid separation using a filter press (*i.e.*, yielding approximately 13 tons of Li-bearing solution and 9 tons of dry filter cake). The filtrate is then purified by sequential addition of lime milk (Ca(OH)<sub>2</sub>) and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) to raise the pH to 12 and precipitate Mg in the form of its hydroxide.<sup>18,38</sup> This step is conducted in a stirred reactor and is designed to ensure a clean Li solution ready for precipitation. The slurry is filtered again to remove the newly formed impurities and produce a dilute Li<sub>2</sub>SO<sub>4</sub> solution, which is then concentrated by evaporation. Heating to 100 °C removes approximately 80% of the water, reducing the volume of solution. The concentrated solution is stored in buffer tanks before being transferred to precipitation reactors.

In the first precipitation step, Na<sub>2</sub>CO<sub>3</sub> is added to the Li-rich solution at 95 °C under agitation at 200 rpm to precipitate Li<sub>2</sub>CO<sub>3</sub>. The yield of this step is typically 75%, and the result-



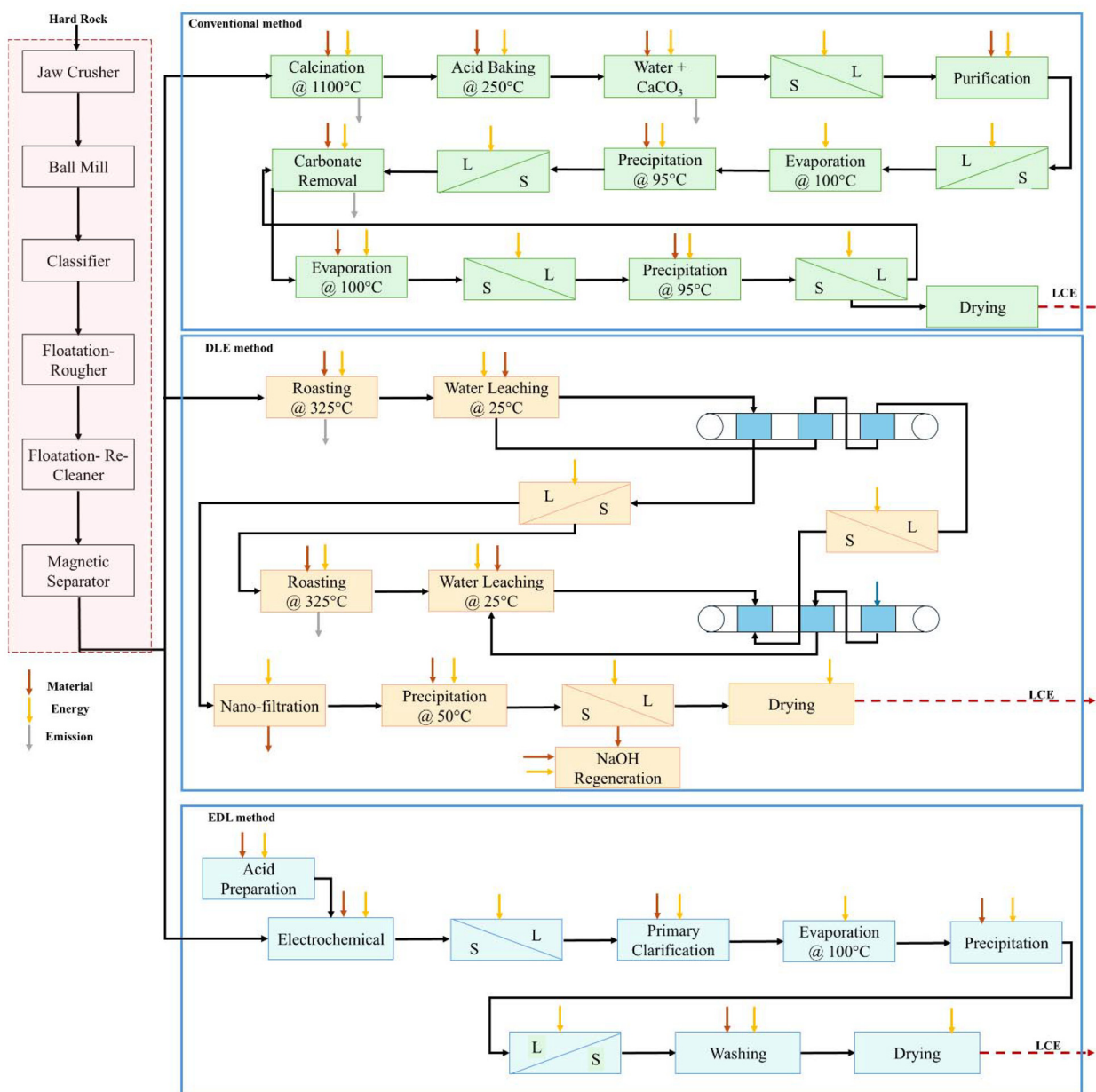


Fig. 1 System boundaries (blue bond) for producing 1 ton of LCE from spodumene via the conventional, DLE, and EDL methods (cradle-to-gate). All energy and reagent inputs within the process boundary are shown; transportation is excluded.

ing slurry is separated *via* filtration, recovering approximately 80% of LCE product. The remaining Li in the solution is recovered through a secondary loop. First, residual carbonate is removed with the addition of  $\text{H}_2\text{SO}_4$  to the filtrate, releasing  $\text{CO}_2$  and converting carbonate species into soluble sulfates. The resulting solution is then evaporated and crystallized, leading to the formation of sodium sulfate ( $\text{Na}_2\text{SO}_4$ ) and a concentrated Li stream. Sodium sulfate is removed *via* filtration, and the remaining Li is precipitated by adding  $\text{Na}_2\text{CO}_3$ . This second-stage precipitation recovers an additional amount of crude LCE. Finally, all recovered  $\text{Li}_2\text{CO}_3$  is dried to obtain battery-grade LCE (*i.e.*, 1 ton).

The recent patented direct lithium extraction (DLE) method from  $\alpha$ -spodumene for producing LCE involves alkaline roasting, water leaching, nanofiltration, and  $\text{CO}_2$  precipitation (illustrated in Fig. 1). The material and energy balances were developed for a functional unit of 1 ton of battery-grade LCE. The process begins with the chemical activation of  $\alpha$ -spodumene through alkaline roasting using NaOH at 325 °C. This step replaces the conventional high-temperature calcination (1100 °C) and  $\text{H}_2\text{SO}_4$  baking (250 °C) with a lower-temperature NaOH-based reaction to form water-soluble lithium-sodium silicates. The reaction time is approximately 2 hours in a rotary kiln. The mass ratio of NaOH to spodumene is





1.5 : 1 by weight.<sup>12,13,39</sup> A 100% transformation to water soluble Li compounds is assumed, with no Li losses in this step. Following roasting, the treated solid is subjected to ambient-temperature water leaching with a solid-to-liquid ratio of 35% by weight. This step solubilizes Li into aqueous solution. Agitation is maintained at 100 rpm for a short residence time of 1 minute.<sup>12,13</sup> The leachate contains  $\text{Li}^+$ ,  $\text{Na}^+$ , and a small fraction of  $\text{Al}^{3+}$ , as well as other impurities. The Li recovery in this stage is assumed to be 75%.<sup>12,13,39</sup> The slurry is filtered to separate the Li-rich aqueous solution from undissolved solids. The filtrate proceeds to nanofiltration, while the solid residue (mainly aluminosilicates) is roasted again to recover the remaining Li. The remaining solid residue is subjected to a second-stage NaOH roasting at 325 °C for 10 minutes with a lower NaOH : solid ratio of 1 : 1. This step maximizes Li recovery, as some Li remains entrapped in the aluminosilicate matrix after the first roasting. A second water leaching step is applied to the newly roasted solids at ambient temperature and with stirring at 200 rpm. The solid-to-liquid ratio is reduced to 30% to increase extraction efficiency. This step recovers additional Li (~24%) into the solution, bringing the cumulative Li extraction yield (from two-stage roasting and leaching) to over 99%. The resulting solids, rich in alumina and other multivalent cations, can be processed further for aluminum recovery or landfilled, depending on the downstream processing flow sheet. The combined aqueous streams from stage 1 and 2 form the input to nanofiltration. To purify the Li-rich solution before precipitation, nanofiltration (NF) is used to separate monovalent ions from multivalent ions ( $\text{Al}^{3+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ). Thin-film polyamide membranes, such as NF270, are used under room-temperature conditions and operated at a pressure of 10–12 bar. The Li recovery in the NF permeate is 98%, while more than 90% of multivalent ions are rejected, significantly improving the purity of the final product.<sup>40,41</sup>

Lithium carbonate is precipitated from the NF-permeate solution by sparging  $\text{CO}_2$  gas at 40 °C in the presence of  $\text{Li}_2\text{CO}_3$  seed crystals, which promote nucleation and particle growth. Carbon dioxide reacts with  $\text{LiOH}$  in the solution to form solid LCE.<sup>42–44</sup> The solution is agitated for 60 minutes. The resulting slurry contains  $\text{Li}_2\text{CO}_3$  crystals and a mother liquor, from which the  $\text{Li}_2\text{CO}_3$  is recovered using a filter press, which produces a moist cake with <10% moisture. The cake is optionally washed with decarbonized water to remove residual impurities and dried at 100 °C in a tray or rotary dryer to yield a final product of 1 ton of battery-grade LCE with more than 99.5% purity. The NaOH regeneration system consists of two stirred tank reactors operating in series. The first reactor (60 °C, 30 min residence time) converts spent NaOH to sodium carbonate using  $\text{CO}_2$ . The second reactor (80 °C, 60 min residence time) employs lime causticizing to regenerate NaOH with 98% efficiency, requiring only 2% fresh NaOH makeup.<sup>45,46</sup> NaOH regeneration requires 1970 kWh per day of electricity (equivalent to 43% of total plant electricity use) and lime as the primary reagent.

The electrochemical direct leaching (EDL) pathway, as shown in Fig. 1, bypasses the high-temperature calcination of

conventional acid baking by directly extracting lithium from ground  $\alpha$ -spodumene ore at ambient temperature using a sulfuric acid electrolyte and an *in situ* promoter. In this process, first,  $\alpha$ -spodumene ore is crushed and ground to a particle size of about 50  $\mu\text{m}$  to increase reactivity.<sup>47</sup> After that, the ground solids are suspended in an aqueous electrolyte of 0.5 M  $\text{H}_2\text{SO}_4$  at room temperature, with agitation at 500 rpm. This process is followed by the addition of hydrogen peroxide ( $\geq 0.1$  wt%) to promote electron transfer between the ore and the electrolyte. This process utilizes a 3D graphene-oxide/carbon-felt current collector reinforced with Nafion and decorated with Au nanoparticles, which catalyzes *in situ*  $\text{H}_2\text{O}_2$  formation under an  $\text{O}_2$ /air sparge.<sup>47</sup> Electrochemical leaching is then performed in sparged and agitated reactors operated at a potential of 0.95 V, which is the optimal condition for maximizing lithium dissolution. After 12 h, 92.2% of the lithium is leached into solution as  $\text{Li}^+$  ions, leaving behind an Al–Si residue. Solid–liquid separation is performed with a thickener and pressure filtration to yield a clear  $\text{Li}_2\text{SO}_4$ -bearing liquor. The solution is then subjected to impurity removal and concentration steps. In the first purification stage, lime milk is added to raise the pH to above 11, which leads to precipitating divalents ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) as hydroxides, which are removed by filtration.<sup>48</sup> The clarified solution is concentrated by evaporation at 100 °C to reduce its volume and increase the lithium concentration.

LCE is precipitated from the concentrated sulfate solution by adding soda ash. This step is performed at 95 °C and pH around 10, with seed crystals of LCE added to enhance particle growth.<sup>37,49</sup> The resulting slurry is filtered, producing LCE cake. Hot washing with DI water at 90–95 °C removes entrained mother liquor and improves purity. Finally, the washed cake is dried in a tray or rotary dryer at 100 °C, yielding 1 ton of battery-grade LCE (>99.5% purity). The EDL parameters were adopted from Zhang *et al.*<sup>47</sup> Based on this study, a lithium leaching efficiency of 92.2% and faradaic efficiency of 83% at an optimized potential of 0.95 V. Lower voltages (0.8–0.9 V) resulted in incomplete Li extraction (76–88%), whereas higher potentials (1.1–1.2 V) marginally increased leaching efficiency but significantly raised energy demand (up to 3.1 kWh  $\text{kg}^{-1}$   $\text{Li}_2\text{CO}_3$ ).<sup>47</sup> To maintain consistency with experimentally verified optimum conditions, the 0.95 V configuration was selected for the present TEA–LCA comparison. The EDL inventory includes the electrode material dosage, consisting of gold nanoparticles (0.05 kg per batch) and carbon felt (7.54 per batch), and their cradle-to-gate production impacts were modeled using the background LCA database.

## 2.2 LCA method

This study follows the ISO 14040/44-based LCA framework, consisting of four key phases: (1) goal and scope definition, (2) life cycle inventory (LCI), (3) life cycle impact assessment (LCIA), and (4) interpretation. The goal of LCA is twofold: first, to quantify the environmental burdens associated with energy and chemical inputs across all process stages of LCE production from spodumene; and second, to compare the



environmental performance of the conventional, EDL, and DLE methods using a consistent and comprehensive approach. The system boundaries are defined as cradle-to-gate, covering all steps from concentrated spodumene input to final  $\text{Li}_2\text{CO}_3$  product, excluding upstream mining and mineral processing as well as downstream battery integration. A visual representation of the LCA boundary is provided in Fig. 1.

The functional unit for this study is 1 ton of battery-grade LCE. Inventory data were compiled from experimental process mass and energy balances (developed as part of this study), with additional background data sourced from the Ecoinvent 3.9 database *via* SimaPro 9.4 software. Direct process inputs include thermal and electrical energy, reagents (*e.g.*,  $\text{H}_2\text{SO}_4$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{NaOH}$ ,  $\text{CO}_2$ ), process water, and emissions. Equipment use, maintenance, and capital infrastructure are excluded from the system boundary due to their marginal contribution per functional unit.

The impact assessment was conducted using the TRACI 2.1 method (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts), specifically the US2008 sub-method within SimaPro. TRACI, developed by the U.S. Environmental Protection Agency, provides a midpoint-oriented characterization of environmental impacts across multiple relevant categories. These include global warming potential (kg  $\text{CO}_2$  eq.), acidification (kg  $\text{SO}_2$  eq.), eutrophication (kg N eq.), smog formation (kg  $\text{O}_3$  eq.), carcinogenic and non-carcinogenic toxicity (CTUh), respiratory effects (kg  $\text{PM}_{2.5}$  eq.), ecotoxicity (CTUe), and fossil fuel depletion (MJ surplus energy). These indicators offer a robust and regionally relevant assessment of environmental trade-offs between the two LCE production routes. The cradle-to-gate scope in this study covers all foreground and background processes from the delivery of concentrated spodumene feed to the production of 1 ton of battery-grade lithium carbonate. Construction, decommissioning, and transportation of inputs to the processing facility and internal handling are considered negligible at the modeled scale and therefore excluded.

### 2.3 TEA method

The techno-economic analysis (TEA) of LCE production from spodumene was performed for three process configurations: the conventional, EDL, and DLE methods. The models were developed for an industrial-scale facility operating 350 days per year over a 15-year project life, with production scales ranging from 1 to 10 tons of battery-grade LCE per day. The analysis accounted for U.S.-based economic parameters, assuming a place factor of one, to reflect domestic labor, utility, and regulatory costs. The project lifespan of 15 years was selected to reflect the typical service period of lithium carbonate production facilities, accounting for capital depreciation, equipment renewal, and market investment horizons commonly used in prior TEA studies.<sup>50,51</sup> The operational lifespan of 350 days per year represents standard continuous industrial operation, allowing for routine maintenance downtime and process calibration.

Capital expenditure (CAPEX) included equipment costs, installation, and indirect capital costs, while operating expenditure (OPEX) was divided into direct and indirect categories. Direct OPEX covered variable costs, including reagents (*e.g.*,  $\text{H}_2\text{SO}_4$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{NaOH}$ ), process water, electricity, and natural gas, for calcination, roasting and leaching processes. Indirect OPEX accounted for labor, maintenance, insurance, and waste management. An hourly wage of \$30 was used to estimate operational labor costs, and labor requirements were scaled based on process complexity and automation level. All input prices and costs were assumed to remain constant over the plant's lifetime.

Economic performance was evaluated using the present net value (NPV), rate of return on investment (ROROI), and payback period. The NPV was calculated (eqn (1)) by discounting the net cash flows at an 8% rate over the project's life, applying a 25% tax rate. The ROROI was defined as the average annual non-discounted net cash flow divided by the fixed capital investment (FCI). The LCE product price was introduced as a stochastic variable in Monte Carlo simulations to assess economic robustness under market uncertainty, alongside fluctuating electricity and gas prices and varying production scales. All assumptions, formulas, and key process parameters used in the TEA model are detailed in the SI Tables S3–S8. The Rate of Return on Investment (ROROI), reflecting the profitability of the process in relation to FCI, was also calculated using eqn (2).<sup>52</sup>

$$\text{NPV} = \sum \text{discounted cumulative cash position} \quad (1)$$

$$\text{ROROI} (\%) = \frac{\sum \text{nondiscounted cash flow/plant life time}}{\text{FCI}} \times 100 \quad (2)$$

### 2.4 Monte Carlo simulation and sensitivity analysis

To assess the economic viability of LCE production under uncertainty, a Monte Carlo simulation was conducted. The analysis focused on three production scenarios: 1, 5, and 10 tons of LCE per day, considering key economic variables, including the market price of LCE and combined utility costs (electricity and natural gas). According to the USGS 2025 report,<sup>10</sup> the price of LCE fell sharply from \$46 000 per ton in 2023 to \$14 000 per ton in 2024. If this downward trend continues, prices could approach \$5000 per ton by the end of 2025. Therefore, for the current study, we selected a range of \$5 000–\$15 000 per ton for LCE costs, which reflects recent market dynamics and provides a realistic basis for our Monte Carlo simulation. The industrial natural gas cost ranged from \$0.08 to \$0.797 per cubic meter<sup>53</sup> and the industrial electricity cost varied from 5.51 ' per kWh to 32.3 ' per kWh.<sup>54</sup>

For each production scale, 1000 simulation runs were performed by generating random input values using the RAND() function in Excel and transforming them to the desired ranges. The resulting output distributions of profit were analyzed statistically and graphically. Histograms were generated



to visualize the spread of economic outcomes, while cumulative distribution functions (CDFs) estimated the probability of achieving a positive profit. Sensitivity analysis was conducted by calculating Pearson correlation coefficients between input variables and profit, with tornado charts ranking the influence of LCE price and utility cost. Additionally, scatter plots of each input variable *versus* profit were used to examine the nature and strength of their relationships. This simulation-based approach provided a robust framework for evaluating economic risk and identifying the dominant cost and revenue drivers in LCE production under uncertainty.<sup>55</sup> Fig. S1 illustrates the Monte Carlo simulation procedure used to assess the uncertainty in LCE production.

## 3 Results and discussion

### 3.1 Environmental insights

A comprehensive visual comparison of the life cycle environmental impacts for the conventional, DLE, and EDL methods for LCE production from spodumene is provided in Fig. 2a–d, supported by detailed impact breakdowns in Tables S9–S11. These figures reflect results from TRACI midpoint indicators and clearly illustrate how each method distributes environmental burdens across inputs and impact categories.

The conventional method for lithium extraction from spodumene shows a greater environmental burden in the extraction phase, which accounts for 57% of the normalized impact. This is primarily due to the energy-intensive processes required for roasting and leaching, resulting in high contributions to categories such as smog, ecotoxicity, and acidification (Fig. 2a). The purification step, while still significant at 43%, benefits from more optimized process controls but remains a notable source of impact due to chemical use and residual emissions. The DLE method concentrates a larger proportion of environmental impact in the extraction step, contributing 62% of the total normalized effect. This is largely due to the intensive chemical inputs and energy required for direct lithium separation from mined spodumene. While DLE can offer advantages in selectivity and potential reductions in some process emissions, it still results in significant environmental burdens associated with extraction-phase activities. The purification phase accounts for the remaining 38%, indicating that DLE improves purification efficiency compared to other methods (Fig. 2b). From an environmental perspective, NaOH regeneration results in measurable hotspot within the DLE system. The unit consumes up to 43% of the total electricity use of the plant. Across different environmental impact categories, the electricity used for NaOH recovery contributes 0–18% of category-level impacts. Lime consumption is the dominant material-related contributor, with its share ranging from 0.5% to 98.7% across the same categories. Lime significantly drives ozone depletion (98.7%), while contributing between ~1–9% to other categories. In contrast, the EDL method demonstrates a marked reduction in extraction-related impacts, with extraction contributing only 42% to the total.

This reduction is achieved through milder, electrochemical reactions that lower both energy and chemical requirements. However, the purification phase assumes a dominant role in the overall environmental profile, accounting for 58% of the impact (Fig. 2c). In the EDL scenario, electrode production contributes approximately  $1.3 \times 10^5$  CTUe to ecotoxicity representing about 50% of the total impact which reflects the relatively high environmental intensity of the gold and carbon materials used in the electrode.

The comparative LCA results, shown in Fig. 2d, provide a clear visual breakdown of environmental impacts for conventional, DLE, and EDL lithium extraction methods across various categories. Conventional processes (gray) dominate most impact categories, comprising over half of the total in areas like smog (45%), acidification (52%), respiratory effects (51%), ecotoxicity (52%), and fossil fuel depletion (53%). This reflects the high energy consumption and intensive chemical use typical of traditional roasting and leaching techniques. DLE (green) generally offers moderate reductions in several impact categories, accounting for significant shares in ozone depletion (55%), smog (36%), and eutrophication (32%), but tends to shift the burden rather than fully eliminate it. EDL (orange) stands out for its lower contribution to ozone depletion (10%), but its relative impact increases in categories such as global warming (16%), carcinogenics (22%), non-carcinogenics (26%), and ecotoxicity (29%). This suggests that although EDL can mitigate some extraction-related impacts, it may introduce new environmental challenges, especially in downstream purification and resource use.

In addition to the relative contribution plots shown in Fig. 2, the absolute life cycle impact values for all three methods are summarized in Table 1. This dual presentation allows direct comparison of the magnitude of each impact category across processes. As shown, the conventional route has the highest total impacts in nearly all categories, particularly global warming ( $1.3 \times 10^4$  kg CO<sub>2</sub>-eq) and fossil fuel depletion ( $1.4 \times 10^4$  MJ), whereas DLE achieves substantial reductions across most indicators. EDL shows the lowest overall energy-related impacts in almost all impact categories, consistent with trends observed in Fig. 2.

### 3.2 Economic viability

The TEA comparison between the conventional, DLE, and EDL pathways for LCE production methods is summarized in Table 2. The conventional and DLE methods show broadly comparable capital intensities, with CAPEX of \$6.8 million and \$6.3 million, respectively (equivalent to \$1.3 per kg and \$1.2 per kg in the first year). By contrast, the EDL method requires a higher upfront investment of \$9.3 million, with a CAPEX intensity of \$1.8 per kg, reflecting the cost of electrochemical infrastructure.

Operating expenditures (OPEX) reinforce these distinctions. The DLE method achieves the lowest annual OPEX (\$1.6 million), compared to \$1.9 million for the conventional process and \$2.0 million for EDL. The shortest payback period for the DLE method (3.4 years) was achieved compared to the





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**Table 1** Absolute life cycle impact results for the production of 1 ton of LCE from spodumene *via* the conventional, DLE, and EDL methods

Impact category	Unit	Conventional	DLE	EDL
Ozone depletion	kg CFC-11 eq.	$8.9 \times 10^{-4}$	$9.6 \times 10^{-2}$	$2.7 \times 10^{-4}$
Global warming	kg CO <sub>2</sub> eq.	$1.3 \times 10^4$	$6.7 \times 10^3$	$4.0 \times 10^3$
Smog	kg O <sub>3</sub> eq.	$4.9 \times 10^2$	$3.4 \times 10^2$	$2.1 \times 10^2$
Acidification	kg SO <sub>2</sub> eq.	$5.7 \times 10^1$	$2.7 \times 10^1$	$2.4 \times 10^1$
Eutrophication	kg N eq.	$5.0 \times 10^1$	$3.0 \times 10^1$	$1.9 \times 10^1$
Carcinogenics	CTUh	$9.1 \times 10^{-4}$	$4.7 \times 10^{-4}$	$4.0 \times 10^{-4}$
Non carcinogenics	CTUh	$4.5 \times 10^{-3}$	$1.6 \times 10^{-3}$	$2.3 \times 10^{-3}$
Respiratory effects	kg PM <sub>2.5</sub> eq.	$1.2 \times 10^1$	$6.5 \times 10$	$4.1 \times 10$
Ecotoxicity	CTUe	$2.4 \times 10^5$	$7.4 \times 10^4$	$1.4 \times 10^5$
Fossil fuel depletion	MJ surplus	$1.4 \times 10^4$	$7.7 \times 10^3$	$4.5 \times 10^3$

Values represent cradle-to-gate results based on TRACI 2.1 midpoint indicators (red represents the highest value and green represents the lowest value).

**Table 2** Key economic performance indicators for both conventional, DLE, and EDL methods for 1-ton LCE production

KEPI	Conventional	DLE	EDL
CAPEX, million \$	6.8	6.3	9.3
CAPEX intensity, \$ per kg 1-year	1.3	1.2	1.8
OPEX, million \$ per year	1.9	1.6	2.0
Payback period, year	4.6	3.4	6
Lifetime OPEX, million \$	28.6	24.4	30.6
Revenue, million \$ per year	4.9	4.9	4.9
ROROI, %	35	41	24
NPV, million \$	16.0	18.9	12.5

conventional method (4.6 years). However, despite its technical innovations, EDL requires 6 years to recover capital due to higher operating costs and capital intensity.

Financial return metrics further emphasize the relative advantages of DLE. The rate of return on investment (ROROI) for DLE reaches 41%, which is higher than the conventional method (35%) and EDL (24%). Furthermore, DLE secures the highest NPV at \$18.9 million, much higher than both the conventional (\$16.0 million) and EDL (\$12.5 million) LCE production pathways. Revenue potential remains equivalent across all three methods (\$4.9 million per year), underscoring that profitability differences are driven by cost structure and efficiency rather than product price. From an economic standpoint, NaOH regeneration contributes approximately 9.2% of the total OPEX of the DLE facility. This cost arises primarily from (i) the electricity demand of the electrochemical regeneration unit and (ii) the continuous requirement for lime in the causticizing loop.

Fig. 3–5 shows the discounted cumulative cash flow over the plant's lifespan for three conventional, DLE, and EDL methods. It can be seen that DLE has the fastest investment return, resulting in the highest net value of \$19 million. The conventional method yields a median economic performance with an investment of more than 5 years, resulting in an ending value of \$16M. The EDL pathway demonstrates the minimum economic performance, characterized by a longer return on investment and the lowest discounted cash flow after 15 years of operation.

The Sankey diagram visualizing the OPEX breakdown per kg of LCE highlights clear distinctions between the conven-

tional (Fig. 3b), DLE (Fig. 4b), and EDL (Fig. 5b) methods for spodumene processing. In the conventional method, labor and supervisory costs dominate (indirect costs), totaling over 2.5 USD per kg LCE, indicative of the complex, labor-intensive stages such as acid roasting, multi-step purification, and extensive handling of hazardous reagents. In contrast, the DLE and EDL methods reduce labor-related costs, likely due to improved process integration, automation.

Utility and consumable costs further distinguish between the approaches. The conventional method leads to higher utility expenses (0.98 USD per ton) due to its high-temperature calcination (1100 °C) and thermal acid baking. In contrast, the DLE method benefits from lower roasting temperatures (325 °C) and simplified thermal profiles, resulting in a 20% reduction in energy use. EDL poses a minimum utility cost due to its avoidance in the use of the furnace in the process. Consumables are also notably lower in the DLE method (0.96 vs. 1.38 USD per ton in conventional), reflecting its elimination of H<sub>2</sub>SO<sub>4</sub> and increased reliance on reagent recycling. The maximum consumables cost is on EDL due to its need for acid and peroxide agent to accelerate the leaching process.

In the conventional method, operating expenses are primarily concentrated in labor and consumables, followed by utilities, driven by high-temperature steps such as calcination at 1100 °C and acid roasting, as well as multiple separation and purification stages. In contrast, the DLE method has a more reagent-intensive cost structure, with consumables making up 20% of total OPEX. Labor and utility demands are notably lower (36% and 17%, respectively) due to simpler and lower-temperature operations. This method offers a more stable and predictable operating profile with reduced exposure to energy and volatility. In the EDL approach, consumables account for the largest share of OPEX at 43%, followed by labor at 30%. This distribution indicates that reagent consumption is the primary cost driver, and optimizing it would be essential to reduce the overall operational expenses of this method.

### 3.3 Monte Carlo cost uncertainty analysis

A summary of key uncertainty indicators derived from Monte Carlo simulations for the conventional, DLE, and EDL methods through three LCE production scales is presented in Table 3. At the smallest scale (1 ton per day), the conventional method shows a negative average profit (−119.8k\$ per year), showing that low-throughput operations are economically unfavorable under traditional processing conditions. In contrast, both DLE and EDL yield positive profits, 652.4k\$ per year and 415.6k\$ per year, respectively, reminding that advanced leaching approaches can make small-scale operations economically viable. As production scale increases, all three methods benefit from economies of scale, but the magnitude of improvement differs: at 10 tons per day, the conventional method reaches 7.93M\$ per year, EDL reaches 11.90M\$ per year, and DLE achieves the highest profitability at 13.15M\$ per year. This confirms that while DLE is the most profitable overall, EDL offers a strong intermediate pathway, outperforming the conventional method across all scales.



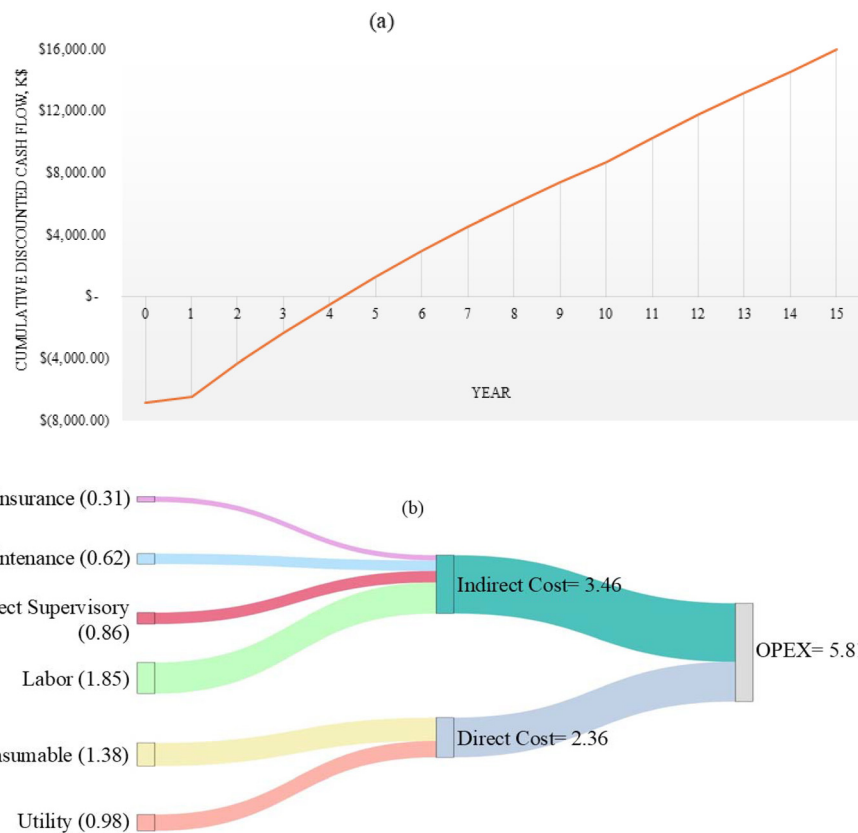


Fig. 3 The discounted cumulative cash flow (15-year) (a) and OPEX (b) values \$ per kg LCE for conventional spodumene-based LCE production method.

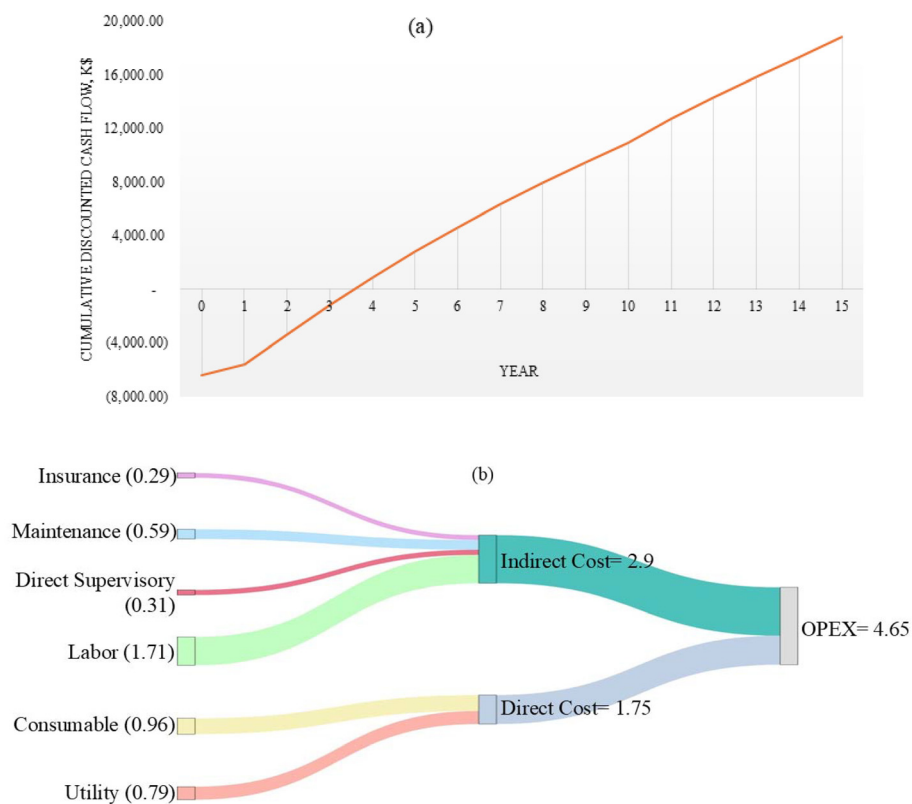


Fig. 4 The discounted cumulative cash flow (15-year) (a) and OPEX (b) values \$ per kg LCE for DLE spodumene-based LCE production method.



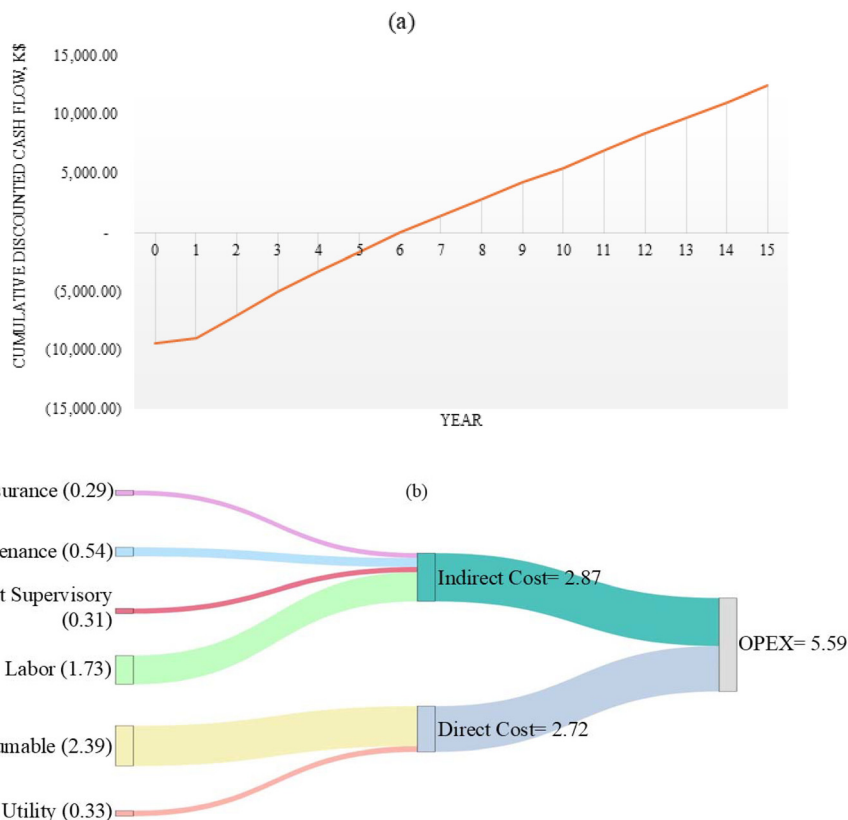


Fig. 5 The discounted cumulative cash flow (15-year) (a) and OPEX (b) values \$ per kg LCE for EDL spodumene-based LCE production method.

Table 3 Monte Carlo simulation results comparing the conventional, DLE, and EDL LCE production processes from hard rock at three scales (1, 5, and 10 tons per day)

Uncertainty indexes	Conventional			DLE			EDL		
	1 ton, LCE per day	5 tons, LCE per day	10 tons, LCE per day	1 ton, LCE per day	5 tons, LCE per day	10 tons, LCE per day	1 ton, LCE per day	5 tons, LCE per day	10 tons, LCE per day
Average profit, k\$ per year	−119.8	2268.0	7932.9	652.4	5793.8	13 152.7	415.6	5352.8	11 896.5
Standard deviation	931.0	4526.4	9238.0	835.8	4274.4	8521.7	813.9	3976.1	8035.9
Minimum profit, k\$ per year	−2248.5	−8455.0	−13 650.4	−1120.9	−2958.9	−4571.7	−1135.0	−2331.9	−3246.7
Maximum profit, k\$ per year	2050.0	13 088.1	29 108.6	2470.3	14 815.3	30 783.1	2050.0	13 102.1	27 858.4
Correlation (LCE price vs. profit)	0.85	0.86	0.85	0.95	0.96	0.96	0.98	0.99	0.99
Correlation (utility cost vs. profit)	−0.53	−0.52	−0.51	−0.34	−0.35	−0.28	−0.18	−0.17	−0.17

Risk metrics show equally important distinctions. Standard deviation values are largest for the conventional process (up to 9.24M\$ per year at 10 tons per day), narrower for DLE (8.52M\$ per year), and smallest for EDL (8.04M\$ per year). These results suggest that the conventional method carries the greatest variability in economic outcomes, while EDL is slightly higher than DLE in terms of predictability and consistency. The minimum profit values emphasize downside risk: at 10

tons per day, the worst-case outcome is −13.65M\$ per year for the conventional method, −4.57M\$ per year for DLE, and −3.25M\$ per year for EDL. This comparison indicates that both advanced methods substantially mitigate extreme losses, with EDL offering the most protection against downside risk.

For all three methods, LCE price is the dominant economic driver; however, its correlation with profit is weakest in the conventional method (0.85–0.86), stronger in DLE (0.95–0.96),



and nearly perfect in EDL (0.98–0.99). These results show that EDL and DLE not only capture higher average profits but are also more strongly leveraged to favorable market conditions. Utility costs, while negatively correlated with profit in all scenarios, show diminishing influence across the methods: conventional (−0.51 to −0.53), DLE (−0.28 to −0.35), and EDL (−0.17 to −0.18). The weaker correlations in the advanced methods imply reduced operational sensitivity to fluctuations in energy costs, with EDL showing the least vulnerability.

## 4 Conclusion

This comparative study evaluated the conventional, DLE, and EDL routes for LCE production from spodumene using an integrated TEA–LCA framework under uncertainty. Both advanced methods outperform the conventional acid-bake process environmentally and economically, though each presents distinct process bottlenecks and trade-offs.

DLE proved to be the most balanced and robust alternative. Under optimal conditions of 325 °C NaOH roasting followed by ambient-temperature water leaching, DLE reduces the global warming potential by approximately 49%, acidification by 53%, and fossil-fuel depletion by 45% compared to the conventional method. Economically, DLE achieved the shortest payback period (3.4 years) and highest return on investment (41%), driven by simplified process integration and reduced thermal demand. The OPEX analysis showed that DLE's operating costs are dominated by reagents (37%), while energy and labor requirements remain 20–30% lower than the conventional process.

EDL, optimized at 0.95 V under ambient conditions with a dilute acid electrolyte, showed even lower direct energy demand and reduced greenhouse-gas emissions (−70% compared to the conventional route). Its profitability (24% ROROI) and NPV (\$12.5M) remain lower than DLE due to higher capital intensity and electricity costs; however, its sensitivity and Monte Carlo analyses revealed the lowest downside risk and highest market-price leverage (profit correlation with LCE price  $\approx 0.99$ ), indicating strong resilience to market volatility.

The Monte Carlo simulation across three production scales (1, 5, and 10 t per day) confirmed that both DLE and EDL maintain positive profits at all scales, unlike the conventional process, which becomes unprofitable at low throughput. The standard deviation and minimum-profit metrics demonstrate that advanced processes reduce financial risk by 30–50% relative to the conventional route.

From a technical perspective, further progress in NaOH regeneration efficiency, reagent recycling, and corrosion resistance is required for continuous industrial operation of DLE. For EDL, electrode stability, module scalability, and energy efficiency remain critical research priorities.

This study demonstrates that integrating TEA and LCA with uncertainty analysis offers a comprehensive framework for evaluating emerging lithium extraction technologies. The findings highlight that as these technologies rapidly evolve to

address the challenges of a highly dynamic and sensitive industry, such as the lithium industry, it is crucial to assess their environmental impacts and economic viability simultaneously. For DLE, future improvements should focus on increasing caustic recovery efficiency, as well as developing corrosion-resistant materials and improving NaOH utilization to reduce reagent intensity. For EDL, progress will depend on developing durable, low-cost electrodes with higher faradaic efficiency and improved current distribution, together with advances in cell design and mass-transfer enhancement to support industrial-scale operation.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The data supporting the findings of this study have been included as part of the supplementary information (SI) provided with the article. All datasets analyzed or generated during the study can be found within the SI, ensuring full transparency and reproducibility of the research. No additional repositories or external databases were utilized for data storage in this study.

Supplementary information is available. Detailed life-cycle inventories for lithium extraction pathways from hard rock; techno-economic parameters and equipment design data; complete LCA characterization results and identified environmental hotspots; CDF uncertainty analysis and full TEA results. See DOI: <https://doi.org/10.1039/d5gc04866d>.

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