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Comparative Life Cycle Assessment of Lithium Mining, Extraction, and Refining Technologies: a Global Perspective

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Abstract

The clean energy transition requires a considerable amount of different minerals, and lithium is one of the most critical elements owing to its use in Lithium-ion batteries for various applications. This led to calling this element "White Oil", predominately extracted from brines and hard rocks. Alternative resource types, such as hectorite and zinnwaldite, become attractive and potentially feasible due to the surging market demand and price. Importantly, each resource type shows unique mineral characteristics and requires different process technologies, resources and energy for lithium extraction and production, resulting in distinctive environmental impacts. Therefore, this paper presents a comparative life cycle assessment (LCA) to quantify the environmental impact of selected lithium production routes: brine (Chile), spodumene (Australia & China), hectorite (Mexico), and zinnwaldite (Germany). The cradle-to-gate LCA models suggest that brine resources have the lowest impact regarding global warming potentials. In contrast, spodumene-based and emerging routes (i.e., hectorite and zinnwaldite) appear more carbon-intensive than brine routes, but it is worth noting the uncertainties of this conclusion due to a lack of high-quality data in the public domain.

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1. Introduction

The clean energy transition is regarded as the key strategy to tackle the climate change crisis, which has become more urgent with a clear sign of breaking climate records, such as global mean surface temperature and sea level rise [1]. To end our reliance on fossil fuel-based energy, the main cause of climate change, renewable energy generators and battery storage technologies will play an essential role in all human activities, which demand numerous non-renewable and rare resources. Lithium is one of the most critical elements owing to its use in Lithium-ion batteries (LIB) for traction batteries in vehicles, stationary storage in electricity grids, and consumer electronics. In 2017, batteries comprised 46 percent of total lithium by enduse and were expected to account for 95 percent by 2030 [2, 3]. Although LIBs are present in different forms of design, formula and composition, the demand for lithium will rise from around 500,000 metric tons of lithium carbonate equivalent (LCE) in 2021 to three to four million metric tons in 2030 [3]. Meanwhile, we have witnessed a soaring lithium price in 2022, \$75,000 per metric ton of lithium carbonate, compared to a fiveyear average of \$14,500 [3]. This led to calling lithium "White

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Oil" which is predominately and currently extracted from bines and hard rocks in Australia, Latin America, and China. Because of its techno-economic significance, lithium industry has also seen a pipeline of new projects in Mexico, United States, Germany, and other countries, extracting lithium from alternative hard-rock assets (e.g. Hectorite and Zinnwaldite) and unconventional brines (e.g., geothermal and oilfield brines) [3].

Although lithium and battery technology are deployed to improve environmental sustainability, there are serious environmental concerns about their production from mining, extraction and purification due to the intensive use of fuels, electricity, and hazardous chemicals. Several papers and reports have created life cycle assessment (LCA) models to assess and compare the environmental impacts of different lithium production routes. Jiang et al. highlighted the significant difference between brine-based and rock-based routes in terms of global warming potentials (GWP), 0.329 and 15.69 kg CO₂eq/kg $Li₂CO₃$, respectively [4]. Although primary data was reported for the lithium refinery process in China, the authors called for further research to improve the inventory data of mining activities and Spodumene production. Ambrose and Kendall projected the environmental impact of lithium production considering the change in resource types and ore qualities in the future [5]. The overall average impact of lithium may change little for the projected period, but site-by-site variability is significant and requires site-specific assessment. More recent studies, including Kelly et al., Schenker et al., and Chordia et al., further proved that the brine-based route is much less carbon intensive than Spodumene-based, but the value and the magnitude of the carbon intensity differ from study to study, especially for Spodumene (all based on production in Greenbushes, Australia) [6-8]. The discrepancy is mainly due to the data sources and the system boundary. However, different processing technology for Spodumene concentrate (e.g., Mt. Cattlin, AU) has yet to be included. Moreover, there is a lack of investigation into the environmental impact of producing lithium from emerging alternative hard-rock assets: Hectorite and Zinnwaldite.

To fill the gap, this paper presents new cradle-to-gate LCA models for producing lithium via Spodumene (Mt. Cattlin,

Australia & China)), Hectorite (Mexico), and Zinnwaldite (Germany). Furthermore, the LCA results are compared with established studies on lithium production via Brine (Chile) and Spodumene (Greenbushes, Australia & China).

The following of this paper is organised as: Section 2 provides an overview of the global lithium extraction and purification industry; Section 3 presents the method and results of this comparative LCA study, followed by a brief discussion on the uncertainties and limitations; Section 4 concludes this work and recommends for future efforts.

2. Global Perspective of Lithium Extraction & Production

Lithium can be extracted in different forms from a range of resources which is usually categorised into two main groups: brines and hard rocks [9, 10]. The extraction processes and technology differ significantly among different categories, subcategories, and even site-to-site, due to dynamic parameters of geological, time, and techno-economic factors. Based on the United States Geological Survey (USGS) report, around 58 percent of the lithium resources come from closed-basin brines, 26 percent from pegmatites, 7 percent from lithium clays, and 9 percent equally from the oil-field, geothermal brines, and lithium zeolites [11-13]. Figure 1 depicts the global explored lithium resources (48 countries) and production in 2021, according to USGS and British Geological Survey [13, 14].

Closed-basin brines, known as brines, are deposits containing considerable amounts of saline groundwater enriched in lithium and other elements like boron, potassium, and magnesium at an economic level. The ratio of elements is critically important for selecting the process method; however, the current producing brine projects, which have a 160 to 1400 ppm lithium content, all use pre-concentration ponds to reduce impurities like sodium chloride, followed with magnesium precipitation by adding lime to the solar-evaporation ponds after pumping out the brine from the playa. From this stage, some projects continue with solar ponds to create a more concentrated brine (around 6.7% Li, as LiCl, like Salar-de Atacama, Chile) before the carbonation process. Furthermore, in some cases regarding the high magnesium-to-lithium ratio

Figure 1: World map of lithium resources with productions in 2021 [13, 14]

and the amounts of potassium and boron, the pre-concentrated brine is sent to an ion-exchange plant to increase the lithium recovery rate to produce different products, such as boric acid, and potassium sulfate besides lithium carbonate [13, 15].

Granitic pegmatites, which are the dominant resource of lithium production nowadays, consist of different minerals for critical elements like lithium, beryllium, tantalum, niobium, caesium, rubidium, and tin. The production process starts with different mining methods regarding the deposit characteristics: after blasting, and hauling the run of mine (ROM), to the processing plant, concerning the geo-metallurgical properties of the ore, a physical (Mt. Cattlin, AU) or flotation (Greenbushes, AU) process is deployed to produce α -Spodumene concentrate, which usually has 5.7 to 6.5 percent of Li2O content. This concentrate undergoes a phase conversion process to deform the α-Spodumene concentrate to β-Spodumene. After this operation, a series of roasting, leaching, precipitation, filtration, and purification steps are utilised to produce lithium carbonate battery grade [16-20].

Lithium-clay deposits are not producing lithium-compound products currently; however, there are several projects in the USA, Mexico, Peru, and Serbia [21]. The American and Mexican lithium-clay (Hectorite) projects will start to produce lithium carbonate in 2022, based on their project reports [22, 23]. Due to the soft characteristic of clays, blasting is unnecessary during Hectorite mining; so, exploitation will be accomplished by stripping the overburden, and then by using a mixture of back-hoe and truck. The run of mine will be transported to the processing plant, whereafter drying the clay ore and crushing it with Raymond mills by using a series of scrubbers and classifiers. So, a feed is prepared for the flotation plant to produce a 0.65 percent lithium concentrate out of a 0.35 percent lithium-clay ore. This is achieved by removing coarse silica particles and calcite during scrubbing and flotation respectively. Afterwards, this concentrate will be roasted in a form of pellets by adding gypsum and lime. After salt roasting, the pellets will undergo the leaching steps and subsequently purification and precipitation stages by utilising the ionexchange method. This operation needs to be repeated for reaching the battery-grade lithium carbonate [22].

Another type of hard rock deposit that will produce lithium products is lithium-bearing micas named Zinnwaldite. There are two projects in the Czech Republic (Cinovec) and Germany (Zinnwald) for this resource type. In Germany, it is planned to produce Lithium fluoride (LiF), which is used for lithium-ion battery electrolytes, in 2022. The mined ore has around 0.3 percent lithium content. After drying and comminution operations, a Zinnwaldite concentrate with approximately 1.3 percent lithium content will be produced by using a series of high-intensity magnetic separators. This concentrate will then be re-grinded and mixed with lime and calcium anhydrite to form Zinnwaldite pellets. These pellets, afterwards, will be sent to the conversion and purification plant, which first will undergo a roasting operation followed by cooling and leaching steps. After transferring lithium ions into the hydrogen fluoride solution by adding chemicals like potassium carbonate and potassium hydroxide, the pregnant leaching solution (PLS) will be purified from magnesium and calcium impurities. Then, it will be sent to the precipitation step by adding potassium

fluoride solution. With these operations, 5112 tons of lithium fluoride with more than 99.4% purity will be produced annually in the Zinnwald plant in Germany. This amount is equivalent to 7280 tons of lithium carbonate (LCE), based on the Zinnwald technical report [24].

Extraction from geothermal and oilfield brines attempts to deploy direct lithium extraction (DLE) method, which may deploy sorption, ion-exchange (IX), solvent extraction (SX), membrane technologies, or precipitation methods [2]. Companies, such as Dow and Albemarle are focusing on oilfield brines, whereas Eramet, Livent, Vulcan Energies and others are developing process based on geothermal brines. However, all these technologies are in lab or pilot scale, thus they are excluded from this comparative LCA at this stage.

3. Comparative Life Cycle Assessment

This comparative LCA study follows the ISO14040 standard, which consists of four phases, namely: goal and scope definition, Life Cycle Inventory (LCI) analysis, Life Cycle Impact Assessment (LCIA), and interpretation. The following subsections are organized accordingly. We used OpenLCA in conjunction with EcoInvent Database 3.7 for this study.

3.1. Goal and Scope Definition

As mentioned earlier, the main goal of this LCA is to compare the environmental impacts of current and emerging lithium extraction routes. Accordingly, the functional unit is producing one metric ton of battery-grade Lithium Carbonate Equivalent (LCE). The scope of this study is cradle-to-gate. Specifically, the LCA models cover life cycle stages from mining, comminution, processing, refining and transport, as shown in Figure 2. Co-products were excluded from this study, so no allocation method was applied.

Five lithium production routes were modelled:

- 1. Spodumene Based: Mt Cattlin, Australia and Zhangjiagang, China;
- 2. Spodumene Based: Greenbushes, Australia and Zhangjiagang, China;
- 3. Brine Based: Salar-de Atacama, Chile;
- 4. Hectorite Based: Sonora, Mexico;
- 5. Zinnwaldite Based: Zinnwald, Germany.

3.2. Life cycle inventory analysis

The life cycle inventory analysis was completed by using mainly existing literature and publicly available technical reports, supplemented by EcoInvent. In this phase, we focused on the identification of input and output flows of each process steps and their quantity as shown in Figure 2. In general, the input flows include different forms of energy (e.g. electricity, diesel, steam), materials, chemical reagents, explosives, and water, whereas the outflows mainly consist of products and wastes. The transportation between site was also accounted. Owing to the availability of high-quality data, the life cycle inventory of brine-based route (Salar De Atacama) and lithium carbonate production in China was recalculated based on [4, 8, 25]. More efforts were invested into developing the inventory

Figure 2: Lithium extraction and refinery processes with inputs and outputs

of Mt Cattlin, Greenbushes, Sonora, and Zinnwald projects based on technical reports and patents [19, 20, 22-24, 26-31]. Specifically, all the inputs and outputs, along with the mass and energy balance and source of energy, are extracted from their NI-43 101 technical reports. Due the page limit, the detailed life cycle inventory of those processes will be published in future articles.

3.3. LCIA from the ReCiPe method

The LCIA has been conducted using the ReCiPe midpoint (H) method for the production of LCE through the selected routes [32]. Figure 3 shows the absolute results of Global Warming Potential (GWP) and water consumption for the different routes. The production routes based on Spodumene from Greenbushes and Mt Cattlin are further distinguished between the share of Spodumene concentration incl. ore extraction at the respective Australian site and refinement of LCE in China.

For Greenbushes, diesel burned for electricity consumption in the processing plant accounts for the largest share of GWP (88.94%) for the α -Spodumene concentrate production. This is mainly due to the energy intensive processes of crushing, grinding, desliming, and flotation, and diesel is used as the main energy source for the remote site. In comparison, only 8.66% of GWP results from mining operations for the supply the designated amount of ore. The rest of the GWP contribution is due to used materials in the processing (e.g. sodium hydroxide and sodium silicate). Overall, the ore extraction and Spodumene concentration leads to a share of 76.53% of total LCE production from this source. Further major contributions to GWP in LCE production in China result from use of electricity (11.32%), soda ash (7.95%), quicklime (2.04%), sodium hydroxide (1.96%) and sulfuric acid (1.50%). It is worth mentioning that the shipping between Australia and China is just 0.11% of the total GWP.

For concentrated α-Spodumene from Mt Catlin process plant, major contribution to GWP results from the exploitation of ore (incl. drilling, blasting, hauling, etc.) accounting for 69.09%. Other main contributions to GWP are due to use of electricity (22.82%) and ferrosilicon production (7.76%). In contrast to the site in Greenbushes, only \sim 30% of the GWP result from Spodumene concentration process due to a different process design and use of electricity instead of diesel in this LCA model. For the entire production of one ton of LCE, only 37.55% result from ore extraction and Spodumene concentration processes at the Mt Cattlin site. The use of

Figure 3 Comparison of GWP and Water Consumption of selected lithium carbonate routes

electricity in China for the further refinement result in 30.15% and the use of soda ash result in 20.9% of total GWP.

The GWP for the production route from Mexican lithiumbearing clays results mainly from the use of electricity (30.02%), natural gas (27.22%) and soda ash (13.69%) for processing of clays to LCE, as well as Hectorite mining (13.51%) and residues from pyro- and hydro-metallurgical processes (11.62%). For the production of one ton LCE from the Zinnwald project in Germany, the total GWP mainly results from the use of quicklime (38.65%), followed by the run of mine (27.3%), the potassium hydroxide (17.6%), and residues from processing and metallurgical processes (13.65%).

LCE production from brine in Chile shows the lowest GWP. Compared to heavy machine-driven mining operations and energy-intensive concentration processes, brine is only pumped to the ground and progressively concentrated via solar evaporation. The highest impact for GWP for this process route results from use of soda ash (66.71%) and electricity (8.54%).

For water consumption, the results of the assessment show similar trends as for GWP. However, the leaching processes for LCE production from Spodumene in China and the use of soda ash, sulfuric acids and sodium hydroxide have highest influence. The water consumption of brine-based LCE production is dominated by the use of soda ash (89.93%) since brine itself is not regarded as water and effects on the groundwater level are not taken into account. For LCE production from Zinnwaldite, water consumption is dominated by the use of potassium hydroxide (28.49%) and the run of mine (25.13%), whereas Mexican clay mining and processing is mostly affected by direct water consumption (49.63%) and the use of soda ash (31.11%).

3.4. Discussion

The LCA models for this study are built on publicly available data, which show limitations compared to first-hand data from industry. One example for these limitations is the 'market for Spodumene' data from EcoInvent, which is approximated based on iron ore mining and limestone crushing. Furthermore, process routes based on Zinnwaldite and Hectorite can only be partly covered, due to missing data. For the example of Zinnwaldite, the run of mine is approximated from EcoInvent data for hard coal mining. However, the use of technical reports and patents allow for appropriate first results.

For the case of Mt Cattlin and brine, the authors have no knowledge about the actual source of electricity. In the model, the respective local electricity mix is used. However, if electricity is generated from diesel generators, similar to the site in Greenbushes, the GWP would result in higher values as well.

The allocation of impacts to co-products is another issue not covered yet, neither in this paper, nor in previous studies. However, it is worth considering that especially the mineral mining results in the production of other co-products, such as tantalum, niobium, tin, beryllium, caesium, rubidium, feldspar and quartz.

Another aspect that is not covered in the environmental assessment is the duration of processes. Although, LCE production from brine appears to result in lowest GWP and water consumption, it is also the process route taking longest time. The extraction of brine concentrate can take up to two years, whereas the production of Spodumene concentrate is done in less than a week. It is therefore worth to conclude that the environmental impacts originating from the hard-rock lithium categories are the price for the increasing demand, speed of production, and delivery to the market.

Compared to previous literature, e.g. [8], the paper shows higher results for the GWP from the production of LCE from Spodumene sourced in Greenbushes. This is mainly due to the inclusion of flotation processes in the model. The results for brine-based process routes are similar to previous studies. The novelty is especially given by the assessment of LCE production routes based on Spodumene mined from Mt Cattlin and future sources of Zinnwaldite from Germany and Hectorite from Mexico. However, due to the discussed limitations of this work, further work needs to be done to gain more accurate results based on first-hand industry data or appropriate process models.

4. Conclusions & Outlook

The paper provides a global perspective of lithium sources, current and emerging production routes of battery-grade Lithium Carbonate Equivalents. The current trend of increasing lithium demand for battery driven vehicles leads to a further diversification of process routes. For this reason, the paper provides a comparative LCA for the production of LCE from various sources.

The results of this study show great variances in the GWP and water consumption of LCE production depending on the source and production route. It can be concluded that under the given approximations, LCE from brine has lowest environmental impact. In contrast, production of LCE in China from Spodumene sourced in Greenbushes, Australia, causes highest GWP and water consumption. However, due to the large influence of energy-intensive process steps in the extraction and concentration of minerals, great potential can be seen in the replacement of fossil energy sources, such as diesel, with renewable energies.

Apart from Spodumene and brine, the two emerging LCE production routes from Hectorite and Zinnwaldite can be regarded as promising for addressing the increasing demand. The results of this study show that the environmental impact of these production routes is not expected to show strong variation from conventional sources. The greatest potential for the reduction of environmental impacts can be seen in a more efficient use of electrical energy and operating materials such as lime.

In order to improve the results of this work, the authors will develop advanced process models for the introduced production routes. The gathering of first-hand data from mining and metallurgical industries is also aimed to validate and improve the results. Once achieved, more comprehensive impact assessments, sensitivity and uncertainty analysis will be conducted and reported.

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