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# Optimal Integration of Polygeneration with Carbon Dioxide Removal

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Deep reduction in industrial greenhouse gas emissions can be achieved through engineering measures such as energy efficiency enhancement, fuel switching, and carbon dioxide removal (CDR). Integrated energy systems like polygeneration plants are inherently efficient, while partial or total replacement of fossil fuels with renewables allows further cuts to be realized. Novel CDR technologies can also be used to generate negative emissions. In this work, we develop an optimization model for optimizing a novel polygeneration system which integrates CDR based on ex situ enhanced rock weathering. The latter relies on exposing pulverized rock to accelerate naturally occurring geochemical reactions of minerals with ambient carbon dioxide and water, and results in the sequestration of carbon as bicarbonate ions in water. Integration with a polygeneration plant allows surplus electricity to be utilized for the energy-intensive rock grinding process, as an alternative to direct energy storage. A mixed-integer linear programming enterprise input-output (MILP-EIO) model is developed and then applied to a case study on design and operation problem. The objective is to determine the optimal design of a zero emissions polygeneration system which is economically feasible. Results indicate that such a result is only possible once $CO_2$ price reaches at least US$ 50 /t.

## 1. Introduction

Mitigating climate change will require the drastic reduction of anthropogenic greenhouse gas (GHG) emissions to net-zero by mid-century (IPCC, 2022). This goal can be achieved through the deployment of different decarbonization measures, such as increased use of renewables and optimization of energy efficiency (Klemeš, 2022). For example, technologies such as polygeneration offer the prospect of meeting growing energy needs within sustainable limits (Jana et al., 2017). However, to offset both historical GHGs already in the atmosphere and the residual emissions from persistent use of fossil fuels, carbon dioxide removal (CDR), also known as negative emissions technologies (NETs), will also be needed (Haszeldine et al., 2018). NETs rely on different chemical, physical, and biological mechanisms to remove $CO_2$ from the atmosphere and store it or its carbon content in another environmental compartment (McLaren, 2012), thus reversing the normal transfer of carbon into the atmosphere due to human activities. A survey of different NET alternatives can be found in the review paper by Minx et al. (2018). As environmental footprints are critical for gauging the sustainability of technologies (Čuček et al., 2012), these metrics have also been applied to assess the large-scale use of NETs (Smith et al., 2016). Process Integration (PI) tools such as Mathematical Programming (MP) models have been proposed to optimize NET portfolios (Migo-Sumagang et al., 2021).

Enhanced weathering (EW) as a NET was originally proposed by Seifritz (1990) and is projected to be capable of removing 300 Gt $CO_2$ by 2100 (Strefler et al., 2018). It relies on the artificial acceleration of natural geochemical weathering reactions of alkaline minerals with $CO_2$ and water. Laboratory experiments in the late 1990s demonstrated the viability of the EW concept (Kojima et al., 1997). There is an abundant supply of alkaline material, including rocks, minerals, and industrial waste, that can be used to capture $CO_2$ at a scale measurable in multiple Gt/y (Renforth, 2019).

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In ex situ EW, alkaline rocks and minerals are reduced to a fine powder to increase the reactive surface area when exposed to the elements. These powders are transported and applied to terrestrial (Beerling et al., 2020) or coastal (Meysman and Montserrat, 2017) application sites at a rate calibrated to match local weather (e.g., precipitation and ambient temperature) conditions (Strefler et al., 2018). The reaction of alkaline minerals with dilute carbonic acid in water forms dissolved bicarbonate ions that are ultimately carried into the ocean for long-term sequestration of the embedded carbon.

A recent large-scale NET portfolio optimization study highlights the advantage of EW over competing NETs when land footprint constraints are considered (Strefler et al., 2021). However, social acceptability may become a significant factor in the eventual commercialization of EW (Spence et al., 2017). Alternative EW-based concepts have also been proposed, including dusting of remote ecosystems with powdered rock (Goll et al., 2021), integration with urban farming (Haque et al., 2021), and closed-circuit mineral looping (McQueen et al., 2020). There is also a potential for integrating EW systems with electricity generation to manage the large energy requirement for grinding rocks and minerals (Renforth, 2012), but this concept has not been fully explored. There have been few studies on the development of dedicated MP models for integrated EW systems. The most notable of which are the supply chain-like EW Carbon Management Network (EW-CMN) models first proposed by Tan and Aviso (2019). A significant research gap is indicated by the limited number of such works.

To address this research gap, a Mixed-Integer Linear Programming (MILP) model is developed in this work for the optimal synthesis of polygeneration systems integrated with EW for carbon sequestration. The model is based on the classic MILP proposed by Grossmann and Santibanez (1980) for generic process synthesis problems. The rest of this paper is organized as follows. Section 2 gives the formal problem statement, while Section 3 gives the MILP model formulation. Section 4 illustrates the use of the model with a polygeneration case study. Finally, Section 5 presents the conclusions and briefly discusses directions for future work.

## 2. Problem statement

There are *m* number of processes being considered for the integrated polygeneration plant and *n* number of material or energy streams. Each process has known fixed input-output ratios which represent process efficiency; each process has an associated cost for integration into the system represented by its variable and fixed cost coefficients; each material or energy stream has an associated price or cost depending on whether it is being consumed or generated from the system; there are known external demand limits for identified material or energy streams. The problem is to determine which processes should be integrated into the system to maximize annual profit and meet exogenously defined stream demands.

## 3. MILP model formulation

The objective is to maximize the annual profit of the integrated polygeneration plant with CDR as indicated in Eq(1). The profit is calculated using Eq(2) which accounts for the revenues generated from the sale of product streams, costs incurred from raw material or energy inputs, and the annualized capital costs from chosen processes. AWH represents the annual working hours, $y_i$ corresponds to the net output of material or energy stream i, $c_i$ is the associated cost for each material or energy stream i, $AF$ is the annualizing factor, $b_j$ is a binary variable which indicates whether process j is selected ($b_j = 1$) or not ($b_j = 0$), $FC_j$ refers to the fixed cost of process j, $VC_j$ refers to the variable cost of process j, and $x_j$ corresponds to the capacity of process j. Eq(3) represents the material and energy balance equation where $a_{ij}$ is the technical coefficient for stream i in process j, $a_{ij}$ will have a negative value if it is an input to the process but will have a positive value if it is an output of process j. Eq(4) ensures that material or energy stream i will be within defined lower ($y_i^L$) and upper ($y_i^U$) limits. Eq(5) activates the binary variable $b_j$ once process j has a required capacity, $x_j$. Finally, Eq(6) defines the $b_j$ to be a binary variable. In addition, material or energy streams which are not externally acquired by the system should be non-negative.

$$\max \text{Profit} \tag{1}$$

$$\text{Profit} = AWH \sum_{i=1}^{n} y_i c_i - AF \left( \sum_{j=1}^{m} b_j FC_j + \sum_{j=1}^{m} x_j VC_j \right) \tag{2}$$

$$y_i = \sum_{j=1}^{m} a_{ij} x_j \qquad \forall i \in N \tag{3}$$

$$y_i^L \le y_i \le y_i^U \qquad \forall i \in N \tag{4}$$

$$x_j \leq b_j M \qquad \forall j \in M \qquad (5)$$

$$b_j \in \{0,1\} \qquad \forall j \in M \qquad (6)$$

This static MILP model is like the one presented by Sy et al. (2018), which is in turn based on the generic formulation of Grossmann and Santibanez (1980). Solving this model presents no significant computational issues, as the global optimum can be readily found using standard branch-and-bound solvers embedded in modern spreadsheet applications and commercial optimization software. Alternative optimal and near-optimal solutions can also be generated for evaluation by adding integer cut constraints (Voll et al., 2015). The use of this model is illustrated in the next section.

## 4. Case study

In this case study, the MILP is implemented in the commercial software LINGO (Schage, 1999) using a laptop with Intel® Core™ i7-6500U CPU at 2.50GHz. Computational time to reach the global optimum was negligible. The case study considers five processes to be included in the integrated natural gas-fired polygeneration system. These processes include a cogeneration module (P1), a boiler (P2), a hot water generator (P3), a steam-water heat exchanger (P4), and a rock crusher (P5) which pulverizes rock for EW application purposes. P5 needs electricity to grind the rock to a particle size of ~10 μm to accelerate weathering to a useful rate (Renforth, 2012). The ground rock is then applied to soil where it reacts with ambient $CO_2$ and water. It is assumed that each t of pulverized rock will absorb 0.85 t of $CO_2$ when used for EW (Moosdorf et al., 2014), even after considering penalties for mining, crushing, and transportation. The technical coefficients and technoeconomic data for processes P1 to P4 were obtained from Sy et al. (2018) while data for P5 were obtained from Woods (2007). Process P1 to P3 have associated $CO_2$ emissions from the combustion of natural gas; P5, on the other hand, "consumes" $CO_2$ via the downstream negative carbon footprint of the powdered rock. Technical coefficients are summarized in Table 1, where negative entries indicate an input to a process and positive ones indicate an output from a process. The cost coefficients are indicated in Table 2. Note that a negative price is indicated for the stream of $CO_2$, which means that positive net emissions of $CO_2$ will be considered as a cost to the system. The limits for the material and energy streams are shown in Table 3, where negative entries denote materials which are sourced externally from the system. For this case study, natural gas and rock are sourced from external suppliers. The lower limit then indicates the maximum amount that can be obtained from the external source. For this case, there is unlimited supply of natural gas, while there is a maximum limit of 50 t/h for the rock (i.e., lower limit for the rock is −50 t/h). For both inputs, the upper limits are set to 0 (since a positive value would indicate a net system output). The superstructure of the polygeneration plant with EW is illustrated in Figure 1.

*Table 1: Technical coefficients of processes in integrated polygeneration plant with EW*

| | | P1 | P2 | P3 | P4 | P5 |
|---|---|---|---|---|---|---|
| Natural Gas | MW | −4.06 | −1.20 | −1.08 | 0.00 | 0.00 |
| Steam | MW | 1.83 | 1.00 | 0.00 | −1.00 | 0.00 |
| Hot Water | MW | 0.53 | 0.00 | 1.00 | 1.00 | 0.00 |
| Electricity | MW | 1.00 | 0.00 | 0.00 | 0.00 | −0.18 |
| Rock | t/h | 0.00 | 0.00 | 0.00 | 0.00 | −1.00 |
| $CO_2$ | t/h | 0.89 | 0.26 | 0.24 | 0.00 | −0.85 |

*Table 2: Cost associated with integrated polygeneration plant with EW (in US$)*

| Process | Fixed Cost | Variable Cost | Stream | Price |
|---|---|---|---|---|
| P1 | 382,500 | 948,347 /MW | Fuel | 20 /MW |
| P2 | 45,500 | 175,000 /MW | Steam | 40 /MW |
| P3 | 7,500 | 39,474 /MW | Hot Water | 30 /MW |
| P4 | 625 | 4,688 /MW | Electricity | 90 /MW |
| P5 | 23,885 | 300,600 /(t/h) | Rock | 25 /t |
| | | | $CO_2$ | −50 /t |

It is assumed that the system operates at 8,000 h/y and that the annualizing factor (AF) is 0.08. Solving Eq(1) subject to the constraints in Eq(2) to Eq(6) results in an annual profit of US$ 4.68 million/y, with the optimal network illustrated in Figure 2. The optimal network only selects processes P1, P2, P3, and P5. Steam is generated by P1 and P2, hot water is supplied by both P1 and P3, and electricity is generated using P1. P5 will require 24.39 t/h of rock to remove all the $CO_2$ generated by P1, P2, and P3. The selection of P5 also results in an increase for demand of electricity. The net amount of steam and electricity generated reached the upper demand limit defined while the amount of hot water generated is at the lower limit.

*Table 3: Material and energy stream limits*

| Stream | Units | Lower limit ($y_i^L$) | Upper limit ($y_i^U$) |
|---|---|---|---|
| Fuel | MW | N/A | 0 |
| Steam | MW | 25 | 50 |
| Hot Water | MW | 15 | 30 |
| Electricity | MW | 5 | 10 |
| Rock | t/h | -50 | 0 |
| $CO_2$ | t/h | 0 | N/A |

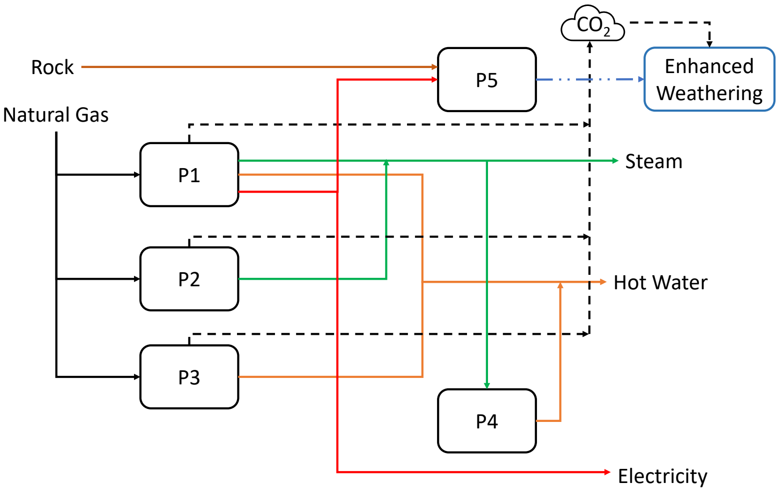

*Figure 1: Superstructure of the polygeneration plant with EW indicates all possible structures for generating the desired products*

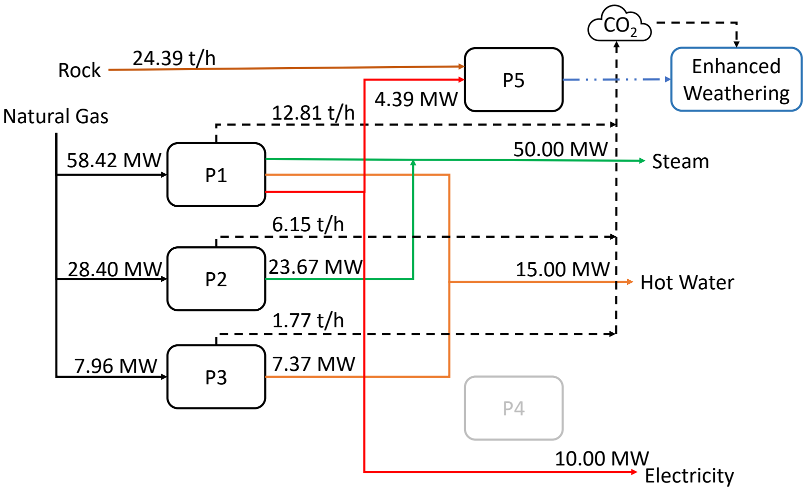

*Figure 2: Optimal carbon neutral network structure for $CO_2$ price at US$ -50/t integrates enhanced weathering in the solution*

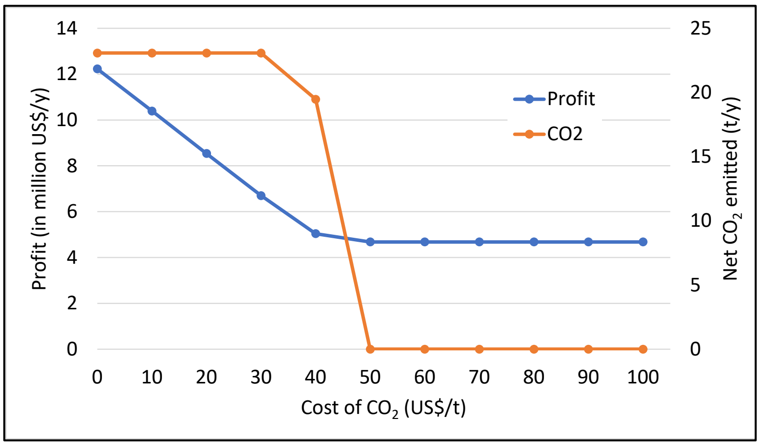

*Figure 3: Change in profit and net $CO_2$ emitted as a function of $CO_2$ cost shows that net zero emissions is achieved at $CO_2$ price of at least US$ 50/t*

A sensitivity analysis was then conducted to determine how variations in $CO_2$ cost will affect the integrated polygeneration system in terms of profit and $CO_2$ emitted. The optimal solution changes are presented in Figure 3 where the cost of $CO_2$ is varied in the range from US$ 0/t to US$ 100 /t. EW is not selected at low $CO_2$ price levels of up to US$ 40 /t. However, once the $CO_2$ price reaches US$ 50 /t, the optimal system design includes the rock crusher for EW, and the entire system becomes carbon neutral. The sensitivity analysis shows that carbon pricing significantly affects the sustainability of the proposed CDR system.

## 5. Conclusions

A mixed-integer linear program for the design and optimization of an integrated polygeneration system with CDR has been developed in this work. This demonstrates the techno-economic feasibility of implementing EW together with polygeneration systems to further eliminate $CO_2$ emissions. The polygeneration plant and EW system complement each other since the polygeneration plant can provide the power needed by the rock crusher, while EW neutralizes any $CO_2$ emission generated by the polygeneration plant. However, GHG reduction targets cannot be met unless the $CO_2$ removal has economic value via carbon tax or credits. Future work can investigate extending the system boundary to integrate emissions resulting from transport processes and other phases of the supply chain. Other potential risks to the ecosystem and an examination of ethical issues potentially surrounding this technology should also be investigated in future studies. Additionally, multi-objective optimization models which simultaneously consider economic, environmental, and social aspects of the technology can be developed.

### Nomenclature

Parameters

AF – annualizing factor
AWH – annual working hours
$a_{ij}$ – input/output of stream i in process j
$c_i$ – cost of stream i
$FC_j$ – fixed capital cost of process j
M – arbitrary big number
$VC_j$ – variable capital cost of process j
$y_i^L$ – lower limit for stream i
$y_i^U$ – upper limit for stream i

Variables

$b_j$ – binary variable for the selection of process j
$x_j$ – capacity of process j
$y_i$ – net output of material or energy stream i

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