RESEARCH AND ANALYSIS

Intensified production of zeolite A

Life cycle assessment of a continuous flow pilot plant and comparison with a conventional batch plant

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Abstract

This study investigates on the environmental impact of an intensified technology for the manufacturing of Zeolite A, one of the largest zeolites employed worldwide by volume and value. The technology under consideration is an oscillatory continuous-flow synthesis, developed industrially by Arkema, and currently at pilot-scale. Life cycle assessment (LCA) is used in this work to measure the sustainability of this emerging technology in an anticipatory fashion, before its full deployment, with the aim of driving the process development toward the minimization of the environmental footprint. The assessment explores the full life-cycle of the production system and comprises comparative analysis, scenario analysis, and a hotspot analysis. Finally, the continuous-flow technology is benchmarked against the environmental impact of a conventional batch production of zeolite A, based on a full-scale commercial plant. The results evidence that significant benefits would stem from shifting from batch to continuous-flow production. The comparative analysis reveals that the extent of the latter advantages depends on the impact category under consideration and directs the next steps of CF system's process development toward pivotal aspects such as the recirculation system to further reduce the system's environmental impacts. Regardless of the chosen production technology, a large share of the total environmental impact hinges on the production of NaOH, a building block of the synthesis, and hence is hardly mitigatable. On the whole, the findings of this work emphasize the need of prioritizing LCA during the development phase of emerging technologies and underline its efficacy to prevent waste of resources and capitals.

KEYWORDS
continuous-flow, industrial ecology, life cycle assessment (LCA), prospective assessment, process intensification, zeolites

1 | INTRODUCTION

Process intensification (PI) has experienced a ceaseless growth over the last two decades. PI is considered to be one of the most promising routes for the development of sustainable chemical processes (Van Gerven & Stankiewicz, 2009). There are several definitions of process intensification in the literature, each one capturing a different aspect of its application. Among these, however, applications and practice have contributed forging over the years a broader definition of PI as: "Any chemical engineering development that leads to substantially smaller, cleaner, safer and more energy efficient technology" (Raey et al., 2013; Stankiewicz & Moulijn, 2000).

To this end, advanced continuous-flow (CF) technologies, both at micro/milli-scale (Elvira et al., 2013; Kralisch & Kreisel, 2007; Roberge et al., 2009) and larger scale (McGlone et al., 2015; Lawton et al., 2009; Harvey et al., 2003), are considered examples of process intensification, and are often developed as alternatives to conventional batch technologies. A number of advanced CF technologies have been proposed over the past recent years as alternative, intensified solutions to the manufacturing of products in various industrial areas. The targeted systems are often those batch-type technologies that have been the conventional way of production for many years and hence are often not prone to changes. The transition to continuous processing promises improvements over the century-old batch processing technology with lower energy consumption, reduced waste generation and exposure hazards, smaller environmental footprint, lower investments, and more controllable product quality (Horizon 2020—EU Framework Programme for Research & Innovation, 2016).

In this paper, we focus on the manufacturing of zeolite Linde Type A (Zeolite LTA or Zeolite A), one of the largest zeolites employed by volume and value (Collins et al., 2020). The most common form of zeolite LTA has a Si/Al ratio of 1, which means a potential high cation exchange capacity (Xue et al., 2014). Other characteristics include thermal stability, high selectivity, nontoxicity, and good mechanical strength (Auerbach et al., 2003). This product is widely used in detergent industry for water softening (Ghadamnan et al., 2019), as well as to remove calcium and magnesium from hard water to prevent precipitation of insoluble salts on piping and equipment (Cruz et al., 2018). It is also used in industry for the dehydration of ethanol/water mixtures near the azeotropic composition (Morigami et al., 2001), which constitutes one of the most expensive steps in biofuel production (Huang et al., 2014). Moreover, zeolite LTA is widely used for dehydration of natural gas and sweetening (Shirazian & Ashrafizadeh, 2015). Finally, the use of zeolite as inorganic membranes in industrial applications and their environmental performances have been investigated in recent studies (Navajas et al., 2018).

The conventional production method is batch-type where alkaline solutions of sodium silicate and sodium aluminate are mixed together and then hydrothermally treated at around 100°C (Milton, 1953). An increasing attention is being paid to the intensification of this process, focusing on the transition to a continuous process that allows controlling the mean size and particle size distribution of crystals. Besides, a continuous installation will be more compact and more flexible, offering the possibility to increase the production capacity by duplicating the operation unit. To this end, Arkema is developing a CF installation able to match and enhance the current zeolite LTA production system. The continuous installation is characterized by the in-line mixing of the reactants and the in-line seeding of the synthesis gel solution. Moreover, a continuous oscillatory baffled reactor is integrated to carry out the hydrothermal treatment at high temperature.

The application of CF technology to the synthesis of zeolite A is an example of PI and is being investigated with increasing attention because of its interesting characteristics like product quality controllability, facility of operation, and efficient recovery of energy and valuable material streams (Ju et al., 2006). However, an intensified process is not automatically synonymous with lower environmental footprint. In fact, not all processes reach commercial scale, inevitably causing loss of capital and human resources. Additionally, technology development is often expensive, and their implementation is labor and time-intensive (Patel et al., 2012).

In light of this, emerging technologies require filtering since the first stages of their development in order to avoid waste of resources that translates into poor environmental performances, increased operational expenditure and reduce competitiveness. To this end, an efficient screening and selection of emerging technologies at early stages requires a high level of processual knowledge, and this poses several challenges regarding the availability of data and the inherent uncertainties caused by process designs yet to be completed (Bergerson et al., 2020b, 2020c). Such knowledge and data, in fact, are usually only available at later stages of their development (Grimaldi et al., 2020a).

This calls for the need of "prospective" assessments that quantifies the environmental impacts of a system since early stages of the process development, that is, on the lower end of the technology readiness level (TRL). This approach renders possible the filtration of emerging technologies from early stages. Furthermore, the outcomes of the assessment serve as input to the technology development for optimizing it toward minimal environmental impacts and maximal economic impact. Finally, the performances of the system under analysis can be benchmarked against the standard industrial practice, thus quantifying the benefits of adopting such innovative production technologies in place of conventional ones.

Over the past recent years there have been numerous attempts to define a life cycle assessment (LCA) methodology for the assessment of emerging technologies, and coin a unified term to univocally identify this approach as well capture by Guinée et al. (2018). These attempts have contributed to render even more unintelligible the identification of such approach to LCA. There exist in fact several definitions and proposed methods in literature that aim at the same objective. Ex-ante, prospective, and anticipatory LCAs (Villares et al., 2017; Wender et al., 2014; Schrijvers et al., 2014; Ravikumar et al., 2018; Cucurachi et al., 2018; Roes & Patel, 2011) share the common objective of screening of emerging technologies since the early stages of their development by assessing the impacts that the selected technology would have once fully deployed at commercial scale. In the

author's view, the most comprehensive description of LCA applied to new technology has been proposed by Cucurachi et al. (2018): "performing an environmental life cycle assessment of a new technology before it is commercially implemented in order to guide R&D decisions to make this new technology environmentally competitive with the incumbent technology mix." There exist many challenges in the path leading to fully define a universal approach to this type of assessment. The major challenges are: (1) assuring robustness of the life cycle inventory, (2) lack of practical examples and comparative analyses. Despite the increasing number of publications and general interest of the scientific community on this topic— as captured in the recent special edition of the *Journal of Industrial Ecology* (Bergerson et al., 2020a)—there still exist a general paucity in the practical application of the LCA of emerging technologies and rare are the comparisons with conventionally adopted technologies.

To this end, the field of PI offers a great number of promising technologies that call for all-round assessments able to determine in an anticipatory fashion their future environmental and economic performance. As pointed out before, the environmental impacts of these novel technologies need to be benchmarked with the state-of-the-art technologies, as in the case of identifying the possible associated benefits of intensified continuous flow technologies over batch systems. In response to this, a series of papers has been published on the assessment of novel production systems originating from both academic (Grimaldi et al., 2020b) and industrial R&D (Grimaldi et al., 2020a) at TRLs 3–4 (bench-scale and mini-pilot scale).

Following up on the application of LCA of emerging technologies, in this work, the LCA methodology is applied to the intensified continuous flow production of zeolite A at full industrial pilot scale (TRL 5). The aim is to provide a projection of the environmental performances of this novel production technology, benchmark it against the standard production system conventionally adopted in industrial production, and hence provide an understanding of the consequences of its full exploitation at large scale.

To the best of the authors' knowledge, no LCA on the CF production of zeolite A at large-scale has been published to date. In this work, the synthesis of zeolite A is subjected to assessment: two production systems are analyzed, the conventional batch synthesis and an innovative CF synthesis. The batch system is sized on an existing commercial plant and it is used as the reference system, while the CF system refers to a full pilot plant that is being developed by Arkema, at GRL facilities in Lacq. Both systems are subjected to LCA and are compared.

2 | MATERIAL AND METHODS

2.1 | Framework

The adopted framework (Figure 1) operationalizes the LCA as a tool for: (1) integrating the technology development with metrics measuring environmental impacts, in order to maximize environmental performances of the emerging technology under analysis; (2) helping the decision making process on the deployment of the emerging technology, by benchmarking the projected performances of the latter against conventional production systems at commercial scale.

In order to enable these two functions, the conventional production system is assessed in a retrospective fashion, that is, following the procedure of a conventional LCA, by assessing the commercial scale system that is already available in the market; the output of the assessment are then used as reference lines on which the environmental performances of the emerging technology are evaluated. On the other end, the emerging technology is assessed in a prospective fashion, providing a projection of the future environmental performances of the system before the latter is deployed. As mentioned before, one of the inherent objectives of the LCA on emerging technologies is in fact to benchmark the future environmental performance of novel technologies against conventional technologies, thus acquiring information on the further development steps of the latter technologies and guide the research and development.

To this end, the results of the comparison are used as inputs to the process development. The information generated from the comparative analysis comprises quantitative and qualitative information of environmental performances of the emerging technology with respect to the conventional system.

In summary, the framework can be subdivided into three main phases (as shown in Figure 1): system definition, assessment, comparative analysis on the basis of the framework presented in previous works (Grimaldi et al., 2020b). In the next sections these phases will be analyzed in detail, touching on the methodological recommendations as well as on the procedures followed for the calculation of quantitative data.

- **System definition**

In this step, the process flow diagrams (PFD) are defined and described in detail for both the emerging and conventional technologies. Specifically, the batch system is sized on a commercial plant with a reference production of 1 kton of zeolite A per year. This scale is used as the baseline reference for the CF system. The functional unit refers to an identical zeolite A product; in fact, the CF manufacturing route generates a product of the same quality as the batch system with the advantage of allowing more control over the particle-size distribution of zeolite A. In the case of the CF system, the construction of the PFD is originated from a full pilot plant and scaled out to the reference productivity of the batch system (1 kton of zeolite A per year). The CF reactor used in the current pilot plant setup served as basis for the scale out, and the final PFD design included a set of the latter reactor operating in parallel. As a result, the original pilot plant design was conserved, and the definition of the
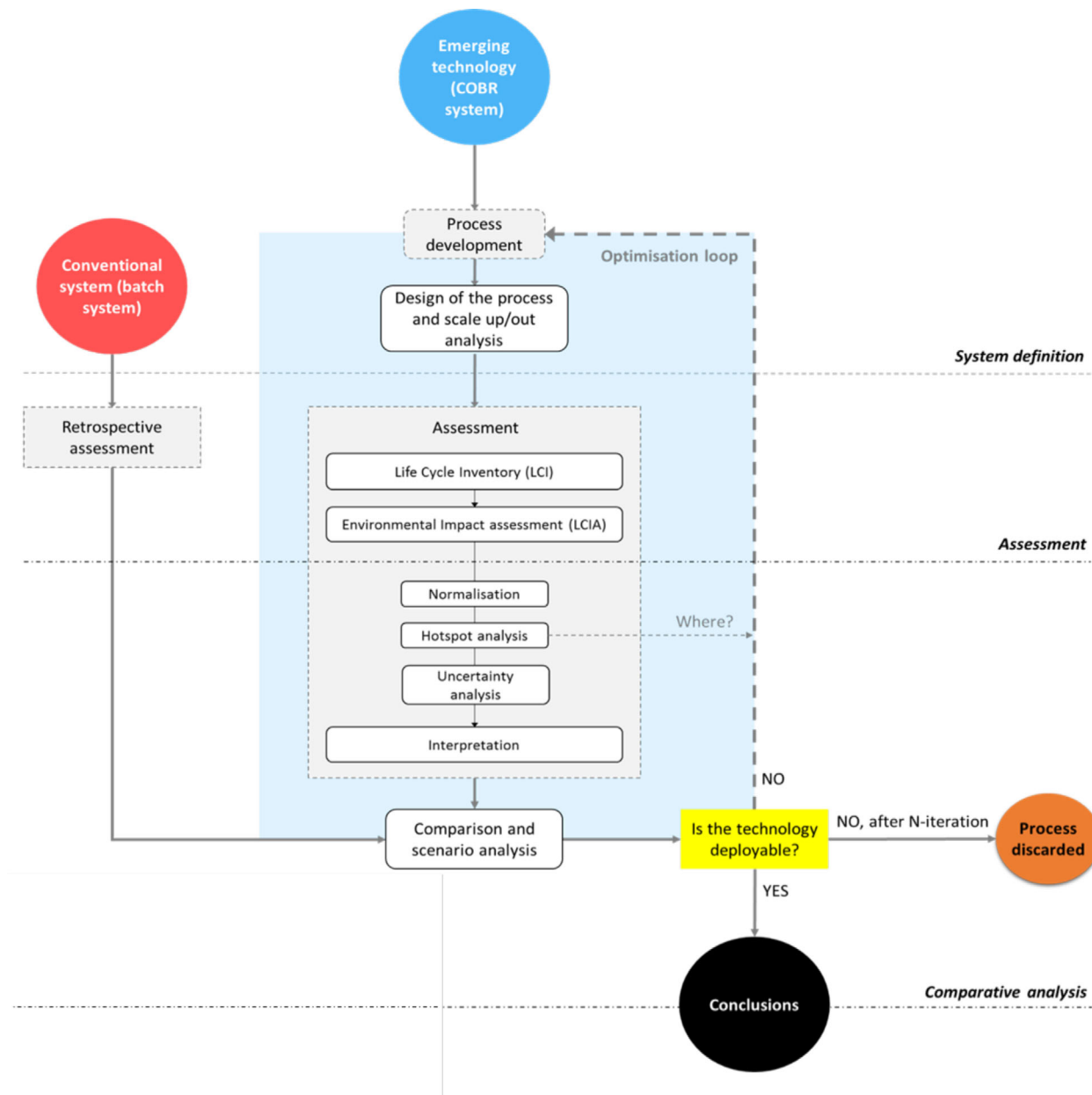
FIGURE 1   Framework: system definition, assessment, and comparative analysis

final PFD of the CF production system did not involve scale up assumptions that would have added uncertainties into the Life Cycle Inventory (LCI).

For both systems, the data necessary to compile the LCI were collected in the Arkema facilities of GRL in Lacq, France. A high level of detail is provided in this phase with specific regard to the emerging technology; as remarked in the previous section the robustness of the data inventory is crucial for enabling a coherent assessment. To this end, the collection of data was undertaken directly on-site by means of a secondment period spent in the Arkema's facilities and through extensive collaboration with technology developers, in order to grant reliable data, quantify the uncertainties and gain general knowledge on the process, necessary for the compilation of the inventorial information.

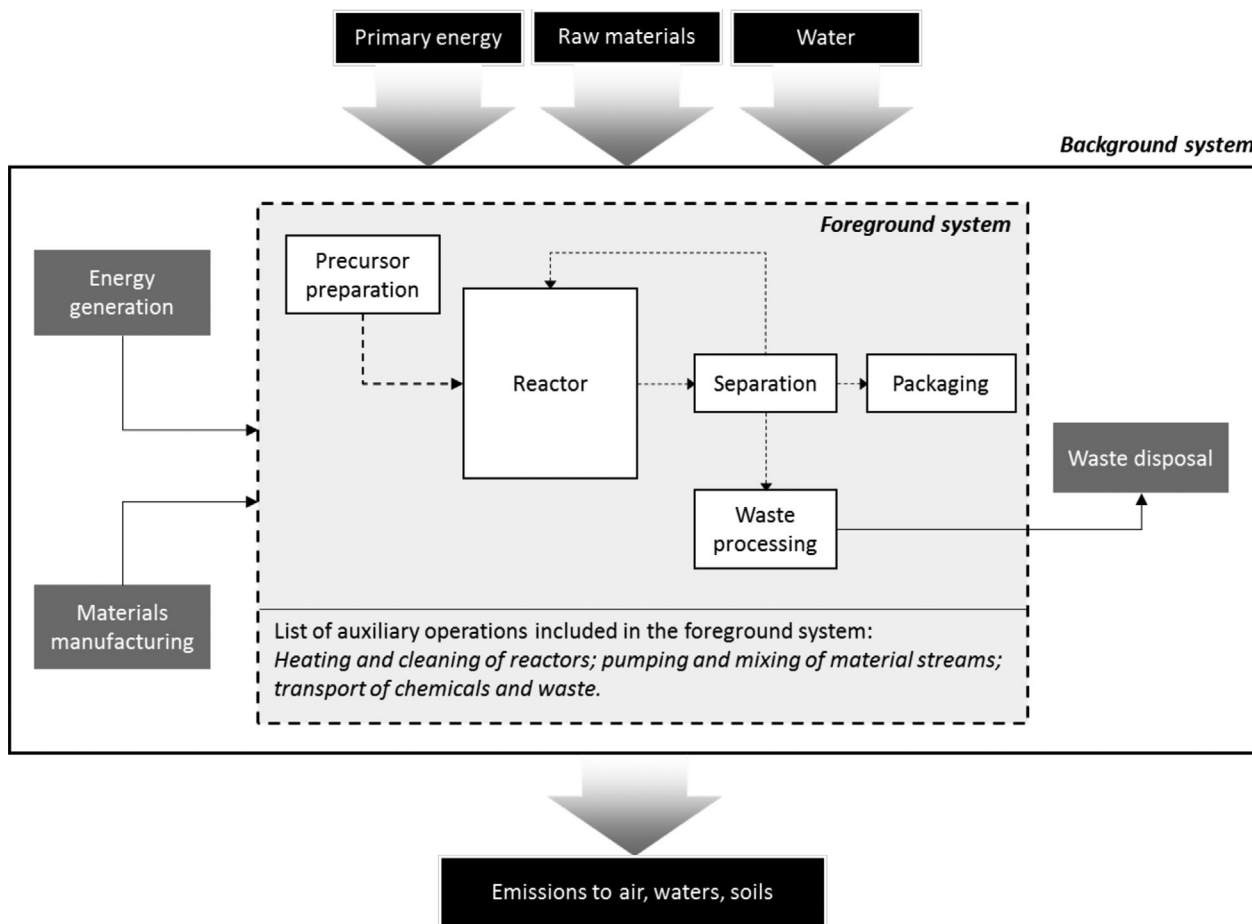The resulting information is organized in the LCI.

FIGURE 2 System boundaries: foreground and background systems

- **Assessment phase**

Life Cycle Inventory

This phase quantifies inputs and outputs of the product systems that are relevant to the assessment. This is achieved prevalently through data collection and integrated with calculations procedures (in accordance with the ISO 14040) (International Organization for Standardization, 2016). The Functional Unit (FU) for this study is 1 kton of zeolite A. This FU is used for the batch and CF production systems as they have the same productivity.

The system boundaries are reported in Figure 2. These consist of two sub-systems: background and foreground systems. The latter includes the set of activities that take part directly to the production: precursor preparation, pumping, heating, mixing, separation, cleaning, packaging, transport, and waste disposal. The background system comprises the activities that support indirectly the synthesis such as extraction of raw materials, chemical production, electricity production, and waste treatments.

The same FU and system boundaries are used for both CF and batch system, in order to assure a coherent comparison.

Environmental impact assessment

The environmental impacts are calculated through the life cycle impact assessment (LCIA) method "ILCD/PEF recommendation 1.09" (Hauschild et al., 2011). This method has been proven to be reliable, is recent, and has received large consensus in the last few years. It uses midpoint indicators: in other words, the quantitative modelling of the environmental consequences of an emission stops at a mid-point in the cause-effect chain. This contains the uncertainties during the calculation of the environmental impacts as less assumptions are made on the effects on the environment. The LCIA associates an impact on the environment to each one of the flows that have been identified in the LCI. The environmental impact is expressed for various impact categories: these cover different aspects of the environmental consequences of a given emission. The selected impact categories for this study are reported in Table 1.

The software adopted for the built of the LCA model is GaBi ts 8.7 (SP36). Processes and materials used in the model comes from the most recent version of the databases "Professional + extensions (II, VI, IX, XVII)" and "Ecoinvent 3.6" in compliance with the ISO standards. All the results reported in this work refers to 1 kton of zeolite A product, which is the chosen FU.

TABLE 1 Impact categories used for the LCIA and related abbreviations

Impact category	Units	Abbreviations
Acidification	[mole H$^+$ eq.]	A
Climate change, excluding biogenic carbon	[kg CO$_2$ eq.]	CC
Ecotoxicity freshwater midpoint	[CTUe]	EcoTOX
Eutrophication freshwater midpoint	[kg P eq.]	E fw
Eutrophication marine midpoint	[kg N eq.]	E mw
Eutrophication terrestrial midpoint	[mole N eq.]	E t
Human toxicity midpoint, cancer effects	[CTUh]	HT c
Human toxicity midpoint, noncancer effects	[CTUh]	HT nonc
Ozone depletion	[kg CFC-11 eq.]	OD
Particulate matter	[kg PM2.5 eq.]	PM
Photochemical ozone formation midpoint	[kg NMVOC eq.]	POF
Resource depletion, water	[m^3]	RD water
Resource depletion, minerals, fossils and renewables	[kg Sb eq.]	RD m, f, ren

Note: The impact categories are selected from the LCIA method "ILCD/PEF recommendation 1.09."

- **Comparative analysis**

In this phase the results of the assessment of the batch and CF systems are further analyzed and then compared. The comparative analysis is integrated with an uncertainty analyses and a scenario analysis that investigates on the environmental performances of the batch and CF systems under different efficiencies of the recirculation system in order to test the sensitivity of the system, understand under which condition the CF system would bring advantages over conventional systems, and hence be deployed.

In the comparison, an uncertainty analysis quantifies the uncertainties that affect the system under consideration; these are mainly attributable to the variability of the data collected and to the assumption made for the calculation of the data not directly available for the inventory. Consequently, the results of the LCIA are affected by errors that arise from such uncertainties and that need to be quantified. With regard to the LCIA, the environmental impact is expressed for a number of impact categories (see Table 1). The environmental impact is composed by the contribution of various activities occurring in the life cycle (electricity generation, NaOH production, etc.) that are organized in groups (chemicals, cleaning, energy consumptions, etc.) according to their function. Every activity is linked to a certain group and contributes to the overall environmental impact of that specific group. For instance, the impact of the group "Chemicals" is obtained by summing the environmental impacts of all the activities of the group "Chemicals," which are: alumina trihydrate production, sodium silicate production, NaOH production, and water use. These groups are assigned a level of uncertainty (ε_j, with j being a given group) on the basis of the variability of the data collected and on the assumptions made for the calculation of nonavailable data. The uncertainty is then propagated (ε_{propj}) by taking into account the reciprocal interactions that occur among the groups of the inventory. Each one of these groups (chemicals, cleaning, energy consumptions, etc.) contributes to the overall environmental impact of the production system in a different way for each impact category. The sum of the impacts of each group in a specific impact category is hence equal to the overall impact of the whole production system in that impact category. Therefore an uncertainty error is calculated for each impact category; this uncertainty error is called $\varepsilon_{LCIA, i}$, with i being a given impact category. $\varepsilon_{LCIA, i}$ is calculated as the weighted sum of the propagated uncertainty error ($\varepsilon_{prop\, j}$), with the weight being the % impact contribution of that specific group j in the impact category i considered:

$$\varepsilon_{LCIA, i} = \sum_j \left(\varepsilon_{prop\, j} * X_{i,j} \right)$$

where i = impact category and j = group of the inventory and $X_{i,j} = $ *impact contribution*.

The results of the analysis are embedded in the charts reported in the "Results and discussion" in the form of error bars; further information on the uncertainty analyses is reported in Supporting Information S1.

In addition to the uncertainty analysis, a hotspot analysis is performed to generate the highest level of detail. This investigates on the main activities responsible for the environmental impacts in the life cycle of the two systems. The hotspots analysis provides information on the possible intervention points in the technology development: both in terms of life-cycle phases (e.g., transport, production phase, waste treatment) and in terms of specific activities taking part to the life cycle of the production system (e.g., preparation of precursor, pumping, and hazardous waste incineration). The underlying concept is to enable an "optimization loop" (as shown in Figure 1) with processual information and projected environmental impacts

travelling back and forth between process development and assessment phase, thus contributing to maximizing the environmental performances of the emerging technology before the latter enters the market.

The outputs of these analyses are compared and discussed for both the systems under analysis.

2.2 | System definition

The productive system taken as reference for this study is a batch commercial plant capable of producing around 1 kton of Zeolite LTA (dry weight) per year. The synthesis of zeolite LTA involves the hydrothermal crystallization of aluminosilicate gels (formed upon mixing between alkaline aluminate and silicate solutions). The precursors, sodium aluminate and sodium silicate, are mixed in a batch reactor at around 35°C to form the synthesis gel that is then heated to be crystallized at 100°C and a zeolite slurry is formed. A buffer tank regulates the flow of the zeolite slurry downstream of the reaction step.

Subsequently, the resulting flow is processed into the separation step that is composed by a set of operative units treating the slurry and refining it to obtain the final product. Specifically, the slurry is firstly adjusted in terms of water content by means of filtration where a zeolite cake is obtained. The cake is then dried, obtaining zeolite powder. Afterwards, the resulting powder is treated via grinding and the final product is collected. A simplified representation of the PFD is reported in Figure 3, which shows the process configuration adopted in the batch system as well as in the continuous flow synthesis.

In the flow setup (top section of Figure 3), a set of parallel continuous oscillatory baffled reactors (COBRs) replaces the vessel that served the purpose of the synthesis gel preparation and crystallization in the batch system. These operations occur in one step in the CF reactor. The latter differs also from the batch reactor for the in-line mixing of the reagents and the in-line seeding of the synthesis gel. The concentration of both sodium aluminate and sodium silicate is also lowered in order to grant the right viscosity of the mixture entering the reaction phase. The content of sodium hydroxide is increased accordingly to keep unvaried the alkalinity. The resulting stream is then sent to the COBRs where the growth of crystals is achieved. Precursor preparation and separation phases remain unaltered as the CF system is meant to be integrated into the existing commercial plant.

A recirculation system is currently being tested for both systems and aims at recovering part of the sodium hydroxide from the effluent after the filtration step of the separation ("Mother liquid"). This, in fact, is rich in sodium hydroxide that is required in large volumes during the synthesis. Despite not being fully assessed yet, the recirculation system is considered part of both systems in this study. The resulting variability that such addition brings to the LCI is taken into account by means of a scenario analysis, presented in the next sections, considering different recirculation efficiencies. Specifically, the selected recycling efficiencies are 0% and 30% for the batch system, and 80% and 95% for the CF system; these efficiencies reflect the different technical feasibility of adopting a recycling system in the two systems, and represent the most likely scenarios.

A breakdown of the material and energy requirement related to the two systems described above is reported in Supporting Information S1.

3 | RESULTS AND DISCUSSION

The results of the LCA are organized in two sections, technology comparison and hotspot analysis. Each section presents a different angle on the environmental performance of the zeolite A production.

3.1 | Technology comparison

The comparative analysis of the environmental impacts of batch and CF production (referred to the chosen FU of 1 kton of zeolite A) is shown in Figure 4.

In Figure 4, four scenarios are considered, each one referring to a different recycling ratio of the "mother liquid" (see "System definition"). The results indicate that the environmental impact of these systems is influenced by the recycling ratio. Specifically, lower impacts are achieved with increasing recycling ratios. The recycled stream, "mother liquid," contains large volumes of NaOH that is employed in large quantity in the synthesis. As a result of the recirculation, a lower amount of waste is produced and the use of fresh NaOH is reduced. The latter is used abundantly in the CF system and hence the recirculation system helps mitigating the environmental impacts arising from its production.

As expected, the adoption of a recirculation system in the batch setup contributes to lower the environmental impact across all the impact categories of the study. When shifting from batch to CF production technology, Figure 4 delineates that the environmental advantages stemming from the CF system depends on the considered impact category. The change of environmental impact with respect to the batch system appears in fact to span from large savings in impact categories as eutrophication of freshwater to decreased environmental performances in resource depletion of water. To this end, Table 2 quantifies such differences of environmental performances of the scenarios scrutinized across the different impact categories.
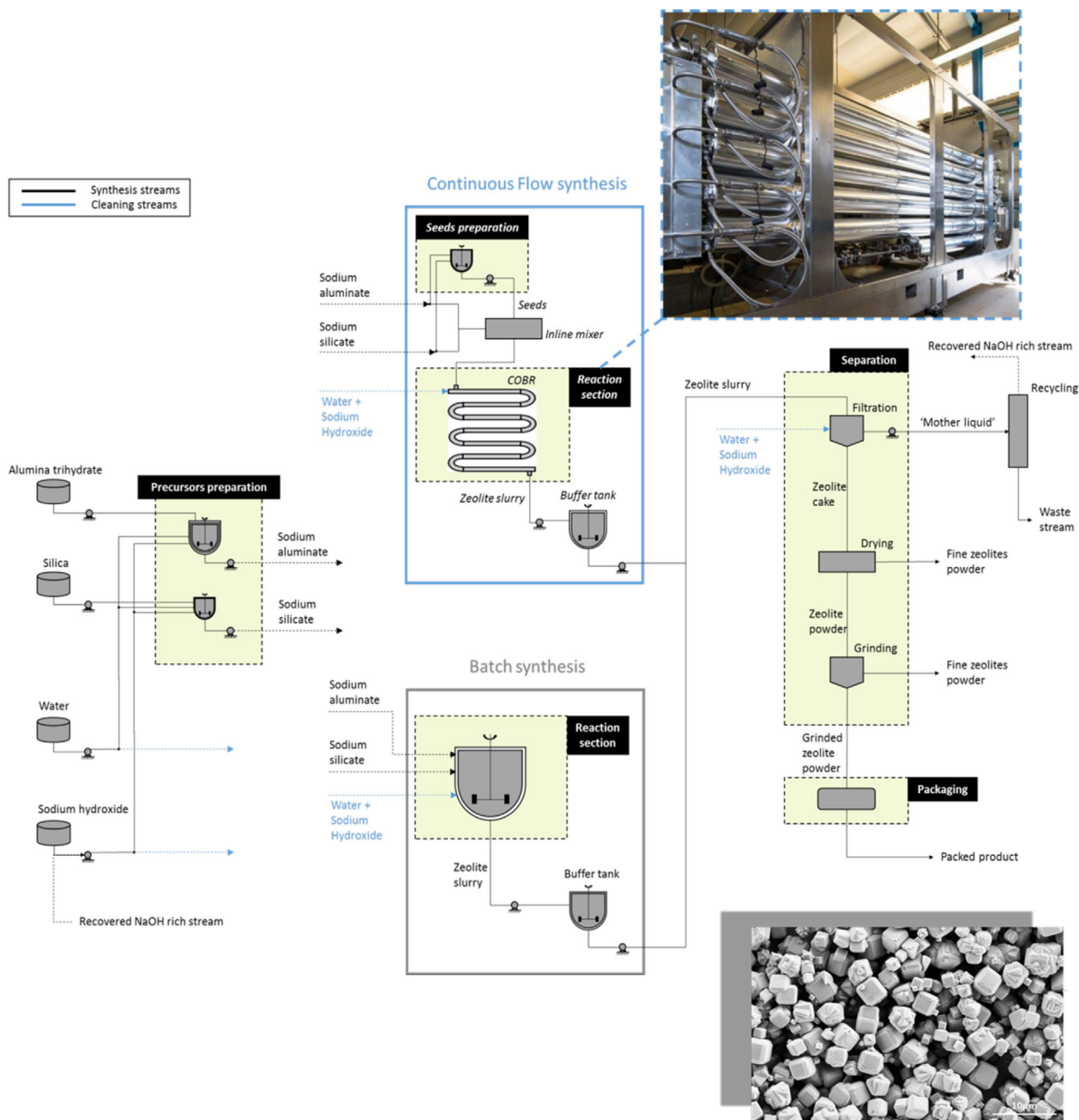
FIGURE 3 Simplified PFD, batch and continuous flow production. The continuous oscillatory baffled reactor (COBR) used at GRL facilities in Lacq is shown in the top-right corner. In the bottom-right corner, Zeolite LTA crystals: scanning electron microscopy image of a sample obtained from the pilot plant

Table 2 reports the percentage changes of the impact of the scenarios analyzed in Figure 4 with reference to the batch system (no recycling) for each impact category of the study. In Human Toxicity (cancer and noncancer effects), Eutrophication of freshwater, Ecotoxicity of freshwater, and Ozone Depletion, the CF production has lower environmental impacts than the batch production. The savings of environmental impact become more robust as the recycling efficiency of the CF system increases. Specifically, when the recycling rate is 95%, the environmental impact of the batch systems is significantly reduced by −48%, −32%, and −22% in E fw, HT non-c, and OD impact categories, respectively. In the rest of the impact categories of the analysis, the CF scenario at 95% of recycling efficiency shows comparable impacts with the reference batch system, with the exception of Resource depletion of water and Resource depletion of minerals, fossil and renewable in which the resulting impact is respectively 3.7 and 1.1 times the reference system's impact.

The comparative analysis of Figure 4 included the result of the uncertainty analysis as introduced in the "Materials and methods" section. Compared to the batch life cycle, larger uncertainties are associated to the CF life cycle, which arise from the lower technological readiness level of
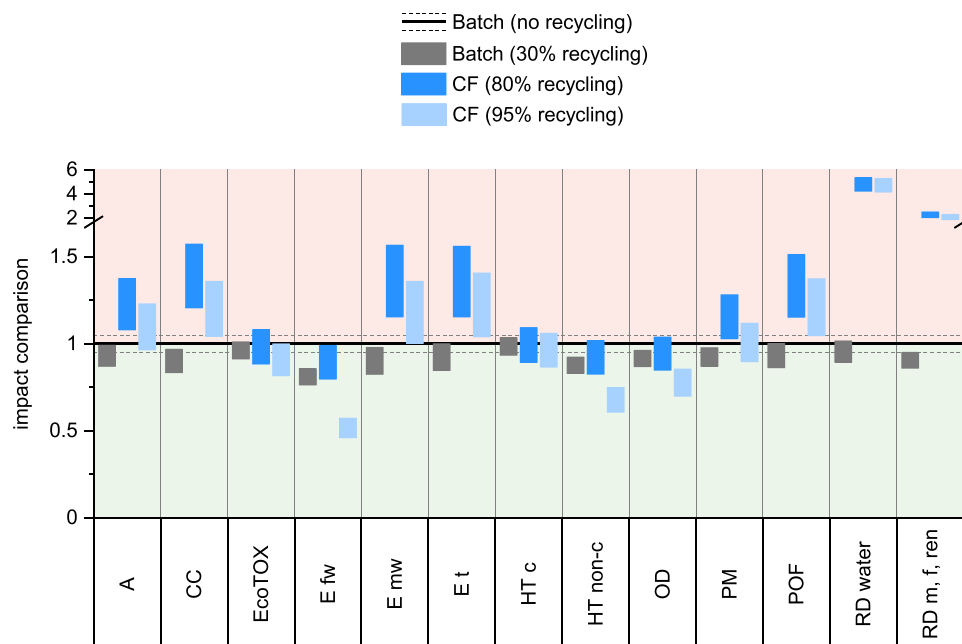
FIGURE 4 Comparison of the environmental performances of the batch and continuous flow systems under variable recycling ratios. Note that the reported impacts are normalized internally on the basis of the conventional batch production system with no recycling. Data underlying this table are available in Supporting Information S2

TABLE 2 Environmental impact of batch and CF production systems under variable recycling ratios

Impact category	Batch (No recycling)				Scenarios (%change on batch no recycling)		
					Batch (30% recycling)	CF (80% recycling)	CF (95% recycling)
A	1.3	x	10^4	*[Mole of H+ eq.]*	-7%	23%	10%
CC	2.0	x	10^6	*[kg CO2 eq.]*	-10%	39%	20%
EcoTOX	3.6	x	10^7	*[CTUe]*	-4%	-2%	-9%
E fw	0.8	x	10^3	*[kg P eq.]*	-19%	-11%	-48%
E mw	2.5	x	10^3	*[kg N eq.]*	-10%	36%	18%
E t	2.3	x	10^4	*[Mole of N eq.]*	-8%	36%	22%
HT c	0.9	x	1	*[CTUh]*	-1%	-1%	-4%
HT non-c	1.0	x	1	*[CTUh]*	-12%	-8%	-32%
OD	1.0	x	10^{-2}	*[kg CFC-11 eq.]*	-8%	-6%	-22%
PM	0.9	x	10^2	*[kg PM2.5 eq.]*	-8%	16%	1%
POF	6.2	x	10^3	*[kg NMVOC eq.]*	-7%	33%	21%
RD water	0.6	x	10^4	*[m³ eq.]*	-4%	379%	371%
RD m, f, ren	3.2	x	10	*[kg Sb eq.]*	-9%	130%	111%

Note: The color code in the heat map is green = lower impact, red = higher impact.

the latter system. To this end, the confidence interval relative to environmental impact of the CF system does not exceed ± 15% whilst being for the ± 9% batch system. The uncertainty analysis depicts that is possible to reach detailed conclusions though the comparative analysis notwith-standing the CF system's early stage of development. To this end, it is worth remarking the importance of direct data collection—in this case under-taken through on-site surveys over a prolonged period spent in the pilot plant location. The latter approach allowed to contain the uncertainties associated to the various categories of the LCI thanks to on-site data collection, repetition of experimental runs, and direct collaboration with
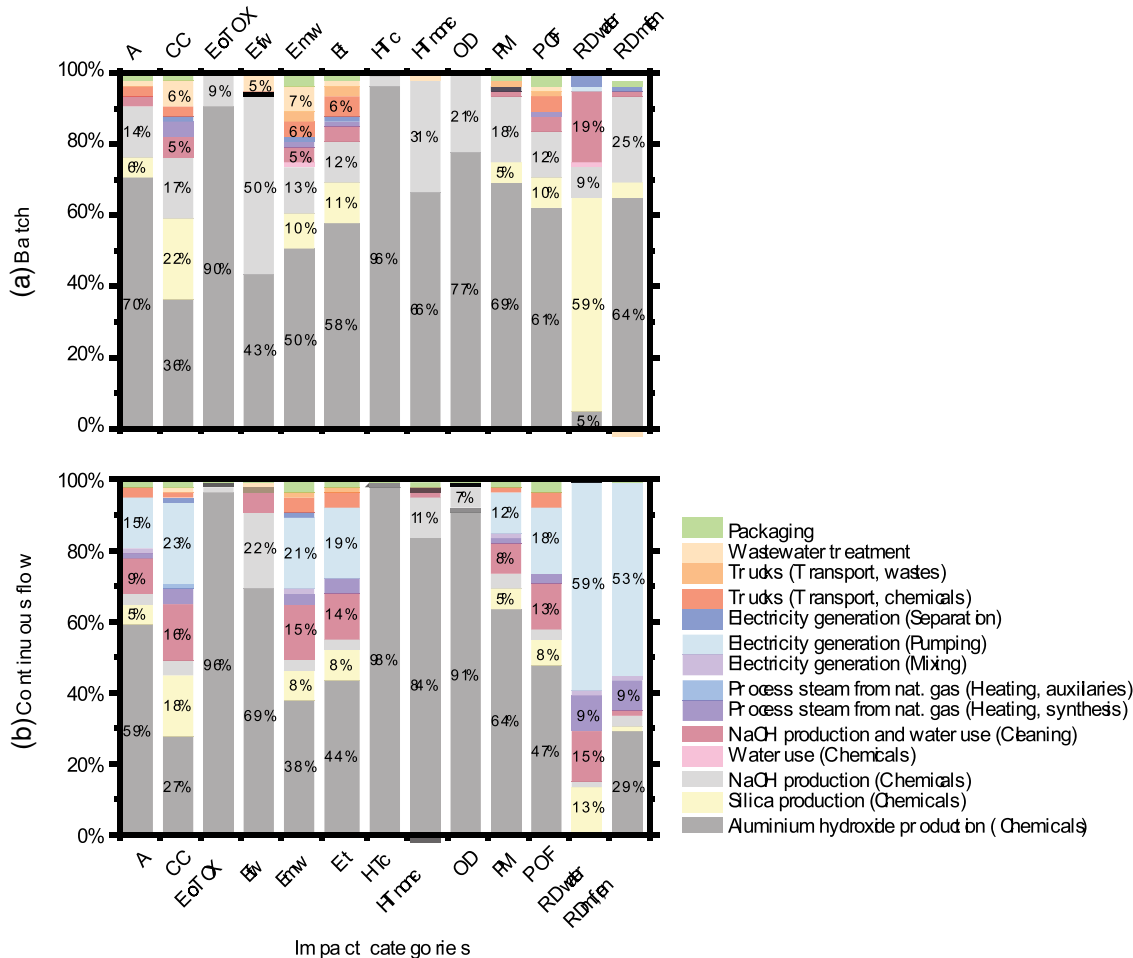
FIGURE 5 Hotspot analysis for: (a) Batch production (30% recycling), (b) Continuous flow production (95% recycling). Data underlying this table are available in Supporting Information S2

technology developers—pivotal aspects when assessing emerging technologies. Even though the level of uncertainties associated to some inventorial group are expected to decrease as the technology development advances—hence allowing more accurate analyses—it is however crucial to perform such early stage assessments in order to guide the process development toward the maximization of the environmental performances of the innovative system. This is only possible with timely analyses, able to identify intervention points and opportunity for optimization across the novel production system and correct the trajectory of the process development in time, without resorting on tardive remediation measures that implicates higher costs and waste of resources.

An in-depth explanation of the latter results of the comparative analysis is presented in the next paragraph that reports the causes behind the environmental impact—and hence the change of the latter—of the batch and CF system in each impact category through a hotspot analysis.

3.2 | Hotspot analysis

The comparative analysis has identified radical differences in the environmental performances of the batch and CF production systems. This section looks into the causes of these differences by means of a hotspot analysis. The hotspot analysis investigates the activities that constitute the life cycle of the selected production technologies and identifies those activities that contribute the most to the impacts. For simplicity, the role that each activity covered in the production of zeolite A is reported after the activity's name, in brackets (e.g., "NaOH production (Cleaning)"). The results of the hotspot analysis are reported in Figure 5.

The production of the chemicals involved in the synthesis of zeolite A take most of the share of the impact of both production systems. Specifically, the production of aluminum hydroxide is the highest contributor compared to the rest of the chemicals, which are mainly silica and sodium hydroxide.

With regard to the production process of aluminum hydroxide $Al(OH)_3$, practically all the $Al(OH)_3$ employed in commercial use is manufactured via the Bayer process (Hind et al., 1999) that entails the digestion of bauxite in a caustic solution of NaOH at high temperatures. The resulting waste is separated and $Al(OH)_3$ is precipitated from the remaining solution. The insoluble bauxite residue—called "Red mud"—is predominantly iron oxide and it is highly caustic because of the residual NaOH (Lee et al., 2009; Paramguru et al., 2004). Red mud has a high environmental impact, with primary regard to Ecotoxicity of freshwater and human toxicity (Figure 5), and has caused several problems in the past as it was stored in lagoons (BBC, 2010; Ruyters et al., 2011). The waste was proven to be useful in ceramic production and in building construction. However, the reuse of this waste is still strongly debated in Europe (Balomenos, 2018). The United States does not approve it due to the dangers it risks to the environment: significant levels of chromium and arsenic were discovered by EPA in red mud samples (EPA). Therefore, the waste is usually disposed in artificial enclosed reservoir.

With regard to other chemicals that are involved in the synthesis of zeolite A, NaOH is the other chemical that contributes significantly to the overall impact. NaOH is directly involved in the synthesis and also it is used as a cleaning agent. From Figure 5a it emerges as a substantial contributor to the overall environmental impact, especially of the batch system. In the CF system, in fact, the weight of its impact over the total impact of the production system is reduced. This is primarily due to the recirculation system, which is more effective in the CF compared to the batch system: this allows the recycling of larger volume of NaOH and consequently reduces the demand of fresh NaOH.

NaOH is also the main source of waste of both production systems. Downstream of the reactor, the synthesis liquid, which is composed predominantly by zeolite A crystals and NaOH, is processed into the separation units. The crystals are recovered and separated from the synthesis liquid, and the resulting stream ("mother liquid," composed mainly by NaOH) is consequently disposed. This occurs via truck transport to waste treatment facilities that processes the mother liquid by means of dilution, neutralization, and discharge into lagoons. Evidently, the resulting environmental impact is directly linked to the volume of NaOH present in the waste stream. In fact, the impact of the waste treatment system is detectable in Figure 5a for the batch system, whilst being negligible in Figure 5b for the CF system, thanks to a better integrated recycling system as explained before.

Finally, the consumption of electricity plays a marginal role in the environmental impact of both production systems. In the CF, however, the resulting impact associated to the production of electricity is slightly higher as a consequence of a pumping system that is more energy demanding. To this end, it is worth remembering that the CF technology hinges on an oscillatory baffled reactor that is powered by a set of pumps controlling the oscillation intensity (frequency and amplitude); thus, the additional energy demand.

Batch and continuous flow systems came out from the hotspot analysis as having radically different impacts on the environment, with the main differences being in the processing of NaOH and the waste disposal. As a consequence of this, the CF system emerges as a more efficient system. It is worth noting, however, that the majority of the environmental impact of the life cycle of zeolite A is attributable to the production of aluminum hydroxide, a factor in common between both the CF and batch production system. The latter takes up more than 50% of the environmental impact across the different the impact categories. Aluminum hydroxide is a building block of the synthesis of zeolite A and the environmental impact of its production is a subject of debate as pointed out before. However, its impact can hardly be lowered by the choice of the production technology and hence, the overall reduction of environmental impact shown by the CF system results contained.

4 | CONCLUSIONS

We performed a life cycle assessment of the production of zeolite A. Two production technologies were subjected to assessment and compared: a conventional batch production, which served as reference for the comparison, and an intensified production technology based on continuous flow. The latter is a novel technology that is currently being developed in an industrial R&D environment and was scaled out conceptually to full scale, starting from an existing pilot plant. The modelling of the life cycle of the two production systems included the production phase as well as all the activities at support of it, such as electricity production, extraction of raw material, and waste treatment.

The LCA highlights the importance of adopting a low impacting production system for the manufacturing of zeolite A. This product is, in fact, massively produced worldwide and hence any reduction of the environmental impact of its production would have tangible effects at large scale. The results of the assessment indicate that a number of advantages would stem from adopting the continuous-flow technology instead of the conventional batch technology, depending on the impact category. The comparative analysis underlines the benefits of the CF system over the batch system in impact categories such as in Human Toxicity (cancer and noncancer effects), Eutrophication of freshwater, Ecotoxicity of freshwater and Ozone Depletion, whilst showing increased impact in Resource depletion of water and Resource depletion of minerals, fossil and renewables. The assessment suggests that relying on an efficient recirculation system is pivotal for maximizing the environmental performances of the production system and hence the next steps of the process development should focus on this aspect. Furthermore, the LCA investigated on the main contributors to the environmental impact of the two systems under consideration through a hotspot analysis. Specifically, the primary advantages that allowed enhanced environmental performances of the CF system in the impact categories identified in the comparative analysis were found to be a lower demand of fresh NaOH and lower volumes of waste produced, which reflects in a lighter transport system and milder waste treatment.

The uncertainty analysis emphasized the capability of the LCA of reaching practical conclusions notwithstanding the CF system's early stage of development. To this end, direct data collection, on-site surveys, and close collaboration with technology developers emerged as fundamental requirements to contain the uncertainties associated to the various categories of the LCI, and therefore allowing the operationalization of the findings of the study.

Finally, it is worth underlining that the environmental advantages that have been identified through the assessment of a single production plant of zeolite A would acquire even more robustness in the case that current and future production facilities in Europe adopted the continuous-flow technology to replace or integrate the conventional batch technology. However, the LCA suggests that there is a limit to the enhancement that can be made to the current production system: this is represented by the synthesis method itself, in other words the chemicals involved, especially aluminum hydroxide, whose production is responsible for a large portion of the environmental impact of the production system.

With the present study the authors want to bring the attention to the importance of prospective evaluations as tools for understanding the future consequences of emerging technologies at the early stages of their development. In this context, process intensification emerges as a powerful way to address and reduce the environmental impact of a process, by enhancing the controllability of the process and containing the generation of wastes. The case of continuous-flow technology applied to the production of zeolite A offers compelling insights about the possibility of a general transition to intensified production technologies, starting from those established processes that still hinge on conventional batch production and that have room for improvement but have not yet undergone a process intensification study. To this end, we envisage the application of LCA at early stages to other novel industrial production systems with the aim of expanding the currently available data on the environmental performances of emerging technologies, demonstrate the feasibility of performing these type of assessment and quantifying the associated uncertainties; the overall objective is to incentivize and facilitate the integration of environmental consideration during the process development of a technology, hence maximizing its performances.

CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABLE ON REQUEST FROM THE AUTHORS

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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