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Comparative Life Cycle Assessment of Electrochemical and Conventional Regeneration Pathways in KOH-Based Direct Air Capture Systems

Georgia Ioanna Prokopou[a], Zoi Drakopoulou[b], Dominik Bongartz[cd], and Alexander Mitsos[eaf*]

a Process Systems Engineering (AVT.SVT), RWTH Aachen University, 52074 Aachen, Germany
b Department of Environmental and Resource Engineering, Technical University of Denmark, 2800, Copenhagen, Denmark
c Department of Chemical Engineering, KU Leuven, 3001 Leuven, Belgium
d EnergyVille, 3600 Genk, Belgium
e JARA-ENERGY, 52056 Aachen, Germany
f Energy Systems Engineering (IEK-10), Forschungszentrum Jülich, 52425 Jülich, Germany
* Corresponding Author: amitsos@alum.mit.edu.

ABSTRACT

Achieving net climate neutrality will likely require negative-emission technologies such as Direct Air Capture (DAC). Potassium hydroxide (KOH) absorption is one of the most mature DAC approaches, but it can cause significant emissions due to natural-gas-based thermal regeneration. Electrochemical regeneration methods, such as electrolysis and electrodialysis, have recently been proposed as alternatives, yet their relative performance and environmental impacts remain unclear. We present a comparative cradle-to-gate life cycle assessment (LCA) of three KOH-based DAC configurations: (i) the established Ca-looping thermal regeneration, (ii) the electrolysis regeneration (DAC-ELY), which co-produces hydrogen, and (iii) the electrodialysis regeneration (DAC-ED). The results show that, expectedly, electricity demand dominates life cycle impacts across all configurations. With the current German electricity mix, the established DAC has the lowest overall impacts, while DAC-ELY and DAC-ED exhibit higher global warming impacts due to the high electricity requirements of electrochemical regeneration. Infrastructure impacts are more noticeable for DAC-ED due to its low current density and large membrane areas. This study highlights that electrification alone does not guarantee lower life cycle impacts and that the sustainability of electrochemical DAC depends on process efficiency, material use, and the expected use of renewable electricity.

Keywords: Lifecycle Assessment, Direct Air Capture, Electrolysis, Electrodialysis, Electrochemical Regeneration

INTRODUCTION

Limiting global warming to 1.5 °C above pre-industrial levels requires reductions in greenhouse gas emissions [1]. Nevertheless, carbon-based resources are expected to remain in use in hard-to-abate sectors such as cement, steel, and aviation, where complete electrification is technically or economically challenging. As a result, residual CO_2 emissions are likely to persist, creating a challenge in achieving net-zero emission targets [2]. In this context, direct air capture (DAC) technologies, which remove CO_2 directly from the ambient air, have gained increased attention [2].

Several DAC technologies are currently under development in the market. The most mature approach is the absorption of CO_2 in a KOH solution with Ca-looping regeneration, which relies on high-temperature thermal regeneration. At the same time, novel electrochemical DAC concepts have recently emerged that replace thermal regeneration with electrically-driven processes, such as electrolysis-based regeneration and electrodialysis-based regeneration. These technologies offer potential advantages in terms of electrification and integration with renewable energy but are currently at lower

technology readiness levels [4].

Techno-economic assessments (TEAs) are widely used to evaluate emerging DAC technologies [5]. However, TEA alone does not capture the environmental implications of these systems. DAC technologies are both energy- and material-intensive, and their environmental performance is strongly influenced by factors such as the electricity supply and infrastructure requirements. A life cycle assessment (LCA) is therefore needed to identify potential environmental trade-offs.

Several studies have investigated the techno-economic and environmental performance of DAC technologies [5]-[8]. However, a comprehensive comparative LCA of the established KOH-based DAC with Ca-looping regeneration and the emerging KOH-based DAC technologies with electrochemical regeneration is still lacking in the literature.

To fill in this gap, we perform a comparative LCA of three KOH-based DAC technologies: DAC with Ca-looping regeneration, DAC with electrolysis-based regeneration, and DAC with electrodialysis-based regeneration. We thus aim to provide insights into their current environmental performance, identify key drivers of impacts, and highlight ways for improving the sustainability of electrochemical DAC systems.

SYSTEM DESCRIPTION

As mentioned earlier, three KOH-based DAC technologies are studied in the present work, which differ in the sorbent regeneration method: (i) the established Ca-looping regeneration (DAC-Established), (ii) the electrolysis regeneration (DAC-ELY), and (iii) the electrodialysis regeneration (DAC-ED). A schematic representation of the studied technologiesis given in Figure 1.

Direct Air Capture with Ca-looping Regeneration

This technology is commercialized by Carbon Engineering [3], exhibits a high TRL of 7-8 [9], and will be considered as the benchmark for the comparison of the other technologies. It consists of two connected chemical steps. In the first step, CO_2 from the atmosphere is brought in contact with a KOH solution in an air contactor unit, producing an aqueous solution of K_2CO_3. In the second step, the solution enters a pellet reactor in which it reacts with $Ca(OH)_2$, producing $CaCO_3$ and KOH. Then the $CaCO_3$ enters a calciner and is decomposed to CaO at high temperatures of 900 °C, while releasing CO_2. The heat needed for the release of CO_2 is provided by burning natural gas [10]. Finally, the CaO is hydrated in a slaker unit to regenerate the $Ca(OH)_2$ that is fed back to the pellet reactor. If the additional CO_2 released from the natural gas burning is not captured, then the process could potentially lead to positive direct CO_2 emissions.

Direct Air Capture with Electrolysis Regeneration

DAC with electrolysis regeneration is a technology at a low TRL but has been proposed as a promising alternative to the established thermal process [11], as it is powered by electricity and can integrate renewable energy [4].

The process consists of two coupled chemical steps, with the first being the same as in the DAC-Established. In the second step, KOH regeneration and CO_2 release are achieved in an electrochemical reactor. This reactor comprises an anode, an anolyte compartment, a cation exchange membrane (CEM), a catholyte compartment, and a cathode [11]. The $CaCO_3$ solution produced from the first step is fed to the anodic side of the electrochemical reactor. By applying current, the water oxidation reaction (WOR) occurs at the anode, where gaseous oxygen and protons are produced according to Reaction (1), while the water reduction reaction occurs at the cathode, producing gaseous hydrogen and hydroxide anions according to Reaction (2):

$$H_2O \rightarrow 2H^+ + 2e^- + 0.5O_2 \quad (1)$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

Simultaneously, mainly K^+ migrate through the CEM from the anolyte to the catholyte. This results in an acidic environment in the anolyte and in an alkaline environment at the catholyte. The acidic environment in the anolyte drives the conversion of carbonate species into gaseous CO_2, which is released in the form of bubbles, while the alkaline environment in the catholyte enables the regeneration of the KOH [11]. Due to the production of O_2 and the release of CO_2 in the anolyte, an additional pressure swing adsorption unit (PSA) is required after the electrochemical reactor to separate the two gases [4].

Direct Air Capture with Electrodialysis Regeneration

Electrodialysis regeneration is another electrochemical approach that has been investigated for DAC. It exhibits a TRL of approximately 4 [5], and similarly to the electrolysis regeneration, the operating principle is based on the pH gradient across ion-exchange membranes.

As in the electrolysis regeneration, the process consists of two main steps, with the initial CO_2 capture step being identical to that of the DAC-Established process. In the second step, the $CaCO_3$ solution is fed into an electrodialysis regeneration unit, which exhibits some differences compared to the electrolysis regeneration. The electrodialysis reactor consists of a single anode and cathode, while in between multiple Bipolar Membranes (BPM) and CEM pairs are arranged in series [4].

By applying current, the reactions (1) and (2) occur at the anode and cathode, respectively. In addition, the

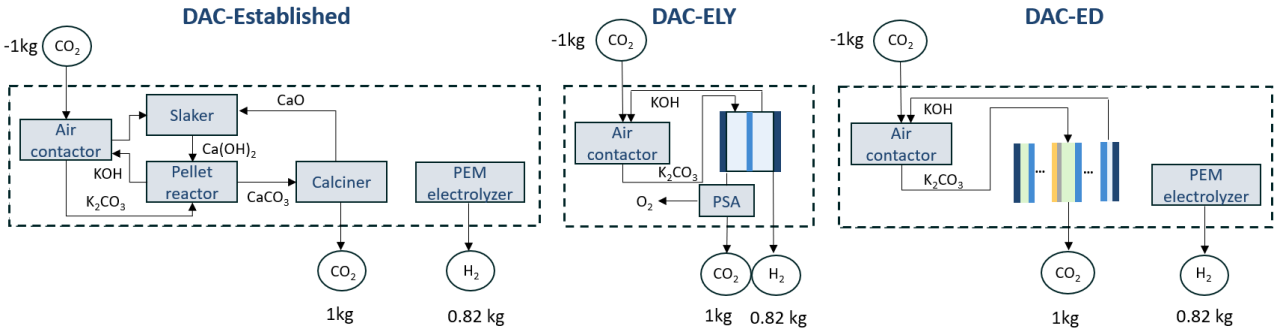


Figure 1. Schematic representation of the cradle-to-gate system boundaries for the DAC with Ca-looping regeneration (DAC-Established), the DAC with electrolysis regeneration (DAC-ELY), and the DAC with electrodialysis regeneration (DAC-ED).

BPMs, which consist of an anion exchange layer and a cation exchange layer, result in water dissociation into OH^- and H^+. This generates acidic and alkaline environments on opposite sides of the membrane, establishing a pH gradient across the CEM [4]. Similarly to the electrolysis regeneration, in the acidic compartments, CO_2 is released in the form of bubbles. In contrast to the electrolysis regeneration, CO_2 is released in chambers that are physically separated from the anode, where oxygen is generated. As a result, no additional PSA unit is required for gas separation. Moreover, the large number of BPM-CEM pairs between the two electrodes allows operation at a lower current density compared to the electrolysis unit. Consequently, the production rate of hydrogen is considered negligible compared to the release of CO_2 [4].

MATERIALS AND METHODS

The LCA was conducted in accordance with the ISO 14040/14044 framework, incorporating the main phases of goal and scope definition, life cycle inventory analysis, life cycle impact assessment, and interpretation [12].

Goal and Scope Definition

The goal of the present study is the comparative LCA of the three DAC technologies. We assess the environmental impacts by performing a cradle-to-gate analysis, including the infrastructure and air capture processes, while excluding end-of-life treatment due to the unknown fate of the final products, as they are similar among the technologies.

The DAC-ELY co-produces hydrogen as a valuable product during electrochemical regeneration. Under the operating current density of 0.14 A cm^{-2}, the hydrogen co-production is 0.82 kg H_2 per kg CO_2 captured [4]. To ensure functional equivalence among the compared systems, multifunctionality is addressed by expanding the system boundaries [13]. In this context, a PEM water electrolyzer is included within the system boundaries for the DAC-Established and the DAC-ED cases to generate 0.82 kg of H_2. Thus, the functional unit is 1 kg of CO_2 and 0.82 kg of H_2.

Life Cycle Inventory Analysis

The life cycle inventory analysis was performed using literature process modeling and experimental data for the different DAC technologies. GaBi [14] was employed as LCA software to calculate the environmental impacts. Background data for material and energy flows associated with process infrastructure were obtained from the Ecoinvent database [15]. For all the assessed technologies, the electricity supply for the operation was assumed to be provided by the German electricity grid (unless otherwise stated), and the lifetime of the equipment and the plant is assumed to be 20 years. In the following, the assumptions of the inventory modeling and the data used for the different DAC technologies are discussed in detail.

PEM Electrolyzer

A PEM electrolyzer operating at a current density of 1.5 A cm^{-2} is assumed to produce H_2 in the DAC-Established and the DAC-ED, according to Zachert et al. [16]. Infrastructure data for the PEM electrolyzer stack and balance of plant (BoP) are taken from Bareiß et al. [17]. Due to the lack of inventory data for iridium at the anode, platinum was assumed as a proxy based on available data from the Ecoinvent database [15]. Moreover, the Nafion membrane material was approximated as tetrafluoroethylene (TFE) [18].

Direct Air Capture with Ca-looping Regeneration

Operational data for the DAC-Established process are taken from Keith et al. [10]. The thermal energy required for sorbent regeneration is assumed to be supplied by natural gas combustion. The oxygen required for combustion is assumed to be provided by the PEM electrolyzer; therefore, no additional air separation unit was

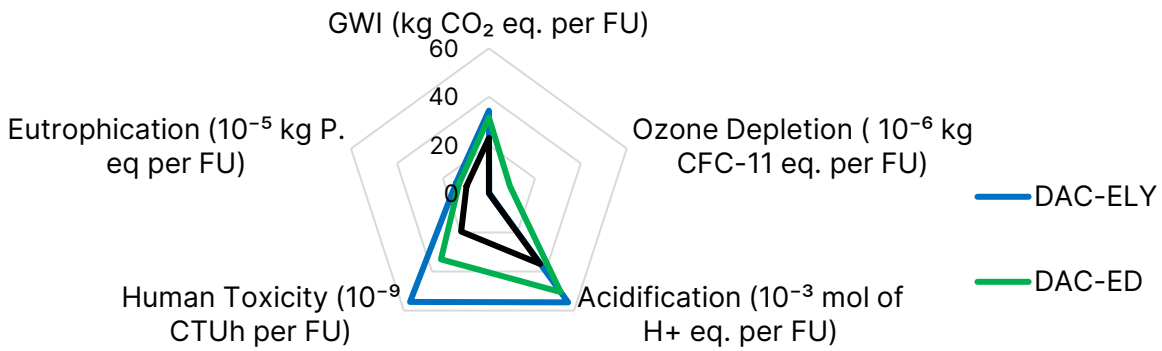


Figure 2: Comparative life cycle environmental impacts of the DAC with Ca-looping regeneration (DAC-Established), DAC with electrolysis regeneration (DAC-ELY), and DAC with electrodialysis regeneration (DAC-ED), assuming the German electricity grid. The DAC-Established exhibits the lowest life cycle impacts in all assessed categories, while the DAC-ELY shows the highest scores in all categories except ozone depletion. The functional unit (FU) is 1 kg CO_2 captured and 0.82 kg H_2 produced. GWI refers to the global warming impact.

included in the system boundaries [19]. Emissions from natural gas combustion were excluded from the inventory, as the CO_2 produced during sorbent regeneration was assumed to be fully captured. Only emissions associated with natural gas production and transportation were considered [19]. Infrastructure data for the air contactor and the regeneration units are taken from Madhu et al. [20]. The pellet reactor, slaker, and calciner were approximated as steel tanks for the purpose of infrastructure modeling [20].

Direct Air Capture with Electrolysis Regeneration

An electrolysis regeneration unit operating at a current density of 0.14 A cm^{-2} is assumed according to the operational data from Liu et al. [4]. A PSA unit for the separation of CO_2 and O_2 is included [4], with infrastructure data for the PSA taken from Burgers et al. [21]. Infrastructure data for the electrolysis regeneration unit stack and the BoP are obtained from Bareiß et al. [17], with modifications to reflect the specific operating conditions of the DAC system. In contrast to the PEM electrolyzer, a nickel catalyst is used at the cathode instead of platinum due to the alkaline operating environment [11]. Similarly to the PEM electrolyzer case, platinum is used as a proxy for iridium at the anode and TFE for the Nafion membrane.

Direct Air Capture with Electrodialysis Regeneration

An electrodialysis regeneration unit operating at a current density of 0.012 A cm^{-2} is assumed according to the operational data from Liu et al. [4]. Each regeneration unit is assumed to consist of 20 CO_2 recovery-alkali regeneration pairs [4]. Infrastructure data for the electrodialysis regeneration unit stack and the BoP are obtained from Bareiß et al. [17]. Similarly to the electrolysis regeneration case, a nickel catalyst is assumed at the cathode, while platinum was used as a proxy material at the anode. TFE is used as a proxy for the CEM, and the BPM is assumed to consist of two Nafion membranes [22].

Life Cycle Impact Assessment

We investigated five typical impact categories: global warming impact (GWI), ozone depletion, acidification, human toxicity, and eutrophication. The environmental impacts were quantified using the Environmental Footprint (EF) 2.0 impact assessment method [23]. The results are normalized to the functional unit of 1 kg CO_2 captured and 0.82 kg H_2 produced.

RESULTS AND DISCUSSION

Environmental Performance across Impact Categories

Figure 2 presents the comparative results for the different impact categories of the three DAC technologies assuming the German electricity grid. Overall, the DAC-Established technology exhibits the lowest life cycle impacts in all assessed categories. This can be attributed to its higher technological maturity and optimization level compared to the more novel electrochemical DAC processes. In contrast, the DAC-ELY shows the highest life cycle impacts in all categories except for ozone depletion. For ozone depletion, the DAC-ED has the highest impact, which can be attributed to the larger number of membranes in the electrodialysis configuration, leading to increased material-related ozone depletion contributions.

Since the global warming impact is one of the most important categories, we focus in the following subsections on its detailed analysis, including process- and infrastructure-level contributions, to explain the observed differences among the assessed technologies.

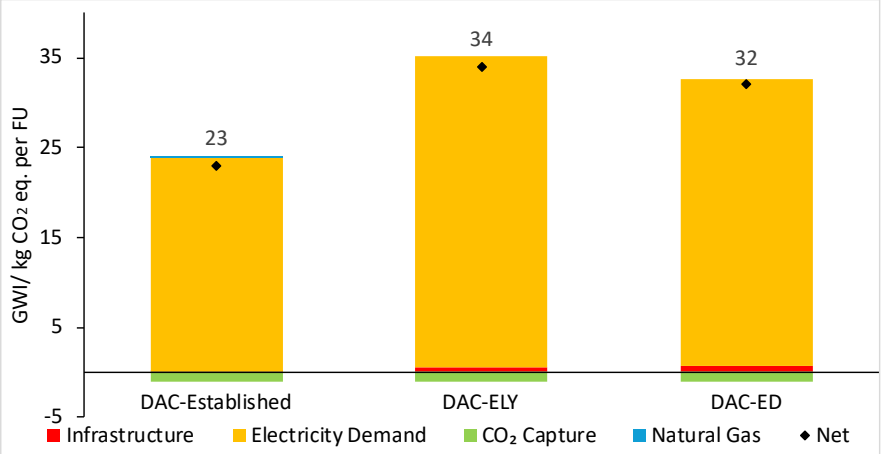


Figure 3: Cradle-to-gate global warming impact (GWI) distribution for the DAC with Ca-looping (DAC-Established), the DAC with electrolysis regeneration (DAC-ELY), and the DAC with electrodialysis regeneration (DAC-ED), assuming the German electricity grid. The main GWI contributor is the electricity demand in all three technologies. The DAC-ELY exhibits higher electricity demand compared to the DAC-Established and DAC-ED. The negative GWI is attributed to the direct removal of CO_2 from the atmosphere through the DAC process. The infrastructure contribution is approximately 2% of the total GWI in the DAC-ED. The functional unit (FU) is 1 kg CO_2 captured and 0.82 kg H_2 produced.

Contribution Analysis of the Global Warming Impact

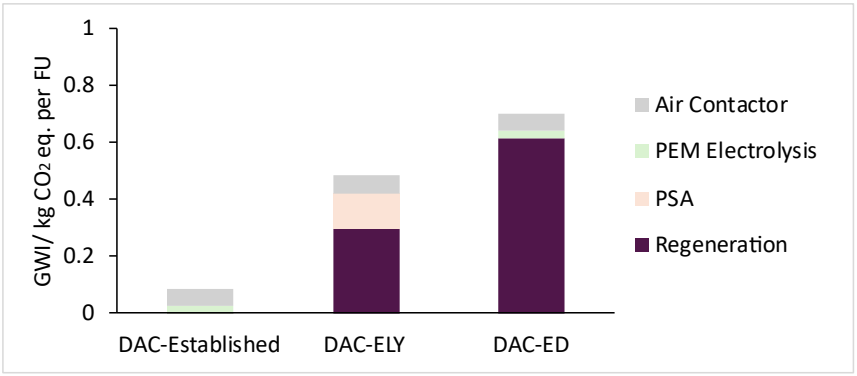
Figure 3 presents the global warming impact distribution for the three DAC technologies. Overall, in all three technologies, the electricity demand dominates the GWI. There is a 47% and 38% increase in the net GWI of the DAC-ELY and the DAC-ED, respectively, compared to the DAC-Established. It should be noted that the high electricity demand in the DAC-Established and the DAC-ED is mainly due to the additional PEM-electrolyzer included to ensure a fair comparison of the three technologies via the system expansion approach.

It can also be seen that there are negative GWI corresponding to 1 kg of CO_2 eq. per kg of FU, reflecting the direct removal of CO_2 from the atmosphere through the DAC process. Moreover, the highest infrastructure GWI is observed for DAC-ED of about 2% of the total impact. Overall, the contribution of the infrastructure is negligible compared to the electricity demand. However, in case there is a larger penetration of renewable energy resources (as analyzed below), the relative contribution of the infrastructure may become more significant. Therefore, it is still of interest to examine the differences in the infrastructure contributions, as follows.

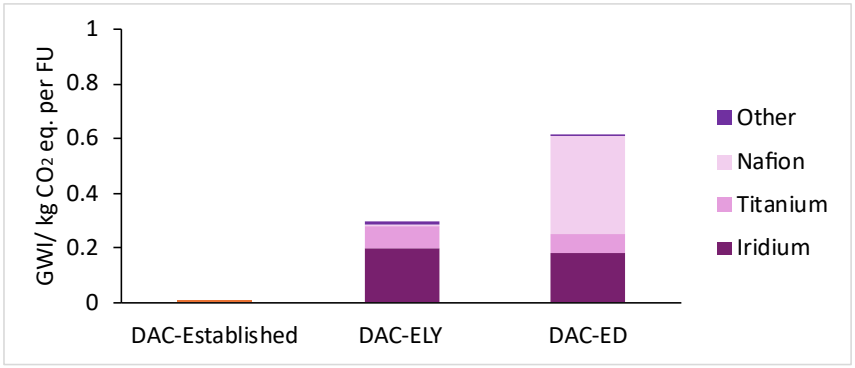
Contribution of Infrastructure to Global Warming Impact

Figure 4a) presents the distribution of infrastructure-related contributions to the global warming impact of the three DAC technologies. The regeneration unit is the main contributor to the infrastructure impact in the case of DAC-ELY and DAC-ED, while DAC-Established has a negligible infrastructure impact. The contribution of the air contactor is identical across all three technologies, reflecting the use of the same design assumptions. At the same time, the PEM-electrolyzer exhibits considerably lower infrastructure GWI compared to the electrochemical regeneration units in the DAC-ELY and DAC-ED systems. This is primarily due to the higher operating current density (1.5 A cm^{-2} compared to 0.14 A cm^{-2} vs 0.012 A cm^{-2}, respectively), which translates into a lower amount of material requirement per functional unit.

Figure 4b) presents the distribution of infrastructure-related contributions from the regeneration units to the global warming impact of the three DAC technologies. For both DAC-ELY and DAC-ED, the main contributors within the regeneration units are the iridium, titanium, and Nafion-based membrane materials. In the case of DAC-ED, the contribution associated with the Nafion membranes is higher than in the case of DAC-ELY. This can be attributed to two factors. First, the DAC-ED regeneration unit operates at a current density approximately eleven times lower than that of the DAC-ELY, resulting in higher material requirements per functional unit. Second, the DAC-ED requires 3 additional membranes per CO_2 recovery-alkali regeneration pair. Therefore, the influence of Nafion membranes on the overall infrastructure-related global warming impact is more pronounced in the DAC-ED system.



a)



b)

Figure 4: (a) Distribution of infrastructure-related global warming impact (GWI) and (b) distribution of infrastructure-related contributions from the regeneration units to the GWI for the DAC with Ca-looping (DAC-Established), the DAC with electrolysis regeneration (DAC-ELY), and the DAC with electrodialysis regeneration (DAC-ED). The regeneration unit has the highest contribution to the infrastructure of DAC-ED, with Nafion-based membranes being the dominant contributing material. The functional unit (FU) is 1 kg CO_2 captured and 0.82 kg H_2 produced.

Influence of the Carbon Footprint of Electricity on the Global Warming Impact

To assess the potential of the novel electrochemical DAC technologies, in Figure 5, we present the influence of the carbon footprint of electricity across different countries on the GW [24]. A lower electricity carbon footprint corresponds to a higher penetration of low-carbon energy sources in the respective electricity mix. The DAC-Established exhibits the lowest GWI across all electricity scenarios. Moreover, the DAC-ELY approaches the GWI of the DAC-ED at lower carbon footprints of electricity, since the electricity demand contribution becomes lower.

The results suggest that, in addition to the larger penetration of renewable electricity, further performance improvements are needed for DAC-ELY and DAC-ED to compete with the DAC-Established technology. Reductions in materials use and infrastructure-related impacts or operation at higher current densities will be necessary to realize the potential of electrochemical DAC technologies.

CONCLUSION AND OUTLOOK

We performed a comparative LCA of three DAC technologies: the established DAC technology with Ca-looping regeneration; the novel technology of DAC with electrolysis regeneration; and the DAC with electrodialysis regeneration. The results have shown that the DAC-Established outperforms the other technologies in all impact categories, when assuming the German electricity grid. DAC-ELY shows the highest GWI due to the higher electricity demand associated with the electrochemical sorbent regeneration. The infrastructure impact is higher in the case of DAC-ED due to both the use of additional membranes and the low operating current densities. Finally, it is concluded that DAC-ELY and DAC-ED need further performance improvement and a larger share of renewable energy to compete with the DAC-Established.

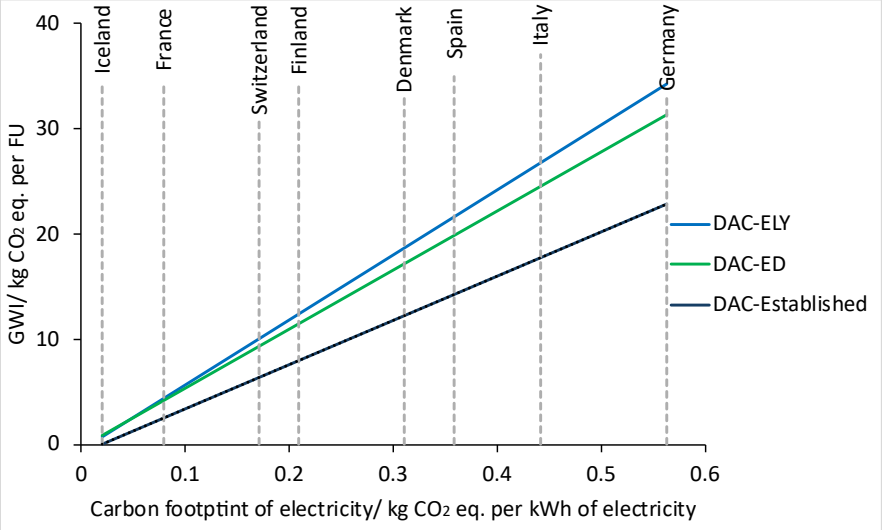


Figure 5: Global warming impact (GWI) with respect to the carbon footprint of electricity for the DAC with Ca-looping (DAC-Established), the DAC with electrolysis regeneration (DAC-ELY), and the DAC with electrodialysis regeneration (DAC-ED). DAC-Established exhibits the lowest GWI across all electricity mix scenarios. DAC-ELY approaches the GWI of DAC-ED in cases of larger penetration of alternative energy resources. The functional unit (FU) is 1 kg CO_2 captured and 0.82 kg H_2 produced.

However, several limitations of this study should be acknowledged. In cases where no dataset was available, proxies were used for the infrastructure, which may not fully capture the specific impacts of the actual materials. Moreover, upstream processes (e.g., metal extraction, refining) were modeled with the average European electricity mix, which may lead to changes if region-specific grids were applied.

Future work should extend the comparison of these DAC technologies beyond environmental performance to include TEA. This will allow for a more holistic evaluation of the three alternatives. Moreover, system optimization with respect to both environmental impacts and costs may help identify trade-offs and pathways for improving the overall performance of electrochemical DAC technologies.

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AUTHOR IDENTIFIERS

Author ORCIDs:
Prokopou G.I.: 0009-0007-2643-6899
Drakopoulou Z.: 0009-0009-8064-1898
Bongartz D.: 0000-0003-1790-0235
Mitsos A.: 0000-0003-0335-6566

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