

Can the use of captured CO₂ lower the environmental impacts of formate production?

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ABSTRACT

The majority of bulk chemicals (e.g. olefins and alcohols) are organic compounds that are almost exclusively produced from fossil feedstocks such as natural gas. Utilisation of carbon dioxide captured from anthropogenic sources, which are both inexpensive and abundantly available, represents an alternative pathway that is drawing increasing attention, mainly for its potential to decreasing emissions of greenhouse gases and resource depletion of chemicals production. Notably, carbon utilisation does not represent an approach to CO₂ mitigation because it only delays its emissions rather than removing it over a long timescale; hence, the relevant question that we aim to address is: “Can captured CO₂ be used as feedstock to reduce the environmental impacts of chemicals’ production?”. As a case study, this work focuses on the production of formate and presents a prospective comparative life cycle assessment (LCA) between the conventional fossil-based pathway and an innovative, CO₂-based process, that involves the electro-catalytic reduction of CO₂ using an ionic liquid as solvent. CO₂ is assumed to originate from a natural gas-fired power plant and captured after combustion, through a conventional monoethanolamine absorption system. Ionic liquids are used to enhance the reduction of CO₂ and its conversion to formate. The study adopts a cradle-to-gate perspective and analyses multiple impact categories including, but not limited to, global warming and resources depletion.

1. INTRODUCTION

The majority of bulk chemicals such as olefins and alcohols are organic compounds that are almost exclusively produced from fossil feedstocks such as oil and natural gas (Centi et al. 2013). CO₂ captured from anthropogenic sources represents an alternative feedstock that has been receiving increasing attention in recent years, not only because it is an abundantly available and potentially inexpensive source of carbon (Aresta et al. 2013; Kondratenko et al. 2013; Assen et al. 2016), but also because it could reduce dependency on fossil fuels and carbon footprint of chemicals’ production (Dominguez-Ramos et al. 2015; Sternberg et al. 2017).

The reduction of greenhouse gas emissions is the single biggest challenge that humanity will face during the next decades. The latest IPCC report indicates that to achieve the more stringent target set by the Paris Agreement (UNFCCC 2015), equal to an increase of 1.5 °C in global temperature by 2100, anthropogenic emissions of carbon dioxide need to fall by 45% from 2010 levels by 2030 and to reach

net zero by 2050 (Masson-Delmotte et al. 2018). Technologies that capture, utilise and storage carbon will play a key role in achieving this target, alongside other low carbon technologies such as renewable energies in the power generation industry and low carbon fuels in the transport sector.

In this work, we aim to understand the environmental performance of carbon capture and utilisation technologies for the production of bulk chemicals. The relevant question that we try to answer is: “can captured CO₂ be used as feedstock to reduce the environmental impact of chemicals’ production?” The life cycle assessment (LCA) methodology is instrumental to this end: it quantifies the environmental impacts of products whilst enabling the identification of crucial trade-offs among the different environmental impacts coming from the different life cycle phases. LCA case studies for CO₂ utilisation have been published for chemicals’ production such as methanol, methane, carbon monoxide and formic acid, and for conversion technologies such as catalysis, electro catalysis and photo catalysis (von der Assen et al. 2013; Von Der Assen et al. 2014; Reiter and Lindorfer 2015; Sternberg and Bardow 2016; Zhang et al. 2017). Here we focus on a novel process using electrochemical conversion of CO₂ in a new type of ionic liquids (IL) to produce formate (Hollingsworth, S. F.Rebecca Taylor, et al. 2015a). In addition, we integrate a power plant into the studied system, as source of CO₂. Formate can in turn be easily converted into formic acid, a widely used chemical in many industrial sectors including textile, agriculture, lather and farming. In 2014, global production capacity of formic acid stood at 950, 000 tonnes/year (Hietala et al. 2016), with over 80% produced from hydrolysis of methyl formate from fossil feedstock. From 2019 to 2024, formic acid production is projected to grow at a compound annual rate of ~4%, especially in response to an increase in the demand in Asia (Intelligence 2019). Therefore, it is crucial to understand whether it can be possible to reduce its environmental impacts by using captured CO₂ as a feedstock.

This article is structured as follows: section 2 introduces the LCA methodology, the goal and the scope of the study, and describes the product system; the results and discussion are presented in section 3, the main conclusions are summarised in section 4.

2. MATERIAL AND METHODS

Life Cycle Assessment

LCA is an ISO standardised and widely used methodology for quantifying the potential environmental impacts associated with products and services (ISO. 2006; ISO 2006). The main feature of LCA is that it considers the whole life cycle of a product, which is from the extraction of raw materials to the management and disposal of wastes (**Figure 1**). LCA quantifies the potential environmental impact on global warming, acidification, eutrophication, ozone layer depletion, human and environmental toxicity, etc.



Figure 1 Phases considered in a Life Cycle Assessment study. (The Project HUB 360)

According to the ISO framework, the LCA methodology consists of four phases:

- **Goal and scope definition phase.** It includes definition of the main purpose of the study and of the functional unit, and identification of the processes that will be considered.
- **Life Cycle Inventory (LCI) phase.** This phase involves collection of data concerning material and energy flows entering and leaving the system.
- **Life Cycle Impact Assessment (LCIA) phase.** Inventory data are translated into environmental impacts by means of characterisation factors (**Figure 2**).
- **Interpretation.** Results of the LCI and LCIA phases are checked and evaluated, and key conclusions are drawn. Usually, the main outcome of an LCA study is a contribution analysis (also known as hot-spot analysis) that enables identification of those elements contributing the most to the overall environmental impact.

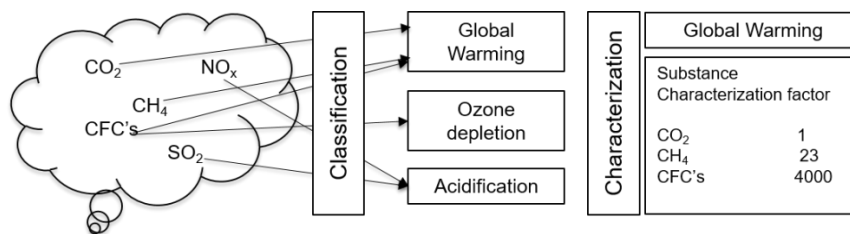


Figure 2 Classification and characterization phase. Adapted from (Moraes et al. 2014).

Goal and scope of the study

This study has a twofold goal. First, it quantifies the environmental impacts of producing formate through a novel process based on the electrochemical conversion of CO₂ captured from a natural gas power plant; it identifies the hot spots in the life cycle and proposes improvements. Second, it compares the environmental performance of CO₂-based formate production with the conventional process based on fossil feedstock with the aim of understanding the potential of using captured CO₂ to lower the environmental impacts associated with formate synthesis.

The study follows an attributional approach whereby potential consequences of decisions based on the outcome of this study are not considered. The boundaries of the product system (see **Figure 3**) follow a “cradle-to-gate” approach; it includes all processes from extraction of raw materials up to the production of formate. The environmental impacts calculated refer to the production of 1 kg of formate; this is commonly referred to as functional unit.

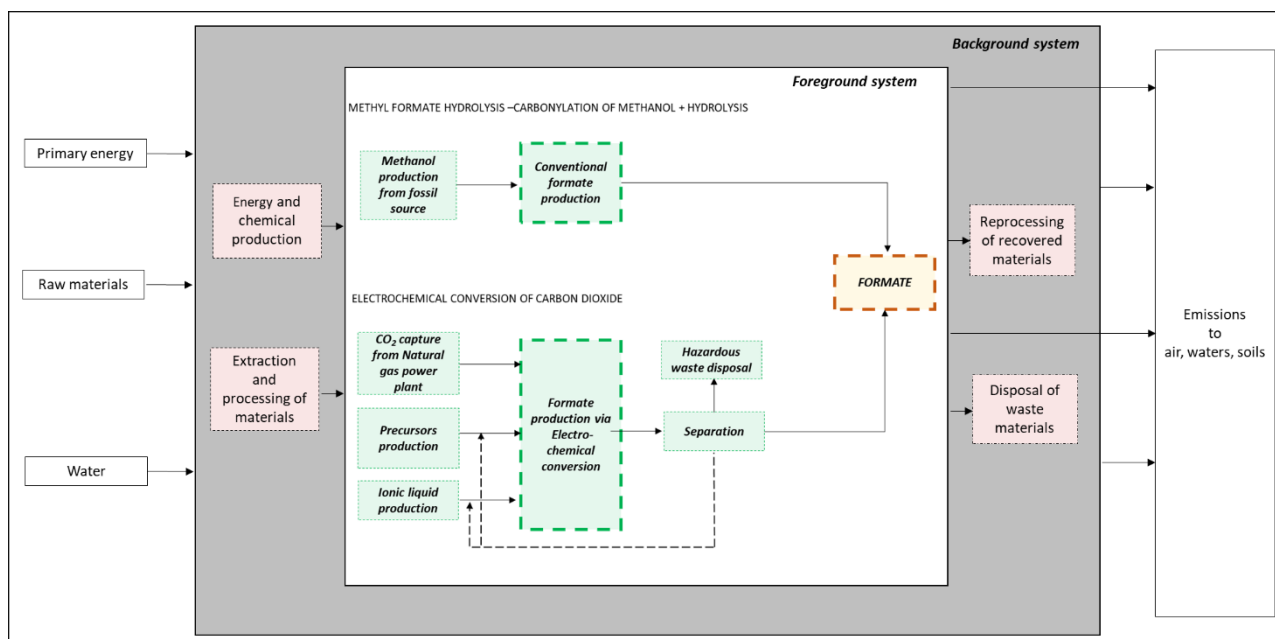


Figure 3 System boundaries used in the LCA study.

Systems description

Electrochemical conversion of CO₂ in ionic liquid

The novel synthesis route of formate is a low-carbon production based on the electrochemical reduction of CO₂ in IL. More specifically, the procedure involves the utilisation of a super basic room temperature ionic liquid, namely trihexyltetradecylphosphonium 1,2,4-triazolide [P₆₆₆₁₄][124Triz], which enables a lower energy pathway to reduce CO₂, than other kind of ILs (Hollingsworth, S. F.Rebecca Taylor, et al. 2015a; Hollingsworth, S. F.Rebecca Taylor, et al. 2015b).

As shown in **Figure 4**, the process uses a platinum and a silver electrode immersed in 8 ml solution of acetonitrile, ionic liquid [0.1 M] and water [5.6 mM] to drive an electric current via an applied potential of 0.7 V, with CO₂ bubbled at a flow rate of 15ml/min. In these conditions, formate is produced with a Faradic efficiency of 95%. (Hollingsworth, S. F.Rebecca Taylor, et al. 2015b)

Downstream of the reaction vessel, the solution is processed into the separation phase, comprised of a gas/liquid separation unit and a distillation column as reported in (Dominguez-Ramos et al. 2015), in which the formate is separated and concentrated. The effluent of the separation, rich in acetonitrile and IL is recirculated back into the system with an assumed recirculation ratio of 99.5% and mixed with fresh synthesis solution. The resulting waste stream coming out of the separation is treated as hazardous waste, which is collected and sent to incineration.

With regards to the CO₂ source, a plausible scenario was considered in which the CO₂ is captured from the flue gas coming out of a combined cycle power plant, using natural gas as a feedstock. More specifically, the CO₂ present in these exhaust gases is captured via absorption using monoethanolamine (MEA): 90% of the CO₂ in the flue gas is captured in this way (Rao and Rubin 2002), together with others pollutants, such as SO₂ NO₂, HCl and NH₃. Subsequently, the captured CO₂ is fed into the IL system. Although UK electricity generation is shifting from coal to renewable, the employment of natural gas has remained strong over the years, covering almost 40% of the national generation of electricity in 2018 (Clark et al. 2019). Such a scenario serves the purpose of contextualising these

emerging technologies within the UK's decarbonisation targets.

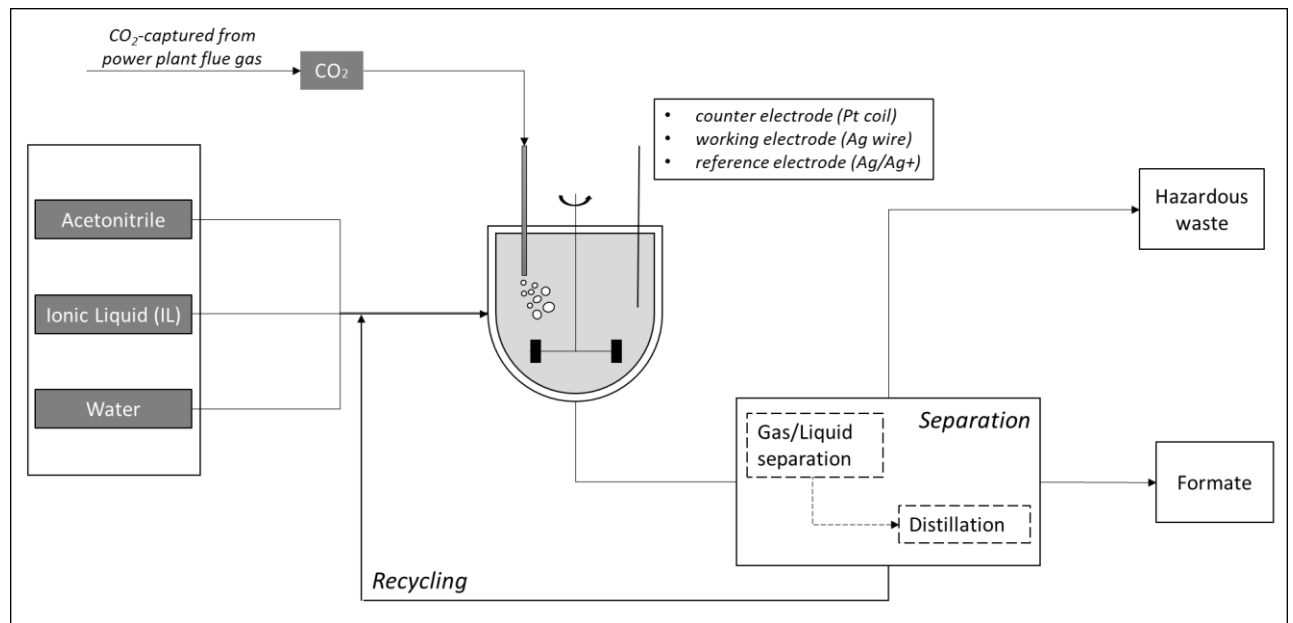


Figure 4 Schematic of the foreground system for the electrochemical conversion of CO₂ in formate at laboratory scale.

Conventional formate production: methyl formate hydrolysis

The reference system is based on the conventional production of formate, synthesized via hydrolysis of methyl formate. The process hinges on fossil fuels, primarily natural gas (Methanol Institute 2019), used as the main feedstock. Natural gas is converted into methanol that is subsequently subject to catalytic carbonylation by means of carbon monoxide. The resulting intermediate, methyl formate, is hydrolysed in presence of an additive (tertiary ammine) helping to overcome the unfavourable equilibrium of the hydrolysis and form formate (Hietala et al. 2016).

The system described is used as the basis for the process benchmarking of the IL system. The resulting differences between the two production routes such as the type feedstock -fossil fuel versus a recycled CO₂ rich waste stream- are presented and assessed in the results and discussion section (section 4). The environmental impacts of the two systems are compared to understand the potential and drawbacks of the electrochemical conversion of CO₂.

3. LIFE CYCLE INVENTORY

The conventional system was modelled using GaBi software (thinkstep AG), thinkstep (service package 36) and Ecoinvent (vv. 3.5) (Wernet et al. 2016) databases.

The electrochemical conversion of CO₂ in IL was modelled on the basis of primary data coming from laboratory experiments, along with literature data.

For the modelling of the remaining processes, such as the production of ionic liquid, carbon dioxide capture, production of electricity, heat, and so forth, secondary data from both scientific literature and

professional databases previously mentioned, were used.

A number of assumptions were required in order to perform the LCA study:

- the synthesis of formate is located in UK; hence, all related datasets used in the model refer to a UK based production;
- unless not already accounted in the dataset, transportation of goods was not considered in the model;
- the separation phase, composed of a gas/liquid separation and distillation, was integrated in the system, considering a highly optimised distillation column with a heat duty as low as 35 MJ/kg (Dominguez-Ramos et al. 2015);
- 99.5% of acetonitrile and IL is recycled back to the electrochemical reaction phase; the resulting 0.5% is sent for waste disposal;
- the wastes coming from the separation phase are identified as hazardous wastes in accordance with the European regulation (Commission of the European Communities 2000), and it is assumed that their disposal occurs via the incineration route.

Economic allocation

Allocation is used in LCA to apportion the environmental impact to each function of a multi-functional process. The natural gas power plant with carbon capture introduced above is a multi-functional process because it generates electricity and also a stream of pure CO₂ that has economic value.

In this work, we follow the ILCD (International Reference Life Cycle Data System) procedure for dealing with multi-functional processes (JRC 2010). For an attributional study and open loop recycling, the procedure envisages partitioning between the functions of the multi-functional process the environmental impacts of the process converting waste into valuable products. This is applied to the carbon capture process: it converts flue gas (waste) into a stream of pure CO₂ (valuable product). In this case, we calculate the partitioning factors based on the economic value of CO₂ and electricity; these are based on the average price of electricity in Europe, equal to 0.2 EUR/kWh (Eurostat 2019) and on an estimated price of CO₂ of 15 EUR/ton based on (Parsons Brinckerhoff 2011). Partitioning factors for electricity and CO₂ are respectively equal to 0.97 and 0.03.

4. LIFE CYCLE IMPACT ASSESSMENT: RESULTS AND DISCUSSION

In the Life Cycle Impact Assessment (LCIA) phase results are calculated for all the selected impact categories. Each impact category represents an environmental issue of concern to which each material and energy flow can be assigned (ISO 14040:2006). In this study the results are calculated by using the ILCD method (EC JRC 2011) and selecting the impact categories reported in **Table I**.

Impacts categories such as particulate matters, land use and ionizing radiation were not considered due to lack of data.

Table 1 List of the selected impact categories used for the calculation of the environmental impacts.

Impact category	Description	Unit	Abbreviation
Acidification	It is mainly caused by air emissions of NH ₃ , NO ₂ and SO _x .	[mole H ⁺ eq.]	A
Climate change, excluding biogenic carbon	Contributions of the greenhouse gases to the global warming and climate change	[kg CO ₂ eq.]	CC
Ecotoxicity freshwater midpoint	Toxic effect on aquatic freshwater species in the water ecosystems.	[CTUe]	EcoTOX
Eutrophication freshwater midpoint	Eutrophication effects in the freshwater compartment.	[kg P eq.]	E fw
Eutrophication marine midpoint	Eutrophication effects in the marine compartment.	[kg N eq.]	E mw
Eutrophication terrestrial midpoint	Eutrophication effects in the terrestrial compartment.	[mole N eq.]	E t
Human toxicity midpoint, cancer effects	Toxic effect on humans referring to potential cancer effects.	[CTUh]	HT c
Human toxicity midpoint, non-cancer effects	Toxic effect on humans referring to potential non-cancer effects.	[CTUh]	HT non-c
Ozone depletion	Depletion of the ozone layer at the stratosphere level.	[kg CFC-11 eq.]	OD
Photochemical ozone formation midpoint	Contributions of VOC (volatile organic compounds) and non-VOC to the formation of ozone at troposphere level.	[kg NMVOC eq.]	POF
Resource depletion, water	Water resource depletion.	[m ³]	RD water
Resource depletion, fossils and renewables	Depletion of mineral and fossil resources.	[kg Sb eq.]	RD m, f, ren

Results and discussion

Figure 5 includes a comparison between the environmental performance of formate obtained via electrochemical conversion of CO₂ (named low carbon production in the chart) and via the conventional process based on the hydrolysis of methyl formate. On the x-axis the graph reports impact categories (acronyms on the lower axis and metrics on the upper axis, see **Figure 5**) and multiplicative factors. The overall score in each impact category is given by values on the y axis multiplied by the factors on the x axis.

The comparison shows that the conventional process performs better than the low carbon one in all categories (including the climate change category) but the ozone depletion and water consumption categories, for which the impact of the low carbon process is ~40% lower. For the remaining impact categories, differences between the two systems range from 1.7 times in the human toxicity (non-cancer effects) category up to 7.6 times in the marine eutrophication category.

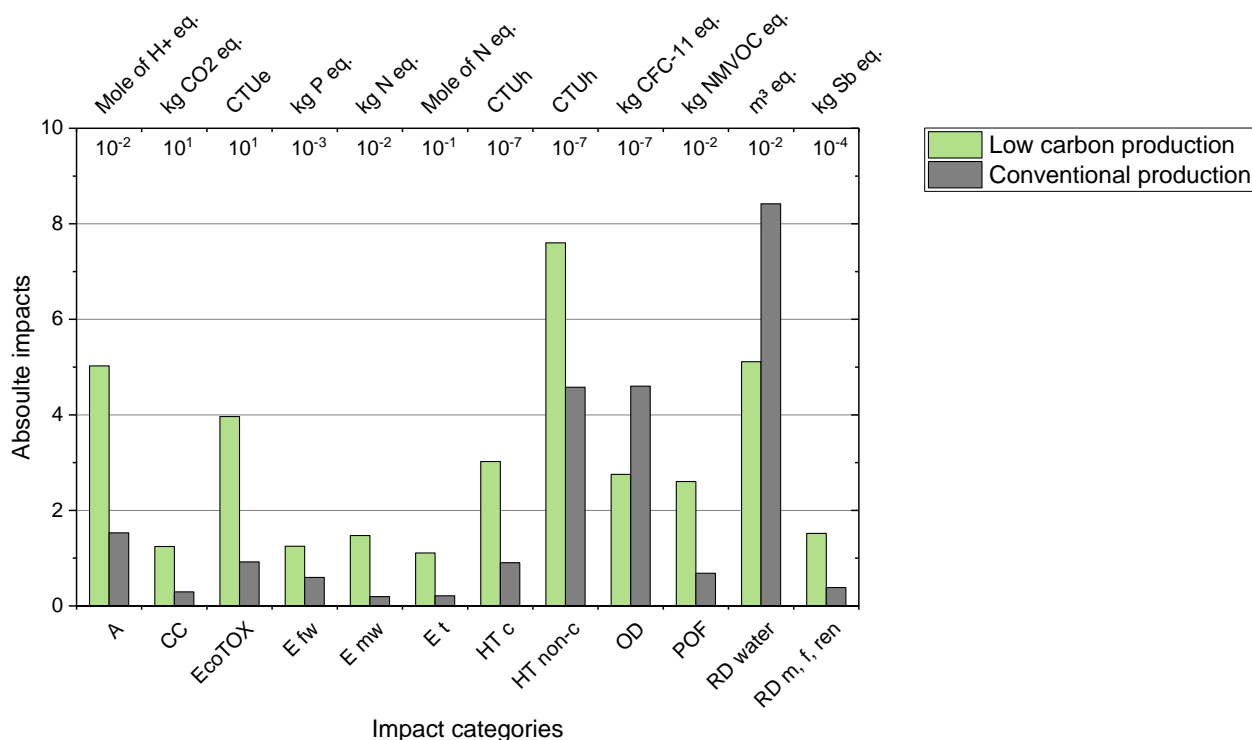


Figure 5 Comparison of environmental impacts between electrochemical conversion of CO₂ (low carbon process) and traditional process of formate production.

The low environmental performance of formate obtained via the electrochemical conversion process should not be surprising. Electrochemical conversion of CO₂ is a process thus far only implemented at laboratory scale with the objective of demonstrating its feasibility and efficiency. The process is thus not optimised in terms of consumption of energy and chemicals and it does not benefit from the economy of scale. On the other hand, the production of formate via hydrolysis of methyl formate is a well-established process implemented at commercial scale and optimised through decades of operation. However, the comparison is useful because it gives an understanding of the environmental performance that the electrochemical conversion process needs to achieve in order to become environmentally preferable.

In **Figure 6** we report a hot-spot analysis on the electrochemical conversion process showing contributions of each element (e.g. electricity, ionic liquid, acetonitrile) of the system to the overall score in each environmental impact category. The chart shows that the contributions of each element vary substantially depending on the environmental issue considered. For instance, ionic liquid [P₆₆₆₁₄][124Triz] contributions range from 6% in the climate change category up to 60% in the ecotoxicity category.

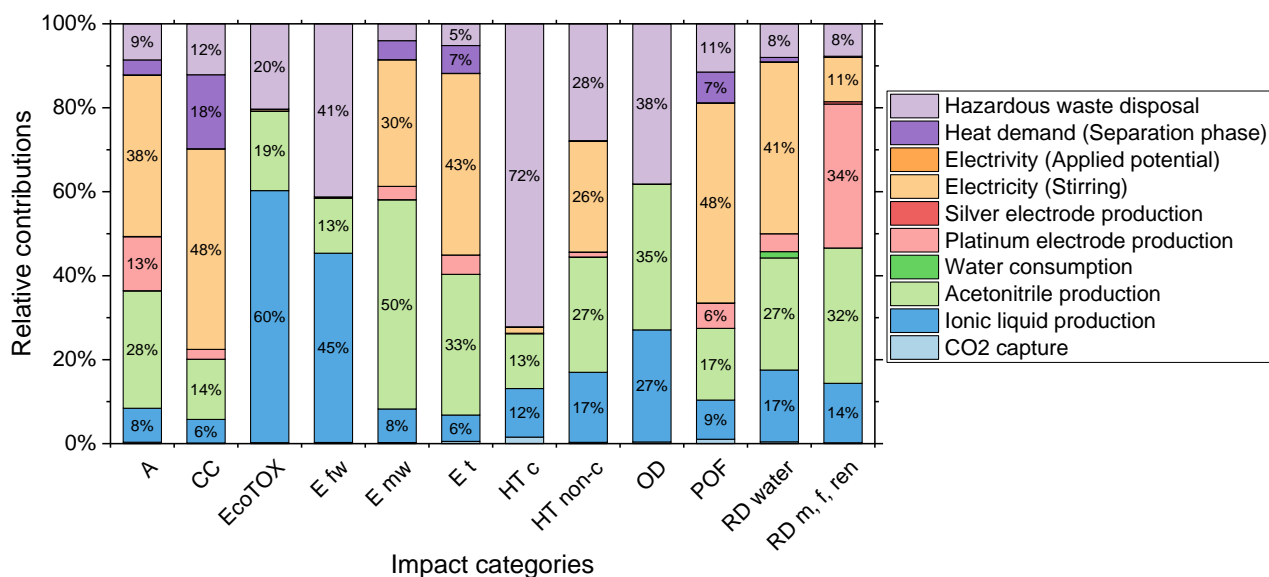


Figure 6 Hot-spots analysis of the elements of the system contributing to the environmental impacts of the low carbon production.

Overall, the greatest portion of environmental impacts is due to production of acetonitrile, production and disposal of ionic liquid, and production of electricity used for stirring. For all, the primary cause is the considerable amount that is required for producing 1 kg of formate; this is expected to diminish when moving from laboratory to commercial scale. Furthermore, the environmental impacts of ionic liquid production are primarily attributed to the substantial amount of chemical required for its synthesis (Cuéllar-Franca et al. 2016); notably, this is another process still at laboratory scale.

With respect to the other elements that are part of the product system, platinum electrodes play a significant contribution for the categories related to resources' depletion and acidification (respectively ~34% and ~13%); whilst thermal energy for the separation phase contributes to approximately a fifth of the impact in the climate change category. The remaining elements which include electricity used to drive the reduction of CO₂, silver electrode, water consumption and CO₂ capture have negligible contributions (lower than 2%). Notably, the small contribution of the CO₂ capture process is due to the price of CO₂ being significantly lower than that of electricity; this entails that only 4% of the environmental impacts of the carbon capture process are allocated to formate's production (see section 3).

5. CONCLUSIONS

This article presented a life cycle assessment study on the production of formate via electrochemical conversion of CO₂ captured from a natural gas power plant. Inventory is based on laboratory scale data for the electrochemical conversion process and on literature data for the other processes. The study adopted a "cradle-to-gate" approach, and the environmental impacts were calculated based on the production of 1 kg of formate (functional unit).

The comparative analysis between formate obtained via electrochemical conversion of CO₂ and the conventional process based on hydrolysis of methyl formate showed that the latter is environmentally preferable in the majority of environmental categories, including climate change. This is due to the still low level of technological readiness of the electrochemical conversion process which is currently at laboratory scale.

The hot-spot analysis revealed that the greatest environmental impacts related to the electrochemical conversion process are due to production of electricity used for stirring, to the production of acetonitrile and to the production and disposal of ionic liquid. These impacts are expected to diminish when moving from laboratory to commercial scale.

To this end, future works should be primarily directed at scaling up the electrochemical conversion process to evaluate its potential performance relative to the traditional process when implemented at commercial scale.

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BIOGRAPHY

Martina Pucciarelli received her MSc in Environmental Science from the University of Pisa (Italy). Her master thesis project focused on quantifying and evaluating the environmental impacts of an auxiliary power unit based on micro-tubular Solid Oxide Fuel Cell, by applying the Life Cycle Assessment methodology. Before joining the department in 2017, Martina worked as trainee at the Joint Research Centre (JRC) - European Commission (Petten, NL), where she applied the LCA methodology to the production of biofuels from micro-algae

Fabio Grimaldi was born and raised in L'Aquila (Italy) where he attended the "Universita' degli Studi dell'Aquila". During his master he spent more than a year abroad, firstly in Zaragoza for 6 months as part of the Erasmus EU programme, and successively in London at UCL where he worked on kinetic modelling on the topic "Energy from waste" for his master thesis' project. After his graduation he was offered a PhD position at UCL as part of Marie Skłodowska Curie-Horizon 2020 framework programme.

Dr. Andrea Paulillo received both his BSc and MSc in Chemical Engineering at the University of Naples "Federico II". During his MSc, he spent six months as Erasmus student at the "Universitat Politècnica de Valencia". In 2015, he moved to UCL to start his PhD on "Operationalizing the use of Life Cycle Assessment to nuclear waste management". After the PhD, Andrea became a Research Associate in Science4CleanEnergy, a Horizon 2020 project that looks at geo-energy operations in the sub-surface where he will lead the efforts on the LCA side.

Professor Paola Lettieri is Academic Director for the UCL East Campus. She is Professor of Chemical Engineering and Vice Dean (Strategic Projects) in the Faculty of Engineering Sciences at UCL. She joined the Department of Chemical Engineering in 2001 as a Research Fellow of the Royal Academy of Engineering, the first female engineer to be awarded a RAEng Fellowship. Prior to this, she spent 5 years at BP Chemicals. Her research interests are in Particle Technology, energy and waste management. She was the recipient of a prestigious Senior Research Fellowship of the Royal Academy of Engineering and The Leverhulme Trust for her work on nuclear waste management. She has authored over 180 refereed papers and 7 book chapters. She is co-author of a book published by Springer on Fluidized bed reactors: processes and operating conditions. She is a Fellow of the Institution of Chemical Engineers; Chair of the IChemE Particle Technology Specialist Interest Group. She is a member of the International Expert Group on LCA for integrated waste management.