Comparative life cycle assessment of synthesis routes for cathode materials in sodium-ion batteries

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Abstract
Sodium-ion batteries (SIBs) are lower cost and more sustainable alternatives for lithium-ion batteries. However, despite the high research attention to the development of the synthesis procedures of the electrode materials for SIBs, there has been less focus on the environmental burdens of each production route which is a vital aspect for large-scale industrial applications. A comparative life cycle assessment (LCA) with a cradle-to-gate approach was performed here to evaluate the environmental impacts of the production phase of a promising cathode material with the chemical formula of Na3MnCO3PO4 (NMCP), which was previously studied in SIBs. LCA was used to compare the environmental impacts of three strategies for the production of NMCP nanomaterials, including ball milling, hydrothermal, and stirring-assisted hydrothermal. Results demonstrated that in hydrothermal-based methods, sodium carbonate showed a considerably high impact in almost all categories owing to its high consumption in these processes. In ball milling and stirring-assisted hydrothermal methods, electricity is one of the main environmental weaknesses. By scaling the results for an equivalent functionality and considering 1 kWh of energy storage capacity as the functional unit, ball milling showed the least environmental impact in all seven categories except acidification, eutrophication, and carcinogenic. Furthermore, Global warming impact as the most investigated category in the field of batteries was in the range of 14–20 kg CO2-eq. per kg of the synthesized NMCP nanomaterials prepared via the three studied methods which suggest the appropriate design of the applied procedures.

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Introduction

Rechargeable batteries are the main bridges for transition from the existing fossil-fuel-based systems toward renewable, sustainable, and clean energy ones. The development of rechargeable batteries is significantly attributed to the improvement in the electrochemical performance of electrode materials (Lopez et al. 2021). However, the electrochemical capabilities of electrode materials cannot be the only deciding factor for their application in energy storage devices and their application should be reconsidered from the viewpoint of their environmental impacts. Among counterpart electrode materials with comparable capabilities, the one with the lowest environmental impact will win for large-scale industrial applications (Sharma et al. 2019).

The evaluation of environmental impacts can be done using life cycle assessment (LCA) studies. LCA is a comprehensive tool to quantify the environmental impacts of a process or a product during its whole life cycle or a portion of its life cycle, and therefore, helping researchers, industries, societies, and policymakers to develop environmentally friendly and sustainable products and processes (Wu et al. 2019).

Sodium-ion batteries (SIBs) are emerging potentially as a less expensive, more sustainable, and environmentally friendly alternative to lithium-ion batteries (LIBs), and they are the only commercialized post-LIBs technology. While the environmental impacts of LIBs are intensively studied, only a few LCA investigations have been done on SIBs and the knowledge base in this field is quite weak (Peters et al. 2016). Most of the studies on SIBs only focus on electrochemical performance and do not assess the environmental burdens associated with the production of SIBs. For closing this gap, Peters et al. (2016) reported the first LCA study on the production of SIBs by using layered transition metal oxide and hard carbon as electrode materials. They found that the environmental impacts of SIBs are competitive with LIBs when delivering a similar lifetime. Moreover, Trotta...
et al. (2022) found that SIBs with bio-derived hard carbon anodes represent the lowest environmental impact in all categories in comparison with their lithium counterparts, plus being 18% cheaper. However, in another LCA study, it was revealed that LIBs show the lowest environmental impact, even compared to the hypothesis of industrial-grade MXenes-based SIBs (Carvalho et al. 2022).

The production process of LIBs and their sodium counterparts is generally divided into three phases of electrode manufacturing, cell assembly, and cell finishing, and accordingly, a wide variety of system boundaries could be selected. Conditionally, the production of cathode materials at battery manufacturing facilities has been suggested to be the largest or second-largest energy consumer, and to develop SIBs with eco-friendly technology, investigating the environmental impact of the manufacturing process of electrode materials, especially cathodes, is crucial (Porzio and Scown 2021). Malara et al. (2021) reported a comparative cradle-to-gate LCA study for evaluating the production process of electrospun Fe2O3-based fibers as SIBs anodes. In a very recent publication, the environmental impact assessment of ten representative Na3V2(PO4)3 cathode materials at a laboratory scale was performed using a cradle-to-gate approach (Rey et al. 2022).

While new promising chemistries such as poly-anionic or Prussian-blue based cathode materials have been developed for SIBs, no environmental assessment is yet available on these battery materials (Peters et al. 2021), except those aforementioned. Na3MnCO3PO4 (NMCP) is a low-cost polyanionic electrode material with high stability, safety, and suitable operating voltage for use as the cathode in SIBs. Additionally, NMCP is one of the few manganese compounds in which both Mn4+/Mn3+ and Mn3+/Mn4+ redox pairs could be activated during the charge/discharge processes, leading to its high theoretical capacity (191 mAhg⁻¹), which is beneficial in making high-energy density batteries (Hassanzadeh et al. 2016a).

In the previous literature reported by one of the authors (Hassanzadeh et al. 2016a, b, 2018; Hassanzadeh and Sadrnezhaad 2021), three facile synthesis procedures including ball milling, hydrothermal and stirring-assisted hydrothermal processes were adopted to prepare NMCP nanoparticles, and then the structural, morphological, and electrochemical properties of the obtained materials were investigated. However, these synthesis methods were not designed according to the strategies of green chemistry and they may involve environmentally harmful chemicals and consume large amounts of energy. Until now, no works have been devoted to specifically examining the environmental burdens of the NMCP cathodes’ fabrication process. Therefore, in the current research, LCA was utilized to evaluate the environmental impacts of the three previously developed synthesis routes of NMCP nanoparticles to inform a cleaner production route.

The current research was aimed to expand the scientific knowledge of the environmental impacts of SIBs, and support battery manufacturers for the future development of eco-friendly cathodes applied in post-LIBs technologies. In this context, this work presents the results of a comparative laboratory-scale LCA on three synthesis methods of NMCP cathode materials, and it is a completion of the previous literature focused solely on the (physicochemical properties and) electrochemical performance of NMCP cathodes.

**LCA methodology**

According to the ISO 14040, the standard LCA approach includes four different steps that are: goal and scope definition, life cycle inventory assessment, life cycle impact analysis, and interpretation.

**Goal and scope**

The aims of this work were to (1) quantify and compare the environmental impacts of the three main routes of NMCP production, including ball milling, hydrothermal and stirring-assisted hydrothermal processes, (2) identify the key processes or inputs/outputs in each fabrication method that had the most significant environmental impact, and (3) identify the most appropriate process for fabricating NMCP cathode from both electrochemical performance and environmental impact perspectives.

As reported by Porzio and Scown (2021), in the field of LCA of batteries, until now, there has not been consensus on how to analyze the environmental impacts, nor how to report the results. Studies use a wide variety of system boundaries, functional units (FUs), primary data sources, and life-cycle inventory, midpoint, and impact categories. Recently, Arshad et al. (2022) reviewed the LCA studies on advanced battery systems by a literature search in the years 2010–2021. As could be seen in their review paper (Table 1), various functional units have been used in different studies. In the current research, two FUs have been used as follows: (1) 1 kg of the synthesized cathode material, and (2) 1 kWh of energy storage capacity. Using the mass of cathode material as a FU is suitable for comparing the environmental impacts of different production routes, even across multiple studies. Since the battery field is an electrochemical performance-driven area, using the energy storage capacity as a FU is beneficial for scaling the results for equivalent functionality. The battery energy storage capacity (Wh) is a performance parameter being the product of the battery voltage (V) and its specific capacity (Ah) (Aghamohammadi et al. 2022). Note that these two FUs were also used in the precedent done by Rey et al. (2022).
The LCA system boundaries define the processes, materials, energy flows, and activities that are involved in the analysis (Papadaki et al. 2017). In the current study, a cradle-to-gate LCA, i.e., from starting materials to the fabrication of NMCP nanoparticles, was investigated, while the use and disposal of the NMCP nanoparticles were not included in the system boundary. The process diagram, system boundary, and the inputs/outputs of each process are presented in the following sections.

**Life cycle inventory**

In this work, a comparative life cycle analysis among the different synthesis methods for the fabrication of NMCP nanoparticles was done using the SimaPro 8.3 software, and the Ecoinvent v3.0 database. Evaluations were performed based on real data obtained from laboratory-scale experiments developed previously by the authors.

The main equipment used in the synthesis processes, including autoclave, planetary ball mill, electric oven, stirrer, etc., as materials, are external to system boundaries, while the used electricity is included inside the boundaries. It has to be noted that except for manganese nitrate, all chemical inputs/outputs used in the synthesis process, are included in life cycle inventory (LCI) databases. Since there are no relevant inventories available in the Ecoinvent dataset for manganese nitrate, its inventory was compiled in the current study, according to the calculations based on the stoichiometric reaction of its production process derived from literature. The consumption of raw materials and the amount of by-products for the production of 1 kg of manganese nitrate are presented in Table S1 (see supporting information) which are calculated based on reaction S1.

### Ball milling synthesis of NMCP

NMCP was prepared by the same ball milling process described previously by Hassanzadeh et al. (2016b). For this purpose, 8 mmol Mn(NO₃)₂·4H₂O (Sigma), 8 mmol Na₂HPO₄·2H₂O (Sigma), and 12 mmol Na₂CO₃·H₂O (RDH) powders were transferred into a container with a ball-to-powder ratio of 30:1. The synthesis was performed in a planetary ball mill at 300 rpm for 1 h as the optimum milling time (Hassanzadeh et al. 2016b). Since NMCP is not soluble in water, the products were dissolved in DI water under magnetic stirring to separate NMCP from soluble by-products. Finally, the dissolution residue was washed with DI water and oven-dried at 50 °C. The process diagram and the inventories for the ball milling synthesis of NMCP when considering 1 kg of synthesized NMCP and 1 kWh of energy storage capacity as the FUs are presented in Fig. 1, Table 1, and Table S2, respectively.

In order to calculate the output and emissions of the process, the occurring reaction between the raw materials should be considered. The possible reaction during the synthesis process could be considered as follows (Hassanzadeh et al. 2016b):

\[
2\text{Mn(NO}_3\text{)}_2\cdot\text{4H}_2\text{O} + 2\text{Na}_2\text{HPO}_4\cdot\text{2H}_2\text{O} + 3\text{Na}_2\text{CO}_3\cdot\text{H}_2\text{O} \\
\rightarrow 2\text{Na}_3\text{MnCO}_3\text{PO}_4 + 4\text{NaNO}_3 + \text{CO}_2 + 16\text{H}_2\text{O} \quad (1)
\]

Stoichiometrically production of 1 kg NMCP according to reaction 1 requires 0.90 kg Mn(NO₃)₂·4H₂O, 0.64 kg Na₂HPO₄·2H₂O, and 0.67 kg Na₂CO₃·H₂O. According to empirical data, as presented in Table 1, the reaction efficiency is about 50%. Therefore, the amounts of reaction...
byproducts (emissions to air and water) were calculated according to the efficiency of 50%, as reported in Table 1.

**Hydrothermal synthesis of NMCP**

Hydrothermal synthesis of NMCP was done according to the procedure reported earlier (Hassanzadeh et al. 2016a, 2018). A clear solution (A) was prepared by dissolving 2 mmol of Mn(NO₃)₂·4H₂O (Sigma) in 5 ml of deionized water. Concurrently, 2 mmol of Na₃HPO₄·2H₂O (Sigma) and 2.34 g of Na₂CO₃·H₂O (RDH) were dissolved in 10 ml of deionized water to form a clear solution (B). Note that the synthesis conditions such as the ratio and concentration of starting materials, the temperature, and duration of the hydrothermal process were optimized in previous literature (Chen et al. 2012a, b; Hassanzadeh et al. 2016a, 2018). After adding solution (A) to the solution (B) under fast magnetic stirring, the solution was transferred into a 25 ml Teflon-lined stainless steel autoclave and heated hydrothermally at 120 °C for 24 h. After cooling to room temperature, the products were washed with deionized water and dried in the oven at 50 °C overnight. Figure 2, Table 2, and Table S3 depict the production scheme, and the process inventories when considering 1 kg of synthesized NMCP and 1 kWh of energy storage capacity as the FUs, respectively.

**Magnetic stirring-assisted hydrothermal synthesis of NMCP**

Magnetic stirring-assisted hydrothermal synthesis of NMCP was performed according to recently published literature (Hassanzadeh and Sadrnezhad 2021). NMCP was fabricated by the similar hydrothermal process described in “Hydrothermal synthesis of NMCP” section. For this purpose, 8 mmol of Mn(NO₃)₂·4H₂O (Sigma) was dissolved in 20 ml of deionized water to form solution (A), while solution (B) was prepared by dissolving 8 mmol of Na₃HPO₄·2H₂O (Sigma) and 9.36 g of Na₂CO₃·H₂O (RDH) in 40 ml of deionized water. Solution (A) was mixed with solution (B) under fast magnetic stirring and then transferred into a 100 ml glass bottle sealed with a cap. The bottle was heated for 480 min in a 120 °C oil bath under magnetic stirring. After cooling the sample to room temperature, the product was washed with deionized water and dried in an oven at 50 °C overnight. Similar to the previous methods, the process diagram and the inventories when considering 1 kg of synthesized NMCP and 1 kWh of energy storage capacity as FUs are presented in Fig. 3, Table 3, and Table S4, respectively.

**Impact assessment**

To fully facilitate the use of results, TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts) method developed by the United States Environmental Protection Agency (EPA) was employed as a midpoint-oriented technique to assess the environmental burdens. The impact categories include ozone depletion (kg CFC-11 eq.), global warming (kg CO₂ eq.), smog (kg O₃ eq.), acidification (kg SO₂ eq.), eutrophication (kg N eq.), carcinogenics (CTUh), non carcinogenics (CTUh), respiratory effects (kg PM2.5 eq.), ecotoxicity (CTUe), and fossil fuel depletion (MJ surplus).

**Results and discussion**

Phase analyses of the obtained products through the three synthesis methods were carried out via the X-ray diffraction (XRD) technique and the results confirmed the formation of the pure NMCP phase, as reported in the previously published literature by the author (Hassanzadeh et al. 2016a, b,
2018; Hassanzadeh and Sadrnezhaad 2021), in which the fabrication methods were the same as those adopted in the current study. The morphological investigations revealed that the ball-milled sample was constituted of micron-sized particles formed from a myriad of needle-like primary nanoparticles with diameters of $\sim 15 \text{ nm}$, as seen in the field emission scanning electron microscope (FESEM) image in Fig. 4a (Hassanzadeh et al. 2016b). Producing extremely fine NMCP nanoparticles via ball milling would result in the enhancement of electronic and ionic conductivities of the prepared material and consequently higher discharge capacities could be achieved. In contrast, the NMCP sample prepared by the hydrothermal method showed a wide range of particle sizes (25 nm to $\sim 4 \mu m$), as observed in Fig. 4b and 4d, and the average particle size was larger than that of the other two samples (Fig. 4a, c). Noted to mention that FESEM and TEM images are used in this figure according to their availability in literature.

Moreover, the performance of SIBs constructed from the prepared NMCP cathodes was reported earlier (Hassanzadeh et al. 2016a, b, 2018; Hassanzadeh and Sadrnezhaad 2021), and is summarized in Table 4. As presented in this table, the NMCP material prepared via ball milling delivered the highest discharge capacity, among the three samples. The NMCP cathodes fabricated by hydrothermal and stirring-assisted hydrothermal methods delivered discharge capacities of 76 and 94 mAh/g, respectively. The difference in the electrochemical performance of the samples could mainly be attributed to the various morphologies of the obtained materials, as compared in Fig. 4. In conclusion, the findings revealed that the stirring-assisted hydrothermal method resulted in the occurrence of mild conditions in both morphological and specific capacity features, as seen in Fig. 4c and Table 4 (Hassanzadeh et al. 2016a, b, 2018; Hassanzadeh and Sadrnezhaad 2021).

In the current study, the comparative LCA study was performed to evaluate the environmental impacts of the production phase of NMCP nanoparticles. Table 5 shows the comparative life cycle impacts in ten environmental categories for the production of 1 kg of NMCP through three different methods of ball milling (B), hydrothermal (H), and stirring-assisted hydrothermal (S). Comparing the data in columns B, H, and S of the table reveals that there is no obvious winner between the three different synthesis methods in terms of environmental impacts.
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Table 3  Life cycle inventory for fabricating NMCP material via stirring-assisted hydrothermal process when considering 1 kg of synthesized NMCP as FU

| Input/output | Consumption | Unit | Unit process |
|--------------|-------------|------|--------------|
| **Inputs**   |             |      |              |
| Mn(NO₃)₂·4H₂O | 0.94 kg     |      | Compiled inventory for manganese nitrate |
| Na₂HPO₄·2H₂O | 0.67 kg     |      | Sodium phosphate [RoW] market for sodium phosphate | APOS, U |
| Na₂CO₃·H₂O   | 4.40 kg     |      | Sodium percarbonate, powder [RoW] market for sodium percarbonate, powder | APOS, U |
| Deionized water | 70.42 L     |      | Water, deionized [RoW] market for water, deionized | APOS, U |
| Electricity  | 44.42 kWh   |      | Electricity, medium voltage [IR] market for | APOS, U |
| **Emission to air** | | | |
| CO₂ (g)      | 0.08 kg     |      | Carbon dioxide |
| **Emissions to water** | | | |
| HPO₄²⁻       | 0.02 kg     |      | Waste water, to water |
| CO₃²⁻        | 0.01 kg     |      | Waste water, to water |
| NO₃⁻         | 0.01 kg     |      | Waste water, to water |
| Na⁺          | 0.04 kg     |      | Waste water, to water |
| Mn²⁺         | 0.01 kg     |      | Waste water, to water |

Fig. 4 FESEM images of a NMCP fabricated by ball milling process (Hassanzadeh et al. 2016b), b NMCP prepared by hydrothermal procedure (Hassanzadeh et al. 2016a), and TEM images of c NMCP synthesized by stirring-assisted hydrothermal method (Hassanzadeh and Sadrnezhaad 2021), d NMCP prepared by hydrothermal method (Hassanzadeh et al. 2018)

Figure 5a illustrates the findings of Table 5. In this figure, the maximum value of three synthesis methods in each of the ten environmental categories has been set as the base for comparison. Therefore, the environmental impacts of the three synthesis routes can be compared in each category. The hydrothermal method has the highest impact in non carcinogenic and respiratory effects categories, while the stirring-assisted hydrothermal method had the most impact in the categories of ozone depletion, global warming, smog, and fossil fuel depletion. The ball milling method showed significant impacts in three categories of acidification, eutrophication, and carcinogenics, while its impact in the other categories was less than or comparable to the hydrothermal-based methods.

When investigating the environmental impacts associated with batteries, by far, the most investigated impact category is global warming as long as transport sector decarbonization is the key factor of electric vehicle market importance (Arshad et al. 2022). The global warming impact category quantifies the amount of greenhouse gases produced in different procedures and accordingly, companies can develop strategies to lower those (Trotta et al. 2022).

Iranian electricity production is still largely dependent on natural gas and a huge amount of CO₂ is released annually from burning fossil fuels. As seen in Fig. 5a, the stirring-assisted hydrothermal method contributes considerably to the global warming impact category, which could be contributed to the high amount of electricity consumption in this method (44.42 kWh according to Table 3). Ball milling and hydrothermal techniques are in the next place in terms of impact on the global warming category, with electricity consumptions of 36.96 kWh and 0.40 kWh according to Tables 1 and 2, respectively. Consequently, to reduce the effects of electricity on global warming, it is recommended to supply electricity from clean energy sources.

Fig S1 shows the impact categories values for the fabrication of 1 kg NMCP material via three different laboratory-scale designs. As revealed in Fig S1, the NMCP materials produced via ball milling, hydrothermal, and
Table 4  Comparison of the electrochemical performance of NMCP materials prepared by different synthesis methods

| Material | Synthesis method | Current rate | First discharge capacity (mAh/g) | References |
|----------|------------------|--------------|----------------------------------|------------|
| NMCP     | Ball milling     | C/100        | 126                              | Hassanzadeh et al. (2016b) |
| NMCP     | Ball milling     | C/30         | 73                               | Hassanzadeh et al. (2016b) |
| NMCP     | Hydrothermal     | C/100        | 76                               | Hassanzadeh et al. (2018) |
| NMCP     | Hydrothermal     | C/30         | 67                               | Hassanzadeh et al. (2016a) |
| NMCP     | Stirring-assisted hydrothermal | C/100 | 94 | Hassanzadeh and Sadrnezhaad (2021) |

Table 5  Overall results for the production of NMCP via different routes based on 1 kg of synthesized NMCP as FU

| Impact category              | Unit            | Ball milling (B) | Hydrothermal (H) | Stirring-assisted hydrothermal (S) | B/H (%) | H/S (%) | S/B (%) |
|------------------------------|-----------------|------------------|------------------|-----------------------------------|---------|---------|---------|
| Ozone depletion              | Kg CFC 11 eq    | 1.54E−06         | 1.29E−06         | 2.04E−06                          | 120     | 64      | 133     |
| Global warming               | Kg CO₂ eq       | 1.53E+01         | 1.42E+01         | 2.00E+01                          | 108     | 71      | 131     |
| Smog                         | Kg O₃ eq        | 8.61E−01         | 9.07E−01         | 1.04E+00                          | 95      | 88      | 121     |
| Acidification                | Kg SO₂ eq       | 5.33E−01         | 1.95E−01         | 1.15E−01                          | 273     | 170     | 22      |
| Eutrophication               | Kg N eq         | 9.21E−01         | 2.89E−01         | 1.08E−01                          | 319     | 245     | 13      |
| Carcinogenicities            | CTUh            | 3.23E−05         | 2.12E−05         | 1.80E−05                          | 153     | 118     | 56      |
| Non-carcinogenicities        | CTUh            | 4.63E−06         | 6.40E−06         | 5.73E−06                          | 73      | 112     | 124     |
| Respiratory effects          | Kg PM2.5 eq     | 1.82E−02         | 1.90E−02         | 1.82E−02                          | 96      | 105     | 100     |
| Ecotoxicity                  | CTUe            | 7.07E+02         | 6.88E+02         | 6.15E+02                          | 103     | 112     | 87      |
| Fossil fuel depletion        | MJ surplus      | 2.54E+01         | 1.74E+01         | 3.39E+01                          | 146     | 52      | 134     |

Fig. 5  Comparative life cycle assessment for producing NMCP; a based on 1 kg of synthesized NMCP as FU, b based on 1 kWh of energy storage capacity as FU
stirring-assisted hydrothermal methods contribute 15.3, 14.2, and 20.0 kg CO₂-eq. per kg NMCP. These values are considerably lower than those reported for ten representative Na₃V₂(PO₄)₃ cathodes for SIBs, which were in the range of 423.9–1380.0 kg CO₂-eq. (Rey et al. 2022). The large difference between the values of global warming impacts in the current study and those reported for Na₃V₂(PO₄)₃, could be attributed to the design of the applied procedures in the current research, which does not require energy-intensive multi-step processes and does not use largely hazardous chemicals.

Comparing the obtained global warming impacts in the current study with those reported previously for LIBS cathodes reveals that the obtained values are nearly close. For example, the obtained global warming impacts were 22 kg CO₂-eq. kg⁻¹ cathode for LiFePO₄ produced by hydrothermal method, 21 kg CO₂-eq. kg⁻¹ cathode for LiNi₀.₄Co₀.₂Mn₀.₄O₂ prepared by aqueous precipitation route followed by calcination (Majeau-Bettez et al. 2011), 21 kg CO₂-eq. kg⁻¹ cathode for LiCoO₂ synthesized by solid-state procedure (Dunn et al. 2015), 32 kg CO₂-eq. kg⁻¹ cathode for LiNi₂Mn₁Co₂O₂, and 21 kg CO₂-eq. kg⁻¹ cathode for LiNiₓCo₁₋ₓAlₓ₂O₅ (Yin et al. 2019). These close values for global warming impacts suggest that the used synthesis procedures are probably at an acceptable level of optimization.

Figure 5b illustrates the results of the comparative LCA of different NMCP production routes when considering 1 kWh of energy storage capacity as the FU. Comparing these results with those shown for 1 kg of NMCP (Fig. 5a) demonstrates that apparently, the environmentally friendlier choice by weight (in each category) does not necessarily translate into the most suitable design when considering battery performance. It is revealed in Fig. 5b that ball milling shows the lowest environmental impacts in seven categories of ozone depletion, global warming, smog, non carcinogenics, respiratory effects, ecotoxicity, and fossil fuel depletion. The high impact of ball milling in three categories of acidification, eutrophication, and carcinogenics could be attributed to the higher amount of emission (mainly NO₃⁻ and HPO₄²⁻) which had the dominant effect in these categories in the ball milling process in comparison with those in the hydrothermal-based methods (as seen in Tables 1, 2, and 3).

To detect the main source of the environmental impacts in each synthesis process, the contribution of the system inputs and outputs (total emissions to air and water) in all three processes were analyzed and the results are shown in Fig. 6 and also Fig. S2. The difference between Fig. S2 and Fig. 6 is that for example, in the ball milling process, the electricity analyzed in Fig. S2 is the sum of electricity consumed in the three-unit processes of ball milling, stirring, and oven drying. In Fig. S2, the contribution of total electricity in each impact category is analyzed, while in Fig. 6 the relative contribution of each unit process (ball milling, stirring, and oven drying) in each impact category is investigated. Similar analyses have been done for hydrothermal-based methods. Note that the inputs are the same in all three methods. In order to know the percentage impact contribution of each component of the process, the 100% columns histogram was used. The three methods’ impact cannot be compared according to the column scale.

As demonstrated by Fig. S2 and also Fig. 6a, for the ball milling method, electricity contributed much of the total impacts in ozone depletion, global warming, and fossil fuel depletion categories, whereas direct emissions contributed more than 80% of the total impact for the acidification and eutrophication categories. Moreover, Manganese nitrate contributed 50%, 60%, and more than 90% of the total impact for the respiratory effects, ecotoxicity, and carcinogenics categories, respectively.

In the hydrothermal method (Fig. 6b), sodium carbonate as input material demonstrates the highest contribution ranging from 40% to about 80% in 7 categories. The reason is due to the high required amount of sodium carbonate to synthesize pure NMCP through hydrothermal-based methods, which is about 10 times higher than the stoichiometric value, as previously optimized (Chen et al. 2012a). And finally, the main contributors to the stirring-assisted hydrothermal process are electricity and sodium carbonate (Fig. 6c). Consequently, it is recommended to supply electricity from clean energy sources or substitute sodium carbonate with an eco-friendlier material.

As seen in Fig. 6a–c, deionized water, stirring, and oven drying exhibit small environmental impacts in all three fabrication methods. Besides, direct emissions are the main contributor to the impact categories of acidification and eutrophication, and manganese nitrate is the main environmental weakness in the carcinogenics category, in almost all three synthesis routes.

In the categories of non-carcinogens, respiratory effects, and ecotoxicity, more than 85% of the environmental impacts originated from the use of raw materials including manganese nitrate, disodium phosphate, and sodium carbonate. Electricity in the hydrothermal process showed little environmental impact while in ball milling and S-hydrothermal processes, it contributed more than 50% of the total impact for the ozone depletion and fossil fuel depletion categories.

Fig. S3 shows the contribution of each unit process for producing NMCP based on 1 kWh of energy storage capacity as FU, via the three aforementioned various methods. The findings are nearly similar when considering 1 kWh of energy storage capacity and 1 kg of synthesized NMCP as FUs (Fig. 6 and S3). In both cases and all three processes, direct emissions are the main environmental weaknesses in acidification and eutrophication categories, and manganese nitrate is the main contributor (about 90%) to the impact category of carcinogenics. The latter could be attributed to the available data of the carcinogenic category in the Ecoinvent
v3.0 database, in which 1 kg of manganese and nitric acid as the main inputs of the manganese nitrate production process contributes 5.26E−05 and 4.17E−08 CTUh, respectively. It is worth noting that the share of deionized water and electricity in the category of carcinogens was minor, and hence, their expression was omitted. Since the amount of consumed manganese was 0.31 kg (as seen in Table S1) and it has a larger effect on the carcinogenic category, the use of manganese can justify the carcinogenicity of manganese nitrate.

Electricity in the hydrothermal process showed little environmental impact while in ball milling and S-hydrothermal processes, it is one of the main contributors to the categories of ozone depletion, global warming, smog, and fossil fuel depletion. In hydrothermal-based methods, sodium
carbonate showed a considerably high impact in almost all categories. These results could be attributed to the high consumption of electricity and sodium carbonate in the related processes.

Noted to mention that laboratory-scale equipment such as oil bath and glass bottle, are not energy efficient and may not be the best available technology for upscaling. For example, instead of using an oil-bath/glass bottle system in the S-hydrothermal process, stirred autoclaves would be appropriate choices to be used on an industrial scale. Furthermore, fossil fuels are the largest sources of energy for electricity generation which produce large quantities of carbon dioxide when burned. Consequently, if part of the electricity is supplied by renewable resources such as wind, the effect of electricity in the global warming impact category would probably be decreased. In addition, sodium carbonate as the main contributor to various impact categories could be substituted by an eco-friendlier chemical.

Sensitivity analysis as a vital part of interpretation in LCA was conducted on the variation of main process inputs in ±10% to explore uncertainty and the variability of environmental impacts to produce 1 kg NMCP. Tornado plots of four specified impact categories for ball milling, hydrothermal, and stirring-assisted hydrothermal methods are demonstrated in Figs. S4, S5, and S6, respectively. As shown in Fig. S4, if the electricity consumption is increased by 10%, the global warming impact category of the NMCP production through ball milling changes by 4.47%. Correspondingly, the respiratory effects impact category increases by 4.98%, if the manganese nitrate consumption is increased by 10%. Moreover, any modification in disodium phosphate consumption by 10% changes the acidification and eutrophication by 0.68% and 0.34%, respectively.

Moreover, the environmental burdens in the hydrothermal synthesis could be increased by 1.79 to 7.36% reliant on the category, where the sodium carbonate is increased by 10% (Fig. S5). This input (sodium carbonate) has a significant contribution in almost all categories, as also revealed in Fig. 6b. Fig. S6 also demonstrates that any modification in sodium carbonate consumption by 10% changes the respiratory effects, global warming, acidification, and eutrophication categories by 4.91%, 4.42%, 4.12%, and 3.71%, respectively.

Finally, and as aforementioned, the best attainable electrochemical performance is not the only determining factor for choosing the appropriate synthesis route for the large-scale production of electrode materials. Instead, the best appropriate route should be adopted according to both electrochemical performance and environmental impact. By considering kWh of energy storage capacity as the FU, the results will be scaled for equivalent functionality. When considering 1 kWh of capacity as the FU, ball milling showed the least environmental impact in all categories except acidification, eutrophication, and carcinogenics. Consequently, the ball milling method could be identified as the most appropriate route for the production of NMCP as the cathode material in sodium-ion batteries.

Noted that the studied processes are not fully optimized for scale-up or mass production capability. Generally, this work provides support for battery developers and assists future advances in the development of sustainable cathodes applied to beyond-Li-ion technologies.

Conclusion

This study evaluated different synthesis routes for the production of a promising cathode material from a comparative LCA perspective. NMCP was chosen as the test cathode material and the environmental impact of its production via three different methods, namely ball milling, hydrothermal, and stirring-assisted hydrothermal was investigated in order to explore what unit processes or inputs/outputs drive impacts for each method and finally which route could have the lower environmental burden. The findings suggested that the most dominant impacts in the ball milling production of NMCP come from electricity, direct emissions, and manganese nitrate, while the main contributors to most impact categories in hydrothermal and stirring-assisted hydrothermal methods are sodium carbonate, and sodium carbonate plus electricity, respectively. When considering 1 kWh of capacity as the FU, ball milling showed the least environmental impact in all categories except acidification, eutrophication, and carcinogenics. Consequently, ball milling could be identified as the most appropriate route for the production of NMCP as the cathode material in SIBs.

The presented analysis which is at an early stage can navigate the developers of electrode materials for energy storage devices in choosing an environmentally friendlier production method when designing new materials, which should be an important goal in materials engineering for energy storage.

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Declarations

Conflict of interest “The authors declare that no funds, grants, or other support were received during the preparation of this manuscript.”

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