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Optimal Design of CO_2 Capture, Utilisation, Mineralisation, and Sequestration Networks within Industrial Clusters

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Holistic development of CO_2 capture, utilisation, and storage (CCUS) networks is crucial for the cost-effectiveness and the widespread deployment of such technologies in the industry. This work proposes a novel framework for the design and optimisation of CCUS networks within industrial clusters. Models for advanced CO_2 capture, utilisation, mineralisation, compression, transportation, and sequestration processes, coupled with economics, are developed and employed. Precipitated calcium carbonate (PCC) nanoparticles produced by a rotating-packed bed process are considered the sole product of CO_2 utilisation and provide revenue in the CCUS network. The framework aims to minimise the total annual cost of the network by adapting the optimal designs of each subprocess and selecting the most suitable CO_2 routes while ensuring a 90 % decrease in CO_2 emissions. A mixed-integer linear programming (MILP) framework is used to solve the optimisation problem. The performed case studies involve 5 industrial emitters from different industrial sectors, 3 sequestration sites, and one mineral deposit site. The results showed that the traditional CO_2 capture-transportation-sequestration chain is favourable when we assume no revenue from the utilisation process, and it offers a 7.2 % lower cost per ton of avoided CO_2 than purchasing carbon permits with the current price. Considering revenue from PCC drives all available $Ca(OH)_2$ into the utilisation process, reducing the network's cost per ton of CO_2 by 10.9 % and 3.9 % compared to the carbon permits and CCS network costs.

1. Introduction

The increasing CO_2 concentration in the atmosphere is identified as the main contributor to global warming. The Paris Agreement sets the goals for a 43 % decrease in greenhouse gas (GHG) emissions by 2030 and carbon neutrality by 2050. To achieve those goals, the systematic adaptation of different low-carbon technologies is imperative despite the current wide dependency on fossil fuel energy (Tapia et al., 2018). CO_2 capture, utilisation, and sequestration (CCUS) are promising technologies to mitigate anthropogenic GHGs, as they can achieve high capture efficiencies and be integrated into existing industrial plants. As such, they can play an important role in the transition into low-carbon technologies and a fossil-free energy economy (Gibbins and Chalmers, 2008). Even so, the widespread deployment of CCUS technologies is slower than anticipated, mainly due to the high costs associated with them (Bui et al., 2018). CO_2 utilisation can greatly facilitate capture cost compensation, as it creates a potential revenue stream for the CCUS network. The most common CO_2 utilisation option in published literature is enhanced oil recovery (EOR). Yet, it is still under debate, as it induces the production of more fossil fuels, and it does not satisfy the goals of a circular economy (Chauvy and De Weireld, 2020). Other utilisation options include agricultural products, synthetic fuels, and minerals (Leonzio et al., 2020). Studying each CCUS subprocess separately is not sufficient to understand the economy of such networks. In the selection of the most cost-effective technological options and the development of the highest-performing network structures, it is crucial to consider a holistic approach encompassing the entire CCUS chain (Leonzio et al., 2020). Mathematical programming enables the optimisation of such complex and scaled-up CCUS infrastructures and is a useful tool for decision-makers, especially in the early stages of the deployment of CCUS technologies (d'Amore and Bezzo, 2017). D'Amore and Bezzo (2017) developed a Mixed-Integer Linear Programming (MILP) framework for the strategic development of a European network for CO_2 capture and

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storage (CCS). The objective was to minimise the total cost of the network, while multiple CO_2 capture and transportation options were included. Al-Mohannadi and Linke (2016) proposed a multi-step approach to the systematic design of CCUS networks for industrial parks. A continuous optimisation method was adopted in the final step, including a variety of utilisation options. Hasan et al. (2014) presented a multi-scale MILP framework for CCUS aiming to maximise the profits from EOR. Several material, process, and supply chain design options were integrated. The employed costing method was based on input-output models that were extracted from detailed Aspen Plus® models for each subprocess. Ostovari et al. (2023) proposed a supply chain network for CCUS by mineralisation, where all captured CO_2 is converted into minerals that are either used as products or stored in abandoned mines. The objective was to minimise the total annual cost (TAC) of the network. Their results showed that the cost of CCUS by mineralisation could be comparable to traditional CCUS when implemented in a network. Although these works offer advancements in CCUS network design and optimisation, they lack detailed techno-economic analyses, as their cost assessment for each CCUS subprocess is mostly based on data from Intergovernmental Panel on Climate Change (IPCC) reports (Metz et al., 2005). Also, utilisation and mineralisation options are either limited within previously studied technological choices (e.g., EOR, agricultural products) or absent.

The scope of this work is to develop a MILP framework that will provide an optimal design of a CCUS network within an industrial cluster. CO_2 capture, mineralisation, utilisation, compression and pumping, transportation via pipeline, and sequestration subprocesses are incorporated into the framework. The economic evaluation of the CCUS subprocesses is based on data from precise techno-economic and process models that are optimised for different values within the operating range. The product from CO_2 utilisation is in the form of precipitated calcium carbonate (PCC) nanoparticles and is responsible for the revenue of the network. Carbonate nanoparticles are gaining increasing attention, and they are a growing market as they find various emerging applications in many industrial sectors (Nessi et al., 2022). To the authors' knowledge, this is the first time that CO_2 utilisation through PCC nanoparticle production in a rotating packed bed (RPB) and CO_2 mineralisation directly from flue gas for capture are integrated within a CCUS network design framework. The framework's objective is to minimise the TAC of the whole network while decreasing the cluster's emitted CO_2 by 90 %.

2. Methodology

2.1 Description of the framework

This work proposes a design framework for cost-optimal CCUS industrial cluster networks while using cost data from optimised, rigorous models for each network subprocess. The framework tests two novel processes, the RPB-based PCC nanoparticle production as a CO_2 utilisation option and the slurry-based CO_2 mineralisation for capture using directly the plants' flue gas. Figure 1 shows the flowsheet of the CCUS network to be designed.

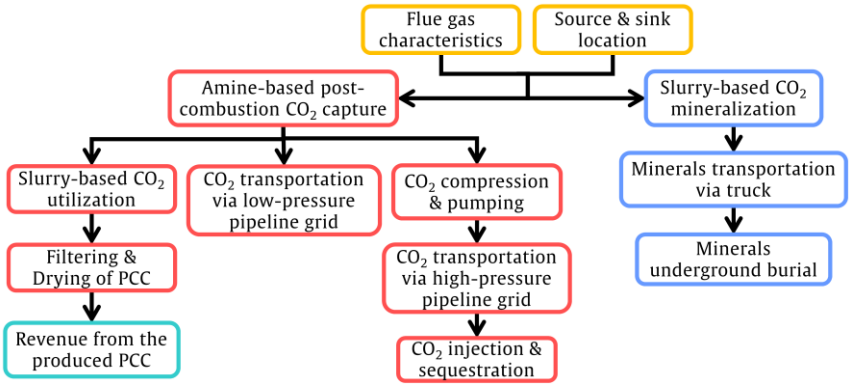


Figure 1: CCUS network flowsheet

The framework takes as input the flue gas characteristics and the location of each industrial emitter, as well as the location of the sequestration and mineral deposit sites. It is assumed that CO_2 capture, utilisation, compression and pumping, mineralisation, and PCC filtering and drying can take place only in any of the emitter plant sites. The decision framework determines for each emitter the selected technology for the treatment of the flue gas. The two choices are calcium hydroxide-$Ca(OH)_2$, slurry-based, CO_2 mineralisation and amine-based post-combustion CO_2 capture. The first technology employs an RPB reactor to intensify the reaction of CO_2 contained in the flue gas with $Ca(OH)_2$ slurry. The reaction produces calcium carbonate-$CaCO_3$, which is stable and non-hazardous for the environment. After this stage, the $CaCO_3$ aqueous solution is left to dry, and then it can be transported via trucks for underground deposit. Common sites for the deposit are depleted mines. The

second technology uses packed-bed (PB) absorption/desorption and an aqueous MEA solution for the separation of CO_2 from the flue gas. After the separation, the exit stream contains almost pure CO_2. There are three choices at this stage: CO_2 conversion to products, transportation to a nearby emitter's plant site, and compression and pumping. CO_2 conversion takes place using the same technology as that used in the CO_2 mineralisation case but in a controlled mode to target the desired particle size distribution; however, it uses the pure CO_2 stream instead of the flue gas. The product of this process is high-quality PCC nanoparticles free from impurities, which are a value-added product with various industrial applications, and usually, it is commercially valued in the range of 1,000 to 2,000 €/t. Filtering and drying stages are required for the removal of moisture from the PCC crystals. CO_2 transportation via a low-pressure pipeline grid pertains to the interconnection between the industrial emitters to either transport CO_2 for utilisation in another emitter's site or to inject CO_2 into the high-pressure pipeline grid. The transportation is performed with the CO_2 in the gaseous phase, as it is not cost-effective to liquefy it for short distances. Finally, compression and pumping pertain to the increase of the pressure of the pure CO_2 stream up to a critical point where it turns into liquid and is then pumped to further increase its pressure. Transportation via the high-pressure pipeline grid is favourable for long distances, as the density of CO_2 is higher compared to its gaseous phase. Finally, when the CO_2 stream reaches the sequestration site, it is injected through wells into the underground cavity, which may be a saline aquifer or a depleted oil or natural gas reservoir, for permanent storage.

2.2 Optimisation problem formulation

The decision variable vector (X) of the MILP optimisation problem consists of N variables with the name CC which denote the amine-based CO_2 capture plants, N variables with the name MIN which denote the mineralisation-based capture plants, N variables with the name CP which refer to the CO_2 compression and pumping, $N \times (N-1) + S \times N$ variables with the name $PIPE$ and $PIPE_{seq}$ which denote the CO_2 transportation via pipeline, $N \times G$ variables with the name $TRUCK$ which denote the mineral transportation via truck from each of the N industrial emitter sites to each of the G mineral deposit sites, G variables with the name GEO which refer to the mineral underground deposits, $S + l$ variables with the name SEQ which refer to the CO_2 sequestration, N variables with the name CU which refer to the CO_2 utilisation, N variables with the name FD which refer to the PCC filtering and drying, N variables with the name PCC which refer to the PCC produced by each one of the N CO_2 utilisation subprocesses, and $S + l$ binary variables (i_{SEQ}) related to the CO_2 sequestration. It must be mentioned that N is the number of industrial emitters, G is the number of mineral underground deposit sites, S is the number of sequestration sites and l is the number of extra sequestration variables needed to capture the process behaviour. For the transportation of CO_2 via pipeline, $N \times (N-1)$ variables, named $PIPE$, refer to the interconnection between the industrial emitter sites (low-pressure pipeline grid), and $S \times N$ variables, named $PIPE_{seq}$ refer to the connection of each industrial emitter site with each sequestration site (high-pressure pipeline grid). For the sequestration subprocess, the total number of variables is $2 \times (S + l)$. This is due to the nonlinear behaviour of the process, which is approximated by piece-wise linear models. All variables of vector X express the mass flow rate of CO_2 that enters the corresponding network subprocess (t/d), except $TRUCK$, GEO, FD, and PCC that express the mass flow rate of PCC (t/h), and i_{SEQ} that are binary.

The objective function to be minimised (OF) is expressed as the sum of all the cluster subprocesses' TAC minus the revenue generated by the produced PCC (Eq(1)). The constraints of the problem are presented in set of Eq(2) and express the mass balances of CO_2, minerals, and PCC for each subprocess and node of the network.

$$\min_{X} \quad OF = \sum_{i=1}^{N} TAC_{CC_i} + \sum_{i=1}^{N} TAC_{MIN_i} + \sum_{i=1}^{N} TAC_{CP_i} + \sum_{i=1}^{N} \sum_{j=1, i \neq j}^{N-1} TAC_{PIPE_{i,j}} + \sum_{i=1}^{N} \sum_{j=1,}^{S} TAC_{PIPE_{seq,i,j}} + \sum_{i=1}^{N} \sum_{j=1}^{G} TAC_{TRUCK_{i,j}} + \sum_{i=1}^{G} TAC_{GEO_i} + \sum_{l=1}^{S} \sum_{j=1, for\ i \neq 3}^{4} TAC_{SEQ_{i,j}} + \sum_{k=1}^{S} \sum_{j=1, for\ i \neq 3}^{4} TAC_{i_{SEQ_{k,j}}} + \sum_{i=1}^{N} TAC_{CU_i} + \sum_{i=1}^{N} TAC_{FD_i} - \sum_{i=1}^{N} REV_{PCC_i} \tag{1}$$

$$\begin{aligned}
&CC_i + MIN_i = COM_i \quad , i \in (1, N) \\
&f_{PCC}(MIN_i) - \sum_{j} TRUCK_{i,j} = 0 \quad , i \in (1, N)\ and\ j \in (1, G) \\
&\sum_{i} TRUCK_{i,j} - GEO_j = 0 \quad , i \in (1, N)\ and\ j \in (1, G) \\
&PIPE_{i,j} + PIPE_{j,i} = 0 \quad , i \in (1, N), j \in (1, N), i \neq j \\
&f_{PCC}(CU_i) - FD_i = 0 \quad , i \in (1, N)
\end{aligned} \tag{2}$$

$$\sum_j SEQ_{i,j} - \sum_k PIPE_{seq,k,i} = 0 \quad , i \in (1,S), j < l, and\ k \in (1,N)$$

$$CP_i - \sum_j PIPE_{seq,i,j} = 0 \quad , i \in (1,N)\ and\ j \in (1,S)$$

$$CC_i \cdot f_{CAPT} - CU_i - CP_i + \sum_j PIPE_{j,i} = 0 \quad , i \in (1,N), j \in (1,N), i \neq j$$

$$\sum_i CU_i - f_{LIME}(CAP_{Ca(OH)_2}) \leq 0 \quad , i \in (1,N)$$

$$PCC_i - FD_i = 0 \quad , i \in (1,N)$$

$$0 \leq \sum_j i_{SEQ_{i,j}} \leq 1 \quad , i \in (1,S)\ and\ j < l$$

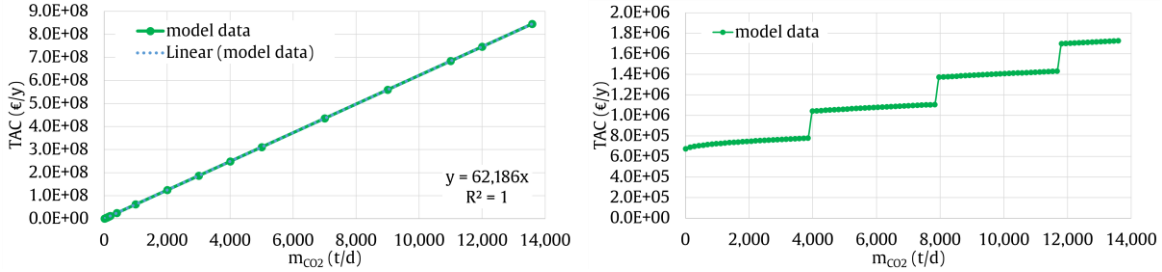
$$0 \leq SEQ_{i,j} \quad , i \in (1,S), j < l$$

$$SEQ_{k,j} - M_h \cdot i_{SEQ_{k,j}} \leq 0 \quad , k \in (1,S), j < l, and\ h \in (1, S + l)$$

where f_{PCC} is the function that correlates the amount of CO_2 that enters in either of the mineralisation or utilisation subprocesses (t/d) with the amount of minerals or PCC that will be produced (t/h), f_{CAPT} is the CO_2 capture efficiency goal, f_{LIME} is the function that correlates CO_2 emissions with the available $Ca(OH)_2$ from the quicklime plant, $CAP_{Ca(OH)_2}$ is the maximum available $Ca(OH)_2$ to be used for PCC production, and M constants are the "big-M" multipliers. The value of M in each constraint is chosen so that it represents the upper limit in the respective independent variable range it refers to.

2.3 Process models

All CCUS network subprocess design and costing is performed using data through optimised rigorous process models. For CO_2 capture, the model using MEA and the conventional PB configuration is attained from Damartzis et al. (2014). For CO_2 mineralisation and utilisation, the model tested in Prousalis et al. (2023) is used. For CO_2 compression and pumping, transportation via pipeline, and sequestration, a detailed analysis by McCollum and Ogden (2006) is adopted. Data for sequestration sites in Greece are attained by Koukouzas et al. (2011). Finally, for the PCC filtering and drying, the cost estimation is performed using the techniques in Walas (1988). All cost values for electricity and equipment purchases are updated to recent values. Figure 2 shows the linear and piece-wise linear models derived for the utilisation and sequestration processes.



(a) (b)

Figure 2: a) Model data and linear function for CU process, b) Model data for CO_2 sequestration in site 2

The linear equations are cost-related in most cases, except for functions f_{PCC} and f_{LIME}. The equations are extracted through linear regression on model data that are optimised for different CO_2 or flue gas compositions and flow rates or different PCC or minerals flow rates, covering the entire operating range for each CCUS subprocess. In most cases, the linear equations present great agreement with the model data (Figure 2a), leading to the assumption that there is no need for a more complex optimisation method. In the cases where the trade-offs are nonlinear, the behaviour is approximated by piece-wise linear functions (Figure 2b).

2.4 Implementation

For the case studies, 5 industrial emitters (N), one mineral deposit site (G), 3 sequestration sites (S), and 6 extra sequestration process variables (l) are considered, leading to a total of 89 decision variables in the vector X. The industrial emitters' flue gas characteristics are presented in Table 1. The cluster consists of a quicklime plant, a cement plant, a pulp and paper plant, a natural gas power plant, and a refinery. The total treated CO_2 in the cluster is 5.5 Mt/y. Maximum $Ca(OH)_2$ availability is restricted to 20 % of the annual capacity of the cluster's quicklime plant, while the capacity is estimated assuming that 1.2 tons of CO_2 are emitted for the production of a ton of $Ca(OH)_2$ (Simoni et al., 2022). Finally, it must be mentioned that both CO_2 capture, utilization, and mineralization processes are designed to achieve 90 % CO_2 conversion efficiency.

Table 1: Industrial emitter's flue gas composition and flow rates

Type	Total flow rate (mol/s)	CO_2 (vol. %)	H_2O (vol. %)	N_2 (vol. %)	CO_2 flow rate (Mt/y)	Source
Quicklime	320	12.3	12.5	75.5	0.055	Kazepidis et al. (2021)
Cement	9,922	16.5	13.2	70.3	2.272	Gerbelová et al. (2017)
Pulp & Paper	4,600	13.3	19.0	67.7	0.849	Gardarsdottir et al. (2014)
NG Power	17,675	4.1	12.5	83.0	1.106	Kazepidis et al. (2021)
Refinery	11,218	7.9	14.9	77.2	1.230	Nazerifard et al. (2023)

3. Results and discussion

3.1 Case study A: Without revenue from PCC

In case study A, the framework is tested without considering any revenue. The algorithm terminated successfully after finding the optimal solution. The cost-optimal CCS network presents the total cost per ton of avoided CO_2 as equal to 76.9 €/t. Comparing this to the current carbon permit cost under EU ETS of 82.9 €/t (end of 2023), the CCS network offers a 7.2 % lower cost per ton of CO_2. The decision variable results are presented in Table 2. The results show that the CO_2 from each industrial emitter is captured on site, then compressed, and finally transported by each one of them separately to sequestration site 2, which is the nearest site to the cluster. The transportation of CO_2 between the industrial emitters or any other sequestration site was avoided. Also, the use of the mineralisation for the capture subprocess and its after-treatment chain was avoided.

Table 2: Optimal network routes for case study A

Decision variable	Value (t/d)	Decision variable	Value (t/d)	Decision variable	Value (t/d)	Decision variable	Value (t/d)
CC_1	150.68	CP_1	135.62	$PIPE_{seq,1,2}$	135.62	$SEQ_{2,4}$	13,591.23
CC_2	6,224.65	CP_2	5,602.19	$PIPE_{seq,2,2}$	5,602.19	$i_{SEQ_{2,4}}$	1
CC_3	2,326.02	CP_3	2,093.42	$PIPE_{seq,3,2}$	2,093.42	-	-
CC_4	3,030.13	CP_4	2,727.12	$PIPE_{seq,4,2}$	2,727.12	-	-
CC_5	3,369.86	CP_5	3,032.88	$PIPE_{seq,5,2}$	3,032.88	-	-

3.2 Case study B: With revenue from PCC

In case study B, a price of 1,000 €/t of PCC is considered. The algorithm terminated successfully after finding the optimal solution once again. The total cost per ton of avoided CO_2 for the designed CCUS network is 73.9 €/t. In this case, the cost per ton of CO_2 is 10.9 % lower than the carbon permit price and 3.9 % compared to the CCS network configuration. The decision variables' values are presented in Table 3. The results indicate that the revenue from PCC is so strong, as all available $Ca(OH)_2$ is exploited in the utilisation process. Once again, no use of the low-pressure interconnection grid or the mineralisation for the capture process takes place.

Table 3: Optimal network routes for case study B

Decision variable	Value (t/d)	Decision variable	Value (t/d)	Decision variable	Value (t/d)	Decision variable	Value (t/d)	Decision variable	Value (t/h)
CC_1	150.68	CP_1	135.62	$PIPE_{seq,1,2}$	135.62	$SEQ_{2,4}$	13,575.61	FD_2	1.33
CC_2	6,224.65	CP_2	5,586.58	$PIPE_{seq,2,2}$	5,586.58	$i_{SEQ_{2,4}}$	1	PCC_2	1.33
CC_3	2,326.02	CP_3	2,093.42	$PIPE_{seq,3,2}$	2,093.42	CU_2	15.62	-	-
CC_4	3,030.13	CP_4	2,727.12	$PIPE_{seq,4,2}$	2,727.12	-	-	-	-
CC_5	3,369.86	CP_5	3,032.88	$PIPE_{seq,5,2}$	3,032.88	-	-	-	-

4. Conclusions

A framework for the optimal design of CCUS networks within industrial clusters was developed. Advanced process and techno-economic models for CO_2 capture, utilisation, mineralisation, compression and pumping, transportation, and sequestration were integrated into the framework through regressed linear and piece-wise linear functions. The results showed that the traditional CO_2 capture-compression-transportation-sequestration chain is more cost-effective than CO_2 mineralisation for capture. In the case studies, the optimal CCS and CCUS networks achieved a total cost per ton of avoided CO_2 equal to 76.9 €/t and 73.9 €/t. These options are favourable compared to purchasing carbon permits with the current price, as they offer reduced costs by 7.2 % and 10.9 %. Future work will involve the integration of different CCUS subprocess technologies, as well as analysis with constraints in raw materials availability and product demand, including uncertainty.

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