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Sustainability analysis of CO_2 capture and utilization processes using a computer-aided tool

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ABSTRACT

CO_2 capture and utilization technologies (CCU) are recently attracting attention as ways to reduce CO_2 emission and generate economic benefits at the same time. Since numerous potential products from CO_2 may be considered and multiple processing pathways are possible for each product, there is a growing demand for a tool that can aid in techno-economic and life cycle CO_2 analyses of a large number of CCU options, in order to identify promising ones. This work introduces a computer-aided analysis tool called ArKa-TAC^3 tailored for this purpose. ArKa-TAC^3 can calculate both techno-economic and CO_2 reduction metrics of CCU processes in a fast and convenient manner. Sufficient flexibility is assured by adopting a superstructure model framework, which allows the user to conveniently describe a CCU processing network composed of multiple processing steps with a large number of technical options. To demonstrate the tool, a CCU process of acetic acid production is designed and its sustainability is analyzed by using it. By implementing the designed process in four different countries, it is verified that the CCU process can be made sustainable by adopting the process substitution strategies its implementation. Some perspectives on potential applications of the developed tool are given.

1. Introduction

Excessive anthropogenic greenhouse gas (GHG) emissions have disrupted the global carbon cycle leading to global warming and climate change. Among various GHGs, CO_2 is the primary GHG as it accounts for the largest portion (77%) of the global GHG emissions [1]. Carbon capture, utilization, and sequestration (CCUS) technologies are attracting attention as a way to abate the GHG emissions until the full conversion to renewable energy takes place. CCUS involves capturing CO_2 from sources like flue gas or industrial off-gas and storing (e.g., in saline aquifers) or utilizing them (e.g., as an extractive agent or as feedstock for conversion into chemicals or fuels) instead of emitting them into the atmosphere. In particular, carbon capture and utilization (CCU) can provide economic incentives that can compensate for the costs in contrast to carbon capture and sequestration (CCS).

One of the recent issues has been the need to evaluate CCU technologies to identify '*sustainable*' options among a large number of candidates [2]. The need arises as numerous products can be made from CO_2, and moreover, these products can be made through multiple processing pathways. Here, 'sustainable' CCU technology means that it is capable of achieving CO_2 reduction with economic viability. Although various ideas for CCU have been suggested, not all of them are sustainable in this sense as most CCU technologies require significant energy inputs, which themselves carry carbon footprints. For instance, methanol production through CO_2 hydrogenation reaction ($CO_2 + 3H_2 \rightarrow CH_3OH + H_2O$) is a representative CO_2 utilization path that can be made sustainable with renewable resources but may be too expensive at present. If the hydrogen feedstock is generated from fossil fuels to improve the economics, it results in a net-positive CO_2 emission meaning that, in the process of converting CO_2 into methanol, more CO_2 is emitted to the atmosphere than consumed [3].

As summarized in Roh et al. [2], a limited number of CCU technologies such as those for producing methanol [4], dimethyl carbonate [5], and sodium carbonate [6], have been analyzed in terms of sustainability, but the sustainability of many other technologies remain unanswered. Theoretically, the number of potential products from CO_2 feedstock stands at more than one hundred [7]. Many more candidate processes need to be examines if every possible reaction path and

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technology are to be taken into account. In order to determine promising CCU technologies systematically, it is important to analyze and evaluate a large number of candidate pathways and technologies fast and efficiently.

Nevertheless, it is difficult to analyze the sustainability of CCU technologies due to the need to consider multiple evaluation metrics. Net profit ($/ton-product) is a key metric for assessing the economic viability while net CO_2 lifecycle emissions (or carbon footprint, mass of CO_2 equivalent emission per mass of product) determine the CO_2 reduction capability. As such, both techno-economic analysis (TEA) and CO_2 lifecycle assessment (LCA) must be performed, but doing so is both time and labor intensive. First, a comprehensive set of data detailing the mass balances, utility consumptions of the CCU processes, and prices and CO_2 lifecycle inventory (LCI) of involved raw materials, products, and utilities should be collected. If a comparative analysis against a reference case is desired, such data for the reference case should be collected also. Sometimes, the LCA and TEA results should be combined to form additional metrics. For instance, CO_2 avoidance cost (dollar per ton of avoided CO_2 equivalent), which is a metric frequently used for comparing the competitiveness of CCUS technologies, requires net cost and net CO_2 reduction, to be derived from TEA and LCA respectively. While commercial tools such as Aspen Process Economic Analyzer® [8], Ecoinvent® [9], and SimaPro® [10] are there to assist TEA and LCA studies, compiling the individual results to prepare appropriate metrics for the evaluation of CCU technologies can be extremely tedious and time-consuming. All the commercial tools for TEA and LCA are applicable to a wide range of technologies and processes. Nevertheless, they are not developed specifically for CCU technologies and therefore have limitations and inconveniences. To the best of our knowledge, there is no tool tailored to the analysis of CCU yet.

To solve this problem, we have developed a computer-aided tool tailored to CCU processes for this purpose. It is called *ArKa-TAC³*, which stands for **Ar**amco-**KA**IST/**T**ool for **A**nalysis of **CO_2** **C**apture and **C**onversion systems. ArKa-TAC³ is a software that enables an easy construction of multiple candidate CCU processing paths and the calculation of key metrics for evaluating the sustainability of a specific pathway in a quick and convenient manner. Adopting the concept of *superstructure model*, which is a network model that encompasses all possible candidates and design options, the software allows the user to describe a CCU processing network encompassing multiple technical options (e.g., processing paths). The concept of superstructure model is described in Section 3. In addition, users can perform both TEA and CO_2 LCA on a selected processing path within a constructed network efficiently, as the software provides the database and the calculation engine.

In the following sections, a methodological framework for evaluating the sustainability of CCU processes is briefly reviewed. Within the framework, the role of ArKa-TAC³ is highlighted. Also, the structure of the software is described with requisite model equations. As a case study, a CO_2 capture and utilization path for acetic acid production is introduced and its sustainability is analyzed using the software. Finally, some perspectives on the application of the developed tool are given.

2. Evaluation framework for sustainable CCU processes

For a CCU process to be sustainable, it should be able to reduce CO_2 emissions in an economically feasible way (as stated in Section 1). Roh et al. [11] proposed a framework for designing a sustainable CCU process, which involves three decision steps considering the CO_2 reduction feasibility, economics, and market size with certain requirements to be met. The requirements concern net CO_2 reduction (related to net CO_2 emission), economic viability, and CO_2 reduction volume and are summarized in Table 1.

The first requirement concerns the *net CO_2 reduction* as reducing CO_2 emissions is the ultimate goal of implementing CCU processes. As shown in Eq. (1), the specific net CO_2 emission ($f_{CO_2}^{NE,j}$) of a given CCU process should be less than that of non-CO_2 based production of an equivalent product or application ($f_{CO_2}^{REF,j}$). The specific net CO_2 emission equals to the total amount of CO_2 (and other GHGs) emitted minus the amount of CO_2 feed. Here, the total CO_2 emission includes direct emissions as waste (e.g. off-gas) as well as indirect emissions for producing required raw materials and utilities (e.g. electricity, steam, or cooling water), and consumption of the product. If the below condition is satisfied, the CO_2 reduction can be achieved by replacing the *reference* process with the CCU process.

$$f_{CO_2}^{NE,j} < f_{CO_2}^{REF,j},\ j = PR\ or\ PD \tag{1}$$

In evaluating the above condition for a given CCU process, two implementation strategies can be envisioned: process substitution (*PR*) and product substitution (*PD*). Process substitution is the replacement of *a non-CO_2-based process* (not utilizing CO_2) by a CCU process that produces equivalent products. Product substitution, on the other hand, is to replace *a non-CO_2-based product* by a different product from CCU for an equivalent function. For a fair comparison, each strategy needs a different form of the specific net CO_2 emission metric (Eq. (34) and (35)). Furthermore, clear demarcation of the system boundary is very important in CO_2 LCA. Fig. 1 represents a general system boundary of CCU processes and their reference cases. It encompasses the CO_2 emission source, CO_2 capture and transport, CO_2 utilization, and transport, use and disposal of the product. In addition, acquisition, manufacture and production of raw materials and utilities should be included. In the case of process substitution, the steps of product transport and consumption can be ignored in the calculation of the net CO_2 emission because such steps are same for both the CCU process and the reference process. However, in the case of product substitution, CO_2 emissions from the whole product lifecycle (cradle-to-grave) should be considered because the CO_2 emissions in the product transport and consumption steps may be different for the CCU process and the reference process [12].

When $f_{CO_2}^{NE,j}$ is negative as well as less than $f_{CO_2}^{REF,j}$, the amount of CO_2 consumed by the CCU process is greater than that of CO_2 generated by the process, and then *standalone implementation* of the new CCU process can contribute to the reduction of CO_2 emission. This means that no substitution with a reference case is needed to achieve the CO_2 reduction.

The second consideration is *economic viability*, which is a point of major concern for both traditional process design and CCU process design. In general, various metrics can be used such as unit production cost or net profit on the basis of mass (e.g. $/ton) or functional unit (e.g. $/GJ). To derive these values, the mass and energy balance and equipment size data of a given CCU process are required, and process simulation can be used to obtain such data. Obviously, positive and high net profit is preferred. Cheaper production cost than a competing product is also desired. On the other hand, if the CCU process is not profitable, this does not necessarily mean that it is infeasible. This is a common misconception. In this case, the feasibility of the process may still be assessed by comparing its *CO_2 avoidance cost* ($\$/ton_{CO2e}$) with that of CO_2 sequestration, such as storing captured CO_2 in saline aquifers or reservoirs, since both are cost-burden options for CO_2 reduction.

Based on the two requirements, the CCU processes can be classified into four cases based on net CO_2 emission and profitability (Fig. 2). The process belonging to the second quadrant (top-left) is an absolutely sustainable (best) option as it can reduce the CO_2 emission with economic benefits. Conversely, if the process is in the fourth quadrant (bottom-right), it is not a sustainable option as it can achieve neither. Most CCU processes tend to place in the first (top-right) or third (bottom-left) quadrant, and these processes can be analyzed by comparison with reference cases or CO_2 sequestration as mentioned above.

The third point of consideration is *potential CO_2 reduction volume*. This concerns the scale of the expected amount of CO_2 reduction by