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Life cycle assessment on calcium zincate production methods for rechargeable batteries

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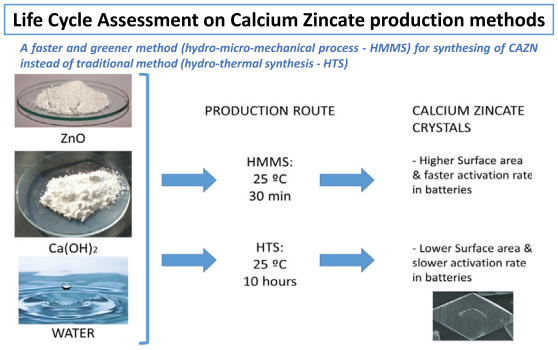
HIGHLIGHTS

- The raw materials shape the environmental results of calcium zincate production.
- Particle size and reaction time are key factors affecting the results of the LCA.
- The reaction time should be considered when calculating chemical LCAs.

GRAPHICAL ABSTRACT



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ABSTRACT

The world's energy transition from fossil to renewable energy is unthinkable without further research in energy storage. Decreasing the environmental impacts from the production of energy storage technologies is essential for achieving a green energy transition. Calcium Zincate (CAZN) is used as active material in rechargeable zinc-based batteries (and other products, such as heterogeneous catalysts for biodiesel or antifungal products). They present a low-cost, safer, alternative to Lithium based batteries and are targeted as replacement solutions for lead-acid batteries.

We propose a novelty in the synthesis of CAZN, the hydro-micro-mechanical process (HMMS). The residence time of this new route is about 20 times lower than the traditional processes, so its production needs less infrastructure and can deliver quicker at an industrial scale. In addition, laboratory tests indicate that HMMS CAZN has more reaction surface area and the activation of the battery is 1.77 times faster.

Using the life cycle assessment (LCA) method, we compare this new process with the current best option, hydro-thermal synthesis (HTS). The cradle-to-gate results per kg of CAZN already indicates that HMMS is an environmentally

Abbreviations: CAZN, Calcium zincate; CC, Climate Change; CC bc, Climate Change, including biogenic carbon; CSTR, Continuous Stirred Tank Reactor; EEA, European Economic Area; EOF, Photochemical Ozone Formation, Ecosystems; EoL, End-of-Life; ET fw, Freshwater ecotoxicity; ET marine, Marine ecotoxicity; ET terra, Terrestrial ecotoxicity; EU, European Union; FC, Freshwater consumption; FE, Freshwater Eutrophication; FD, Fossil depletion; FU, Functional Unit; HCS, Hydro-chemical synthesis; HMMS, Hydro-micro-mechanical synthesis; HMS, Hydro-mechanical synthesis; HOF, Photochemical Ozone Formation, Human health; HT carc, Human toxicity, cancer; HT ncarc, Human toxicity, non-cancer; HTS, Hydro-thermal synthesis; IR, Ionizing Radiation; LCA, Life cycle assessment; LCI, Life cycle inventory; LCIA, Life cycle impact assessment; LU, Land use; MD, Metal depletion; ME, Marine eutrophication; ODP, Stratospheric Ozone Depletion Potential; PMF, Fine Particulate Matter Formation; TAP, Terrestrial Acidification Potential; UK, United Kingdom; UMA, University of Málaga; IPCC, Intergovernmental Panel on Climate Change.

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better alternative for all indicators; especially when considering the normalization of the results with the residence time and the surface area, HMMS delivers better results, with improvements of 97 % in global warming, for instance. With this, we demonstrate that, outside of the cradle-to-gate, variables that make the final products better service units or give more function should be considered as valuable additional information when deciding among alternatives. This also highlights the importance of life cycle thinking when working with chemical processes and substances.
In the sensitivity analysis, we developed 7 scenarios related to the energy demand of the processes, and we incorporated the projection in the European electricity mix for 2030 and 2050.

1. Introduction

Energy storage is an important part of the transition to a carbon-free economy, and it meets several of the Clean Energy for All Europeans package's fundamental objectives. It provides a real technique of enhancing energy efficiency and integrating additional renewable energy sources into electrical networks by balancing power grids and conserving surplus energy (European Commission, n.d.).

Zinc-batteries present a low-cost, potentially safer alternative than other batteries produced from metals such as lithium or sodium, which typically use flammable organic electrolytes. Other properties, such as low potential (leading to a high cell potential), great reversibility (quick kinetics), low equivalent weight, and compatibility with aqueous electrolytes make it a popular battery material (Tsai and Chan, 2013). Calcium zincate (CAZN) can be used in the production of: i) zinc anode of alkaline electrochemical generators; ii) heterogeneous catalysts for the production of biodiesel; or iii) antifungal products. Zinc-batteries electrodes are the most common use of this product.

CAZN is traditionally produced through the following types of synthesis: hydro-chemical synthesis (HCS) (Li and Zhou, 2012; Sharma, 1986), hydro-thermal synthesis (HTS) (Wang et al., 2008), and hydro-mechanical synthesis (HMS) (Yang et al., 2004; Zhu et al., 2003). These syntheses require a long residence time and numerous production steps that make these processes both impracticable and environmentally unfriendly when developed at industrial scale (Caldeira et al., 2017b, 2017a; Lacoste and Thiel, 2016; Li and Zhou, 2012; Sharma, 1986; Wang et al., 2008; Yang et al., 2004; Zhu et al., 2003). For example, HCS uses potassium hydroxide (Li and Zhou, 2012; Sharma, 1986), which is expensive and must be removed at the end of the process. HTS usually requires heating up and stirring a substantial amount of water to 75 °C to reach the acceptable conversion (Wang et al., 2008). HMS is not continuous, cannot be scaled to an industrial scale, and it is energy-intensive (Yang et al., 2004; Zhu et al., 2003).

Hydro-micro-mechanical synthesis (HMMS) is an innovative way of synthesis (Caldeira et al., 2017a, 2017b), patented in 2016 (Lacoste and Thiel, 2016), which might be operated continuously and on an industrial scale. The proposed novelty opens the door to the industrialisation of this product, with a green, fast, and simple process.

In this paper, we are comparing HMMS with HTS. HTS is the most competitive of all the conventional methodologies because it only uses water as a solvent and it has a relatively low residence time, compared with the other processes. We validated the data from scientific and technical literature related to HMMS and HTS syntheses reactions with several experiments. The experiments for HMMS were developed by DEASYL SA (a spin-off company of EASYL SA) in their laboratories in Switzerland and the experiments for HTS were performed at the University of Málaga (UMA) in Spain.

We compare these processes using the Life Cycle Assessment (LCA) methodology. LCA provides a comprehensive and holistic view of the environmental loads of the products or services understudy, covering a wide set of environmental impact categories (Baumann and Tillman, 2004; Molander et al., 2004; Muñoz, 2006; Puig et al., 2013). Operation within companies and production chains of environmental options follow the principles of Life Cycle Management (Fullana i Palmer et al., 2011). This methodology is widely used to measure impacts throughout the lifespan of products in different sectors, also at industrial scale (Arfelis Espinosa et al., 2022; Nabavi-Pelesaraei et al., 2019). In the present article, this is of relevance because, although both processes synthesise the same product (CAZN), its particle size (of the product) depends on the process used, and this will affect downstream processes. Comparing two production processes of the same product allows the LCA practitioner to focus mainly on those stages of the production chain that are different from each other, assuming that all other stages that are equivalent have the same environmental impact between the two processes (Fullana-i-Palmer et al., 2009). As far as known by the authors, this is the first environmental study of the calcium zincate production process.

1.1. Hydro-thermal synthesis (HTS)

According to the scientific literature (Wang et al., 2008), this process consists of heating the stoichiometric mixture of calcium hydroxide ($Ca(OH)_2$) and zinc oxide (ZnO) at 75 °C for 24 h (12 h of reaction and 12 h of aging) with an excess of distilled water. CAZN crystals are retrieved by centrifugation, washed with demineralized water two times, and dried at 50 °C for 2 h in vacuum. Unlike in the HCS, the HTS does not require a strong alkali medium or other solvent than water.

The reaction was replicated by DEASYL SA at the UMA for the purposes of our research. High conversion rates were achieved in only 10 h. In addition, the crystals were obtained at ambient temperature with no need of heating the mixture to 75 °C. To be in the safe side, the HTS inventory data used for the comparison with HMMS are the best ones: the primary data that were tested in UMA. The flow diagram in Fig. 1 presents the steps included in this process.

1.2. Hydro-micro-mechanical synthesis (HMMS)

This method is an evolution of the conventional process of HMS. It consists of introducing a stoichiometric [2·ZnO + Ca $(OH)_2$] aqueous suspension through a high-efficiency continuous-flow mechanochemical reactor filled with an appropriate amount of zirconia oxide (ZrO_2) micro-milling balls. These micro-balls (0.5 to 1 mm in diameter) do not generate any appreciable pollution because of their wear and have a large lifespan of approximately 10,000 h.

The milling step lasts only a few minutes. It is used to activate and reduce the residence time of the reaction which takes place in a Continuous Stirred Tank Reactor (CSTR) in 30 min:

$$2ZnO(s) + Ca\,(OH)2(s) + 4H2O \rightarrow Ca\,(OH)2 \cdot 2Zn\,(OH)2 \cdot 2H2O(s) \quad (1)$$

The best efficiency of the process is obtained for a water mass ratio between 3:1 and 5:1 at ambient temperature for 1–3 min, with a 30 to 90 L/h flow rate and a suspension of 300–600 g/L (Caldeira et al., 2017a, 2017b; Lacoste and Thiel, 2016). This process is described by the flow diagram in Fig. 2.

2. Materials and methods

2.1. Materials

Materials are key to understanding the overall impact of services or products, both per type and per quantity. Therefore, the design stage

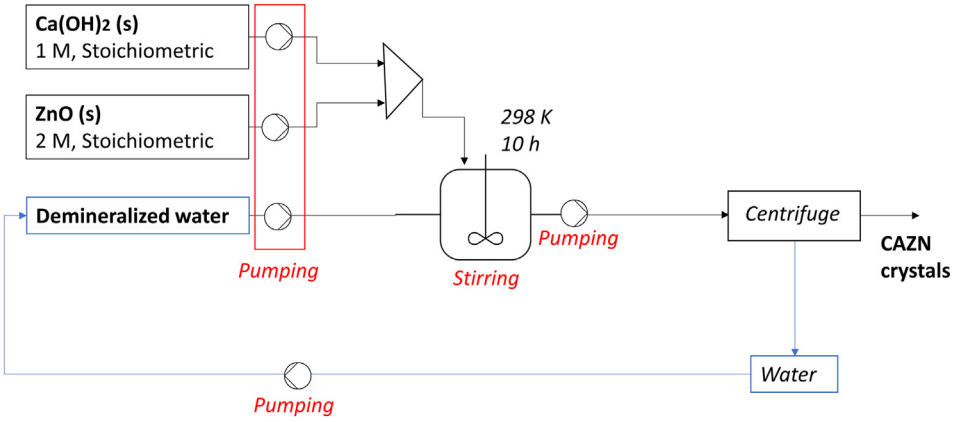


Fig. 1. Boundaries for the hydro-thermal process (Wang et al., 2008).

(such as laboratory conditions) must be given priority attention (Civancik-Uslu et al., 2018).

The materials utilized in the laboratory experiments and considered for the environmental evaluation comply with the criteria specified in the publications and patents of the HTS (Wang et al., 2008) and HMMS (Caldeira et al., 2017a, 2017b; Lacoste and Thiel, 2016) processes: i) the $Ca(OH)_2$ with particle size from 0.01 to 1 μm, and purity of 96 % or greater; ii) the ZnO with particle size from 0.01 to 1 μm, and purity of 99.9 % or greater; and iii) water, which has been purified with a demineralization process.

2.2. Quality of data

Most of the data that we use in the study are primary data tested at a laboratory scale. In addition, academic literature, scientific databases, or modelling complement these primary data. For instance, we use the publication of Piccinno et al., (2016) on advanced process calculations to scale up from the laboratory scale to the industrial scale. This benchmark is widely used in the literature on LCA for chemical reactions (Arfelis Espinosa et al., 2022).

All the secondary data were evaluated qualitatively using the Pedigree matrix (Ciroth, 2009; Weidema and Wesnæs, 1996), all data being sufficiently reliable, complete, and without temporal, geographical, or technological differences. Even though, not having fully operational industrial scale data presents limitations, which are common in chemical LCAs.

2.3. Life Cycle Assessment (LCA)

The environmental impact has been calculated with the LCA methodology, which is based on the ISO 14040 (ISO 14040:2006(en), 2006) and ISO 14044 (ISO 14044:2006(E), 2006) standards, and which is used to compare alternatives (Fullana-I-Palmer et al., 2009).

LCA is structured in four different iterative phases: i) Definition of Goal and Scope, ii) Development of Life Cycle Inventory (LCI), iii) Life Cycle Impact Assessment (LCIA), and iv) Interpretation of results.

The model for the LCA is developed using GaBi Professional Software (version 10.6.1.35), including its datasets (Sphera, n.d.) to characterize and quantify the impact of the raw and auxiliary materials. This is the world's leading LCA modelling and reporting software with the largest LCA databases for chemical products. In addition, AspenPlus (version 12.2) is used to simulate the processes, model the mass and energy balances for the LCI, and check the physical and chemical properties of the substances. This is the most widely used software for chemical process simulation and has extensive databases for this purpose.

The ReCiPe 2016 (version 1.1 midpoint, Hierarchist model) methodology is used for the calculation of LCIA (Huijbregts et al., 2017, 2016). According to a recent review on 47 different LCA of chemical reactions, this is the preferred method in this type of LCA (Arfelis Espinosa et al., 2022), mainly because it brings together the main authors of the most previously used methodologies and because it perfectly summarises the long list of LCI results in its different environmental indicators (Ordikhani et al., 2021). In addition, to maintain simplicity and have visual results, we cut off the emission sources with an environmental impact of less than 1 %.

2.3.1. Boundaries and scope

The present LCA consists of a cradle-to-gate approach, from the extraction of the raw material to the production of CAZN. The End-of-Life (EoL) of CAZN crystals after its use, typically in zinc batteries, is not included in the scope. Regardless of its production synthesis, the rate of Lead acid batteries and accumulators which are collected for recycling in the EU is 90 % (EPRS | European Parliamentary Research Service, 2022), we expect the Zinc based batteries to follow the same pattern. Water treatment, crystals washing, and drying requirements are equivalent in both syntheses, so they can be omitted for the comparison. The same happens with the distribution stage (batteries weight and volume are identical) and use stage (the efficiency of the battery is independent of the CAZN synthesis).

To calculate the environmental impact of the chemical processes, we have considered the following emission sources: i) input material flows;

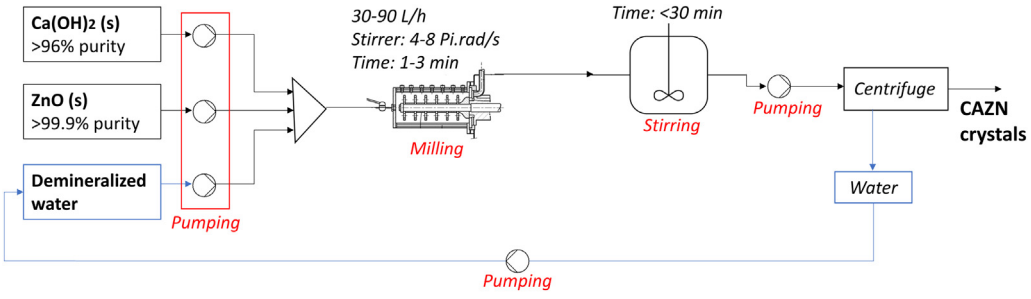


Fig. 2. Boundaries of the hydro-micro-mechanical process (Caldeira et al., 2017a, 2017b; Lacoste and Thiel, 2016).

ii) flows of the energy used to heat or cool the reactants; and iii) energy used in stirring, pumping, or milling the mixture.

2.3.2. Functional unit, allocation factors and normalization

The functional unit (FU) serves as the reference basis of the calculation related with the environmental impacts of the system under study (ISO 14040:2006(en), 2006; ISO 14044:2006(E), 2006). In this case, the FU selected for the present study is 1 kg of CAZN for both processes.

The authors have identified that, in LCAs relating to the chemical industry sector, special care must be taken in the selection of the FU. Although outside of the cradle-to-gate LCA practice, there are variables such as the particle size of the final product or the residence time of the reaction which make the final product with more service units or more function. Therefore, this should be considered as valuable additional information when deciding among alternatives. The result of the LCA with an FU depending only on mass does not take this into account. There are several chemical LCA papers that already perform the study with a flowrate as FU (Battista et al., 2017; Bello et al., 2020; Guzmán et al., 2021; Taher et al., 2020). Another example is the consideration of module D within the EN 15804:2012 + A2:2019 for the consideration of information out of the cradle to gate scope in construction products LCAs (Albertí et al., 2019; European Standards, 2019). In the present paper, the results of the LCA per kg of intermediate product are changed in the sensitivity analyses with the application of the normalization factors described below.

The production routes present differences in the average particle size of the CAZN crystals. The particle size of the crystals directly affects the surface area, and it has an impact on the activation time of the battery. DEASYL SA, compared the standard CAZN crystals synthesized with HMMS (8–12 μm) (Caldeira et al., 2017a, 2017b; Lacoste and Thiel, 2016) with the larger grain size synthesized with HCS (70–1000 μm) (Li and Zhou, 2012; Sharma, 1986), observing three times faster activation of the battery with the HMMS crystals. This phenomenon occurs due to a global feature in the field of interfacial electrochemistry: the more the contact surface area increases, the more the reaction velocity increases. The activation speed with HTS crystals (10–200 μm) (Wang et al., 2008) would be 1.77 times slower than with HMMS crystals, obtained with linear interpolation of the previously tested activation speeds of HMMS and HCS crystals. Furthermore, the difference in residence time of the processes means that the production rate of the HTS process is in the order of 20 times slower than HMMS.

According to the HTS experiments performed in the UMA laboratories, the HTS reactor volume must be about 3.33 times higher than the one for HMMS, as expected from the values of the kinetics of both reactions. For higher productions, several CSTR of equal size in series should be used to get reasonable reaction sizes. Fig. 3 shows the conversion of $Ca(OH)_2$ as a function of the CSTR volume for a CAZN production with the HTS and the HMMS. This difference in reactor volume has been considered to account for the excess water used in each type of process when calculating the mass and energy balances.

2.3.3. Life cycle inventory

The LCI comprises the input and output mass and energy flows. Table 1 presents these inventories for each one of the reactions per 1 kg of synthesized CAZN.

As shown in the bill of materials, HMMS is a process with lower energy (electricity) and freshwater consumption. We have considered the European energy mix of 2017 from professional GaBi databases (Sphera, n.d.) for the study, as well as the projection (without consideration of the consequences of the war in Ukraine, yet) for years 2030 and 2050, according to the "EU Reference Scenario 2016 - Energy, Transport and GHG Emissions - Trends to 2050" published by the European Commission in 2016 (Capros et al., 2016). These electricity projections are mainly used in LCA of the energy sector (Reinert et al., 2021). However, in LCA of chemical reactions, where they also provide important value, they are not so commonly used (Arfelis Espinosa et al., 2022). Years 2030 and 2050 are selected because of the European Commission's plans in the European Green Deal

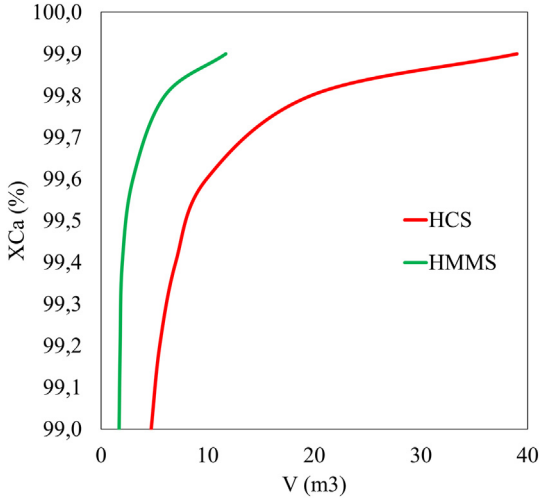


Fig. 3. Conversion by HTS and HMMS as a function of the volume of the reactor.

(European Commission, n.d.), as well as the availability of data on the projected energy mix in these years, both in the software used and in the literature. Inventories of input materials are described below.

2.3.4. LCI of the production of the micro-milling balls for HMMS

The micro-milling balls used in the HMMS are composed of 78–95 % of ZrO_2 and other metals. They cover about 60 % of the total volume of the reactor chamber (Lacoste and Thiel, 2016). The environmental impacts from the ball milling step in the production of HMMS crystals relate to the extraction of zirconium. Also, the environmental impacts of the production of the micro-milling balls are comparable to the environmental impacts of the production of ZnO (Stieberova et al., 2019), one of the raw materials used in HMMS and HTS. In addition, the milling balls in HMMS can be reused for a lifespan of 10,000 h.

Since one cycle in the ball milling step of HMMS lasts few minutes, the impact of the production of the milling balls allocated to one cycle is less than 0.01 % and neglected for this study.

2.3.5. LCI of the calcium hydroxide production

According to GaBi professional database (Sphera, n.d.), calcium hydroxide is produced when water is slowly added to previously crushed or powdered quicklime (calcium oxide). The quicklime is made by burning different types of limestone.

2.3.6. LCI of the zinc oxide production

ZnO is a multifunctional material with unique physical and chemical properties, due to its high chemical stability, high electrochemical coupling coefficient, a broad range of radiation absorption, and high photostability

Table 1
Bill of materials of HMMS and HTS.

Mass balance	HMMS	HTS
In		
$Ca(OH)_2$ (kg)	0.24	0.24
ZnO (kg)	0.53	0.53
Water (L)	3.07	10.13
Out		
Water (L)	2.76	9.117
Calcium zincate (kg)	1.00	1.00
Energy Balance	HMMS	HTS
Pumping (kwh)	$1.01 \cdot 10^{-5}$	$3.06 \cdot 10^{-3}$
Stirring (kwh)	$4.67 \cdot 10^{-4}$	$2.72 \cdot 10^{-2}$
Milling (kwh)	$8.00 \cdot 10^{-3}$	0.00
Centrifugation (kwh)	$2.11 \cdot 10^{-2}$	$5.99 \cdot 10^{-2}$
Total energy consumption (kwh)	$2.96 \cdot 10^{-2}$	$9.02 \cdot 10^{-2}$

(Kolodziejczak-Radzimska and Jesionowski, 2014). There are three main industrial large-scale production processes: i) pyrometallurgical synthesis; ii) hydrometallurgical synthesis; and iii) by-product of other chemical reactions. The pyrometallurgical synthesis is the most utilized process.

Pyrometallurgical synthesis for ZnO production is based on the roasting of zinc ore according to the ISO 9298 standard ("ISO 9298:2017 Rubber compounding ingredients — Zinc oxide — Test methods," 2017). It can be performed through the direct process (the American process) and classified as type A, or through the indirect process (the French process) and classified as type B. The indirect process is the most used in Europe. In this process, the raw material is zinc metal. Metallic zinc is melted in a furnace and vaporized at ca. 910 °C. Then, ZnO is produced as an immediate reaction between the zinc vapor with the oxygen from the air (Kolodziejczak-Radzimska and Jesionowski, 2014).

We have simulated the French process to produce ZnO (Fig. 4) to calculate its environmental impacts, because these are not directly available in the GaBi professional database (Sphera, n.d.). We have considered the French process for both HMMS and HTS, and not the American one, as both routes have been studied in Europe.

2.3.7. LCI of the transport stage

For the transport stage of the raw materials, we have assumed a diesel-driven Euro V truck of 20–26 t gross weight and 17.3 t payload capacity. This capacity is the average value of the alternatives available in the GaBi professional databases (Sphera, n.d.) with 2020 as a reference year. Euro V is the vehicle emission standard for exhaust vehicles sold in the European Union (EU) and European Economic Area (EEA) member states and the United Kingdom (UK) between 2008 and 2012 (Stratstone, 2021). This period corresponds with the average age of trucks in Europe (ACEA, 2021). The Euro V standards are defined in a series of EU directives staging the progressive introduction of increasingly stringent standards.

Since batteries weight and volume are identical for both processes, only raw materials transport is included in the study, where a transportation distance of 100 km has been assumed.

2.4. Scenario building

2.4.1. Energy demand

After selecting the most efficient processes (HTS and HMMS), we have compared their environmental impacts. Thus, we have considered each of the differences that may exist in each reaction (i.e., residence time and particle size) and have completed an exhaustive study with 7 different scenarios of energy demand in different stages of the overall process.

The scenarios have been selected to evaluate the uncertainty generated by some data discrepancies in the different literature sources used for each process or to evaluate different operational conditions that are being studied and optimised for each stage of the processes in the laboratory.

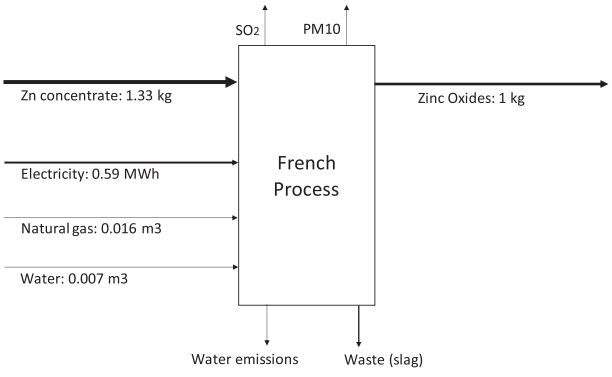


Fig. 4. French Process input/output diagram for ZnO production ("ISO 9298:2017 Rubber compounding ingredients — Zinc oxide — Test methods," 2017).

- 1_HEAT_MID, the temperature in CSTR in HTS is 50 °C. This is the average value between the experiments performed in UMA and the temperature reported in the literature (Wang et al., 2008).
- 2_HEAT_HIGH, the temperature in CSTR in HTS is 75 °C. This is the value reported in the literature (Wang et al., 2008).

For the two first scenarios, a theoretical value of 20 % of energy savings is applied for heating and cooling (Chauvy et al., 2021; Hischier et al., 2005; Pereira et al., 2014; Piccinno et al., 2016). This is associated with optimization and process integration when it is upscaled from the laboratory to the industrial scale. According to the range reported in Piccinno et al. (2016), we add 5 more scenarios:

- 3_CF_LOW, with the lower energy demand value for centrifugation in both HTS and HMMS.
- 4_CF_HIGH, with the higher energy demand value for centrifugation in both HTS and HMMS.
- 5_DRY_YES, considering that the final product is being dried before the transport stage in both HTS and HMMS.
- 6_MILL_LOW, with the lower energy demand value for milling in HMMS.
- 7_MILLING_HIGH, with the higher energy demand value for milling in HMMS.

2.4.2. Electricity mix

The European electricity mix is expected to change due to the more restrictive directives from the European Commission and the European Green Deal implementation (European Commission, n.d.). This aspect influences the impact categories. Therefore, to assess possible future environmental impacts, we developed a sensitivity analysis based on the future electricity grid mix in Europe for years 2030 and 2050 according to EU Energy trends data available in the Professional GaBi Databases (Sphera, n.d.). The electricity mix sources are presented in Fig. 5.

2.4.3. CAZN-transport stage

CAZN crystals used as active material in zinc electrodes through a water-based process coating or paste, to be used in zinc batteries electrodes. It does not seem necessary to remove water after the centrifugation stage so we can reduce the electricity consumption for drying and the water consumption in the use phase. Even though, adding a drying stage also has benefits, taking up between 5 and 6 times less volume when transported from one place to another, as can be calculated from CAZN density and CAZN concentration in zinc-batteries electrodes (Caldeira et al., 2017b, 2017a; Jain et al., 1992).

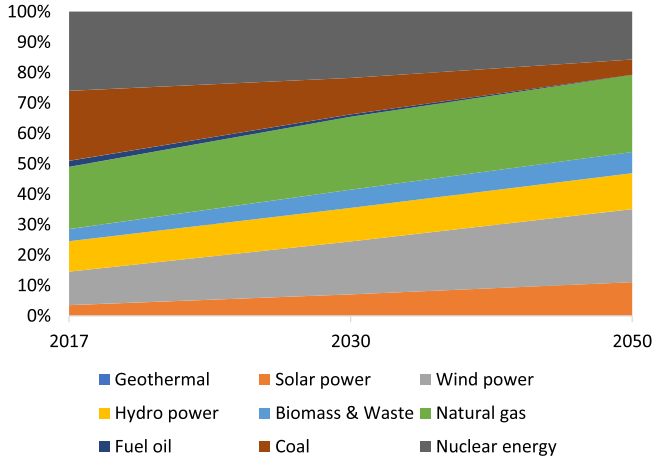


Fig. 5. Projection electricity mix EU27 in 2030 and 2050.

Therefore, we have decided to include a study of the transport phase of the CAZN product to evaluate under which circumstances it is better to transport the product directly in suspension in water, and when it is better to pre-dry the final product to transport it in powder form.

Climate change is the principal environmental impact related to transport. We have used it to calculate the equilibrium distance of the environmental performance of two scenarios: transporting CAZN crystals mixed with water (without a drying stage) and transporting CAZN crystals in powder (with a drying stage).

3. Results and discussion

3.1. General results

The results obtained for the whole LCA of CAZN production with HMMS and HTS show that most of the impacts are related to the extraction and refining of the ZnO because of its high energy consumption. This is presented in Fig. 6.

To compare HMMS and HTS, we have analysed the environmental impacts of only the CAZN production processes under the FU of 1 kg of CAZN. As expected, the results (Fig. 7) are already favourable for HMMS at some midpoint indicators.

When normalizing the results of HTS by the particle size (multiplying by 1.77) and residence time (changing FU from kg to kg/h), the HMMS performs better in all the indicators of the ReCiPe 2016 methodology, as presented in Fig. 8. Reaching about 44 % of improvement in Climate Change indicator when normalizing only by the particle size, and improving by 97 % in the same indicator when normalizing by both the particle size and the residence time.

Moreover, HMMS can perform even better in all the midpoint indicators if freshwater consumption is reduced. It is relevant to state that latest DEASYL SA experiments achieved a 3:1 ratio mass proportion of water-reactants without affecting the efficiency of the reaction.

3.2. Sensitivity analysis

3.2.1. Energy demand

The comparison of the results for all the seven scenarios is presented in Fig. 9. The process (HMMS or HTS) with less environmental impacts is

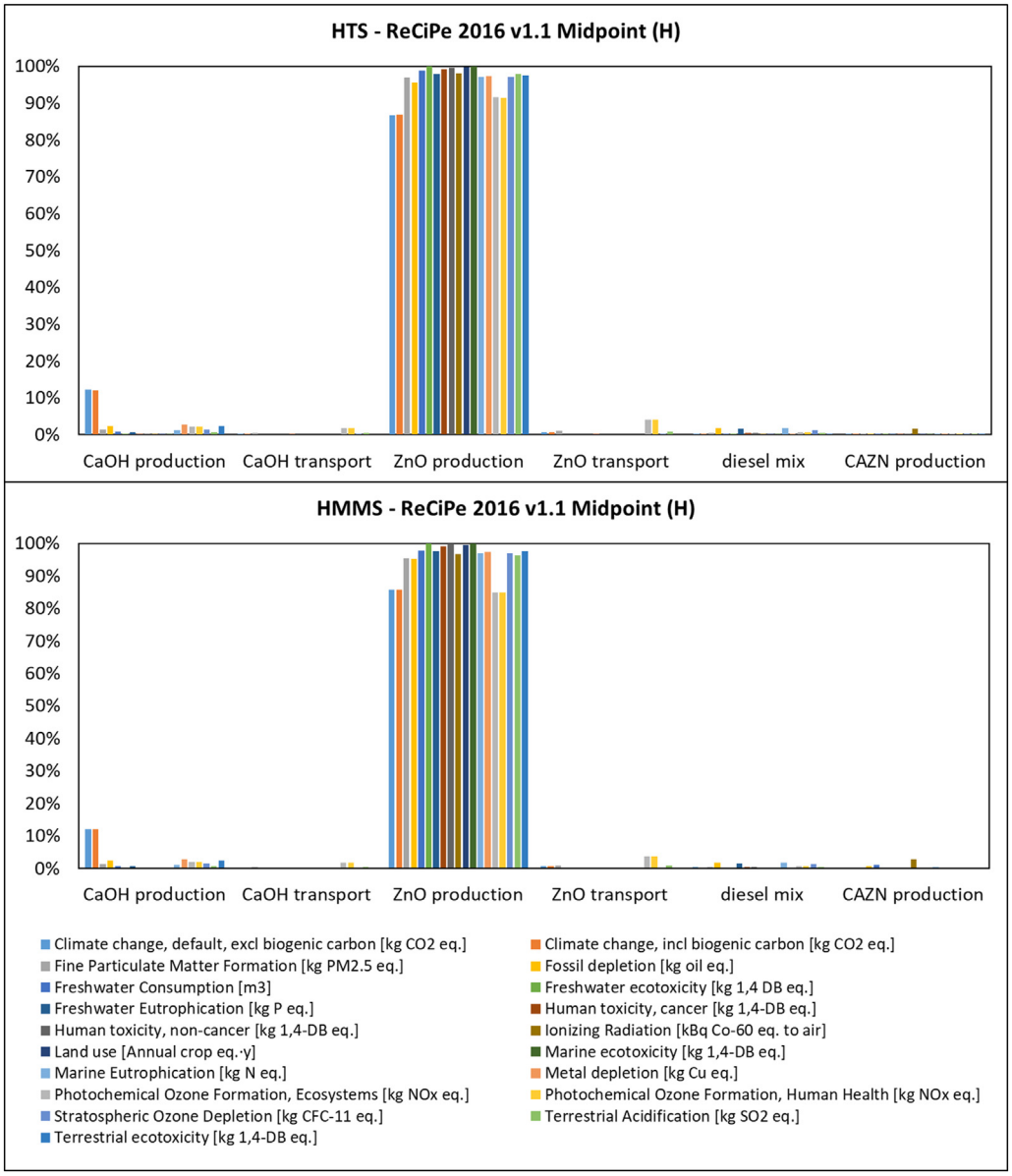


Fig. 6. LCA indicators of CAZN from the extraction of the raw material to transport stage of the final product.

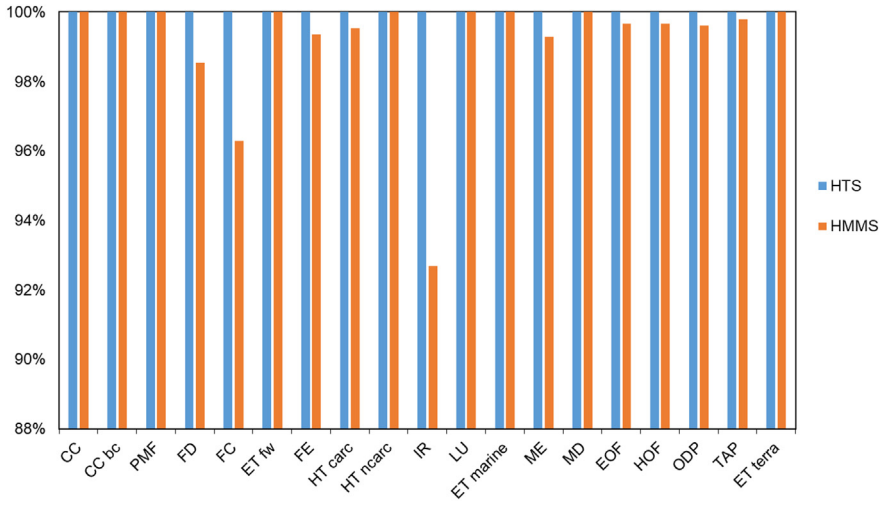


Fig. 7. LCA indicators of CAZN production stage for HTS and HMMS (FU = 1 kg of CAZN).

highlighted in green and the process with more environmental impacts is highlighted in red.

According to Fig. 9, when neither the particle size of the CAZN crystals nor the residence time of the reaction are considered, the HMMS is environmentally better for all the indicators in the scenarios which involve heating in the HTS (scenarios 1 and 2), but only for a few indicators in the other scenarios. For the remaining indicators in scenarios 3–7, the environmental impact is similar for both processes.

Even though, particle size has a direct relationship with activation rate in its use in batteries, as well as a direct relationship with its performance when used as antifungal product or heterogeneous catalyst. Residence time has also a high impact in the economic model of the system since all the infrastructure of the HTS must be upscaled 20 times to reach similar production rate of HMMS.

When considering both, particle size and residence time, the environmental impacts are lower in the HMMS case for all the indicators in all the scenarios.

3.2.2. Electricity mix

We have projected, in Fig. 10, the LCA results with the European electricity mix of 2030 and 2050 (Capros et al., 2016). HMMS shows a better environmental performance than HTS for all the indicators in the next 30 years.

According to the results, only human toxicity and land use indicators are increasing in next years. This is due to the future energy mix foreseen for the years 2030 and 2050 according to the European Commission's green deal (European Commission, n.d.). The increase in the share of energy produced from green and renewable sources leads to a reduction of the environmental impact on most midpoint indicators (i.e., climate change). However, these energy sources are not favourable in absolutely all midpoint indicators. The impact of some clean energy sources on land use (solar energy, wind energy, biodiesel....) is better known (Ritchie, 2022; van de Ven et al., 2021). Also, in the case of human toxicity potential, the electricity produced from wind energy (sharing about 30 % of power generation in 2030 and 47 % in 2050) has higher environmental impact than electricity produced from gas, for instance (Atilgan and Azapagic, 2016).

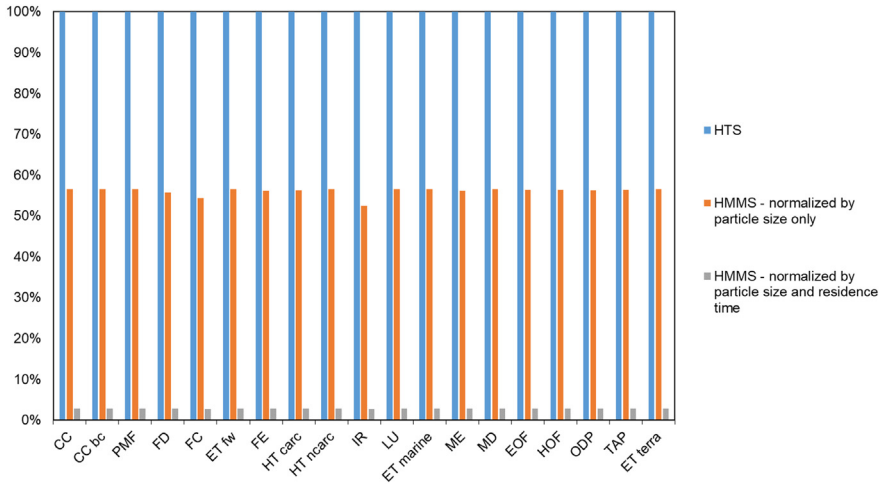


Fig. 8. LCA indicators of CAZN production stage for HTS and HMMS (considering particle size and residence time).
CC: Climate Change [kg CO2 eq.]; CC bc: Climate Change, incl biogenic carbon [kg CO2 eq.]; PMF: Fine Particulate Matter Formation [kg PM2.5 eq.]; FD: Fossil depletion [kg oil eq.]; FC: Freshwater consumption [m3]; ET fw: Freshwater ecotoxicity [kg 1,4 DB eq.]; FE: Freshwater Eutrophication [kg P eq.]; HT carc: Human toxicity, cancer [kg 1,4 DB eq.]; HT ncarc: Human toxicity, non-cancer [kg 1,4 DB eq.]; IR: Ionizing Radiation [kBq Co-60 eq. to air]; LU: Land use [Annual crop eq·y]; ET marine: Marine ecotoxicity [kg 1,4 DB eq.]; ME: Marine eutrophication [kg N eq.]; MD: Metal depletion [kg Cu eq.]; EOF: Photochemical Ozone Formation, Ecosystems [kg Nox eq.]; HOF: Photochemical Ozone Formation, Human health [kg Nox eq.]; ODP: Stratospheric Ozone Depletion [kg CFC-11 eq.]; TAP: Terrestrial Acidification Potential [kg SO2 eq.]; ET terra: Terrestrial ecotoxicity [kg 1,4 DB eq.].

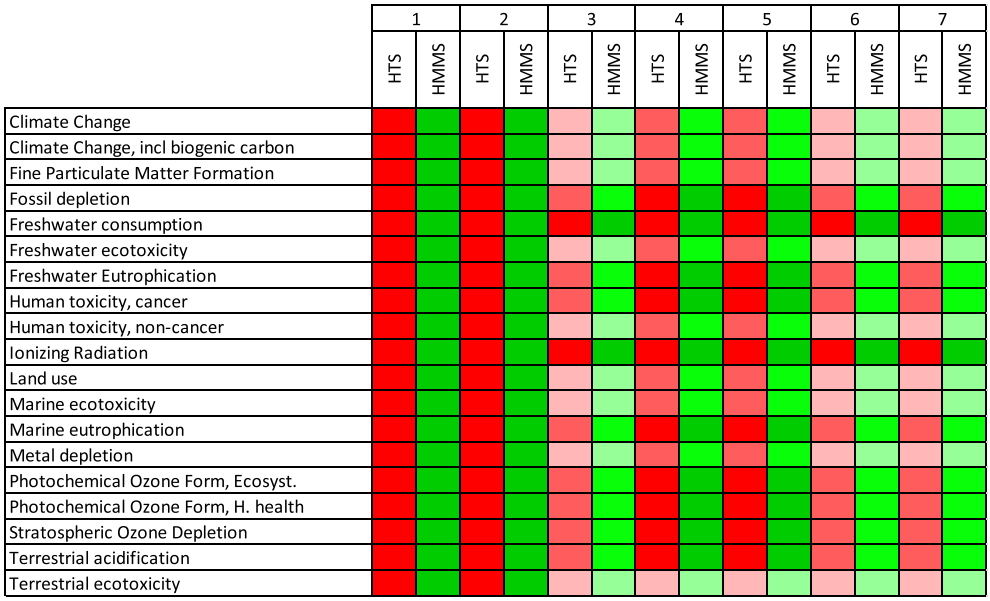


Fig. 9. Sensitivity analysis of the 7 scenarios for HMMS and HTS
HMMS columns: i) dark green is for indicators that are environmentally friendlier for HMMS not even considering surface area or time, ii) medium green is for indicators that are environmentally friendlier for HMMS when considering surface area but not even time, iii) light green is for indicators which are environmentally friendlier for HMMS only when considering both surface area and reaction time, and iv) dark red is for indicators which are less environmentally friendly even considering both surface area and time.
HTS columns: i) dark green is for indicators that are environmentally friendlier even considering both surface area and time, ii) dark red is for indicators that are less environmentally friendly not even considering surface area or time, iii) medium red means less environmentally friendly for HTS when considering surface area but not even time and iv) light red means less environmentally friendly for HTS only when considering both surface area and reaction time.

3.2.3. Transport stage

The equilibrium distance beyond which it is more advisable (from an environmental point of view) to dry the product and transport it in powder form than to transport it dissolved in liquid (in a liquid state) has been calculated to be about: 100 km if we assume drying stage powered with the European electricity mix of 2017; 56 km with the European electricity mix of 2030; and 36 km with the European electricity mix of 2050. For travel distances larger than those km, it is better to dry.

The study has been repeated for the endpoint indicators, which include all the midpoint indicators. The distances obtained are: 80.87 km for Damage to Human health and 85.88 km for Damage to Ecosystems with the European electricity mix of 2017; 45.28 km for Damage to Human health, and 48.09 km for Damage to Ecosystems with the European electricity mix of 2030; and 29.11 km for Damage to Human health and 30.91 km for Damage to Ecosystems with the European electricity mix of 2050.

The Resource availability endpoint indicator does not depend on the transport environmental impacts and, therefore, resulted better for the non-drying stage scenario at any distance.

4. Conclusions

This study works out the life cycle environmental impacts of a new synthesis for CAZN crystals, the HMMS (Caldeira et al., 2017a, 2017b; Lacoste and Thiel, 2016), and compares it with the most competitive alternative, the HTS (Wang et al., 2008).

The results show that extracting and processing the ZnO are the most relevant stages for both HMMS and HTS to shape the LCA results of CAZN productions.

HMMS presented better environmental performance for a cradle-to-gate study with a FU of 1 kg of CAZN only for some indicators (fossil depletion, freshwater consumption, freshwater eutrophication, carcinogenic human toxicity, ionizing radiation, marine eutrophication, ozone formation, ozone depletion, and terrestrial acidification) with similar results between HTS and HMMS in the rest of the indicators assessed. Even though, as reported throughout the paper, we have found that, with a FU depending only on the total mass produced in a cradle-to-gate study, we fail to consider relevant aspects of both processes, which have a direct impact on both the economic model of the system (residence time) and the performance of the final product (particle size). This is a very important point to bear in mind both in the case of this paper and in any LCA that seeks to compare different chemical processes delivering intermediate products.

HMMS environmental impacts are clearly lower when these aspects are considered. This conclusion has been validated in the main scenario, after applying the normalization factors of the FU, as well as in the 7 alternative scenarios analysed.

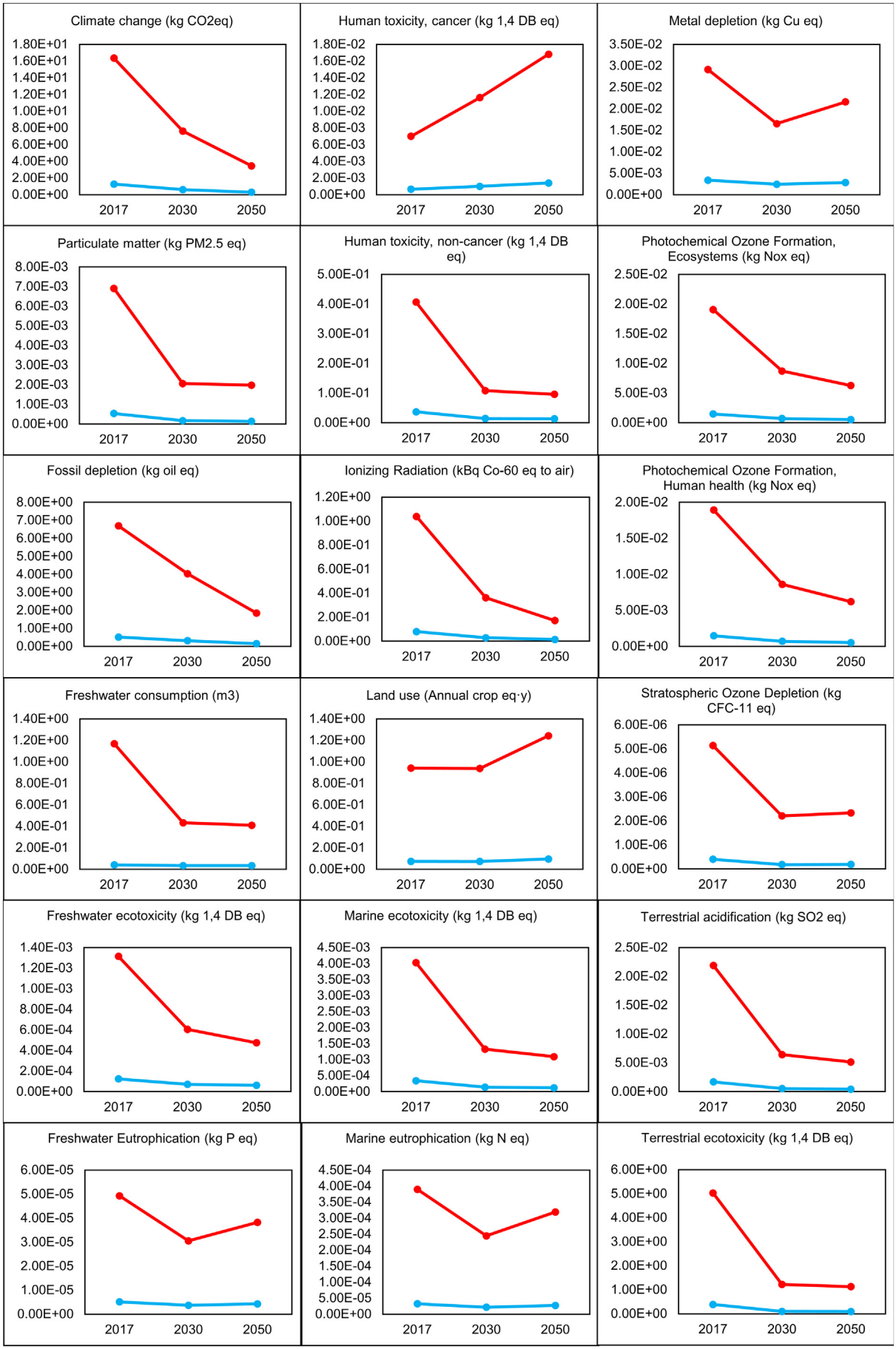
Sensitivity analysis of the electricity mix help projecting the LCA results for the European electricity mix in 2030 and 2050, for which HMMS is performing better for all the environmental impacts.

Finally, the best transport strategy for both processes consists in not drying the final product if the CAZN crystals are to be used quite close to the production facility. For longer transport distances, it is advisable to dry the crystals to decrease the environmental emissions of the transport stage.

This paper has presented a simpler, greener, and faster route to obtain CAZN crystals. The fact that it is a continuous process, foresees a possible scale-up to pilot or industrial scale. In fact, the HMMS process is currently in the process of being scaled up and optimised by the company that provided the experiments for this paper. This could revolutionise their manufacture and their application in energy storage using zinc batteries.

Based on the 6th Assessment Report by IPCC published recently (IPCC, 2022), the electrification in the world will increase, and it will be crucial to create a new and less environmentally harmful technology for energy

Fig. 10. Projection of the LCA results in 2030 and 2050 where red line represents HTS and blue line represents HMMS.


Climate change (kg CO2eq)
1.80E+01
1.60E+01
1.40E+01
1.20E+01
1.00E+01
8.00E+00
6.00E+00
4.00E+00
2.00E+00
0.00E+00
2017
2030
2050
Human toxicity, cancer (kg 1,4 DB eq)
1.80E-02
1.60E-02
1.40E-02
1.20E-02
1.00E-02
8.00E-03
6.00E-03
4.00E-03
2.00E-03
0.00E+00
2017
2030
2050
Metal depletion (kg Cu eq)
3.50E-02
3.00E-02
2.50E-02
2.00E-02
1.50E-02
1.00E-02
5.00E-03
0.00E+00
2017
2030
2050
Particulate matter (kg PM2.5 eq)
8.00E-03
7.00E-03
6.00E-03
5.00E-03
4.00E-03
3.00E-03
2.00E-03
1.00E-03
0.00E+00
2017
2030
2050
Human toxicity, non-cancer (kg 1,4 DB eq)
5.00E-01
4.00E-01
3.00E-01
2.00E-01
1.00E-01
0.00E+00
2017
2030
2050
Photochemical Ozone Formation, Ecosystems (kg Nox eq)
2.50E-02
2.00E-02
1.50E-02
1.00E-02
5.00E-03
0.00E+00
2017
2030
2050
Fossil depletion (kg oil eq)
8.00E+00
7.00E+00
6.00E+00
5.00E+00
4.00E+00
3.00E+00
2.00E+00
1.00E+00
0.00E+00
2017
2030
2050
Ionizing Radiation (kBq Co-60 eq to air)
1.20E+00
1.00E+00
8.00E-01
6.00E-01
4.00E-01
2.00E-01
0.00E+00
2017
2030
2050
Photochemical Ozone Formation, Human health (kg Nox eq)
2.00E-02
1.50E-02
1.00E-02
5.00E-03
0.00E+00
2017
2030
2050
Freshwater consumption (m3)
1.40E+00
1.20E+00
1.00E+00
8.00E-01
6.00E-01
4.00E-01
2.00E-01
0.00E+00
2017
2030
2050
Land use (Annual crop eq·y)
1.40E+00
1.20E+00
1.00E+00
8.00E-01
6.00E-01
4.00E-01
2.00E-01
0.00E+00
2017
2030
2050
Stratospheric Ozone Depletion (kg CFC-11 eq)
6.00E-06
5.00E-06
4.00E-06
3.00E-06
2.00E-06
1.00E-06
0.00E+00
2017
2030
2050
Freshwater ecotoxicity (kg 1,4 DB eq)
1.40E-03
1.20E-03
1.00E-03
8.00E-04
6.00E-04
4.00E-04
2.00E-04
0.00E+00
2017
2030
2050
Marine ecotoxicity (kg 1,4 DB eq)
4.50E-03
4.00E-03
3.50E-03
3.00E-03
2.50E-03
2.00E-03
1.50E-03
1.00E-03
5.00E-04
0.00E+00
2017
2030
2050
Terrestrial acidification (kg SO2 eq)
2.50E-02
2.00E-02
1.50E-02
1.00E-02
5.00E-03
0.00E+00
2017
2030
2050
Freshwater Eutrophication (kg P eq)
6.00E-05
5.00E-05
4.00E-05
3.00E-05
2.00E-05
1.00E-05
0.00E+00
2017
2030
2050
Marine eutrophication (kg N eq)
4.50E-04
4.00E-04
3.50E-04
3.00E-04
2.50E-04
2.00E-04
1.50E-04
1.00E-04
5.00E-05
0.00E+00
2017
2030
2050
Terrestrial ecotoxicity (kg 1,4 DB eq)
6.00E+00
5.00E+00
4.00E+00
3.00E+00
2.00E+00
1.00E+00
0.00E+00
2017
2030
2050

storage capacities. This will improve the flexibility of the electricity grids, involve more renewable energy in the electricity production system, and create the possibility for involvement of electrical vehicles. Future research by the authors will be based on the development of ex-ante LCA for production of specific energy storage products that will use the proposed technological process.

CRediT authorship contribution statement

Sergi Arfelis: Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Irene Malpartida:** Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Valentin Lair:** Conceptualization, Validation, Investigation, Resources, Writing – review & editing. **Vincent Caldeira:** Investigation, Writing – review & editing. **Ilija Sazdovski:** Validation, Writing – review & editing. **Alba Bala:** Methodology, Validation, Resources, Writing – review & editing. **Pere Fullana-i-Palmer:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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