

Article

# Techno-Economic and Environmental Evaluations of Decarbonized Fossil-Intensive Industrial Processes by Reactive Absorption & Adsorption CO<sub>2</sub> Capture Systems

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Abstract: Decarbonization of energy-intensive systems (e.g., heat and power generation, iron, and steel production, petrochemical processes, cement production, etc.) is an important task for the development of a low carbon economy. In this respect, carbon capture technologies will play an important role in the decarbonization of fossil-based industrial processes. The most significant techno-economic and environmental performance indicators of various fossil-based industrial applications decarbonized by two reactive gas-liquid (chemical scrubbing) and gas-solid CO<sub>2</sub> capture systems are calculated, compared, and discussed in the present work. As decarbonization technologies, the gas-liquid chemical absorption and more innovative calcium looping systems were employed. The integrated assessment uses various elements, e.g., conceptual design of decarbonized plants, computer-aided tools for process design and integration, evaluation of main plant performance indexes based on industrial and simulation results, etc. The overall decarbonization rate for various assessed applications (e.g., power generation, steel, and cement production, chemicals) was set to 90% in line with the current state of the art in the field. Similar non-carbon capture plants are also assessed to quantify the various penalties imposed by decarbonization (e.g., increasing energy consumption, reducing efficiency, economic impact, etc.). The integrated evaluations exhibit that the integration of decarbonization technologies (especially chemical looping systems) into key energy-intensive industrial processes have significant advantages for cutting the carbon footprint (60-90% specific CO<sub>2</sub> emission reduction), improving the energy conversion yields and reducing CO<sub>2</sub> capture penalties.

**Keywords:** fossil-intensive industrial processes; decarbonization technologies; reactive absorption/adsorption  $CO_2$  capture systems; modeling; simulation; and process integration; techno-economic and environmental assessments

## 1. Introduction

Global warming and climate change are fundamental issues nowadays. In order to significantly reduce global warming for long-term sustainable development, the greenhouse gas emissions (especially the fossil ones) need to be significantly cut and decoupled from economic growth [1]. Along this line, the industrial and transport sectors are facing important modifications and restructuring with the aim of reducing the fossil energy sources as required for the development of an economy with a low carbon footprint. Among industrial applications with significant greenhouse gas emissions, the heat, and power generation, iron and cement production, and various fossil-based chemical systems are the biggest contributors. To illustrate the major importance of these industrial sectors and to consider the



global greenhouse gas emissions one can mention that the coal-based power generation is responsible by more than 10 Gt CO<sub>2</sub> from the 33.1 Gt CO<sub>2</sub> emitted globally in 2018 [2], production of iron and steel counts for about 6% of global CO<sub>2</sub> emissions [3], and the cement production counts for 5% of global CO<sub>2</sub> emissions [4]. Accordingly, the fossil-intensive industrial processes need significant changes in forthcoming years to efficiently contribute to the global effort of reducing carbon emissions.

As possible technical and scientific options to cut the fossil carbon emissions, a broad range of measures can be applied, ranging from promoting renewable ene.g., increasing the energy conversion, and utilization yields too large scale deployment of carbon capture, utilization and storage (CCUS) technologies [5]. These technologies are seen as important options for the medium time horizon to allow a smoothly transition from the current fossil-based economy to a future low carbon one. For the integration of the CO<sub>2</sub> capture process into various energy-intensive applications, several conceptual options are already available and widely evaluated in the literature, e.g., pre-, post- and oxy-fuel combustion methods [6]. Once captured, CO<sub>2</sub> can be used as raw material for various processes (e.g., production of various chemicals and fuels), stored in appropriate geological formations (e.g., saline aquifers) or used for increasing the oil/gas recovery yields [7].

This work is assessing some key fossil fuel-intensive industrial applications in view of energy and cost-effective process decarbonization. The selected industrial applications are power generation, iron and steel production, cement production, as well as producing chemicals which can also be used as decarbonized energy carriers (e.g., hydrogen). The chemical absorption (scrubbing) method [8] and the innovative chemical/calcium looping cycles based on reactive adsorption systems [9] were evaluated as decarbonization technologies. Apart from the carbon footprint reduction, the overall energy conversion yields, as well as other techno-economic and environmental performance indicators, represent important elements in the present evaluations. Similar non-carbon capture plants are also assessed to quantify the various penalties imposed by decarbonization (e.g., ene.g., raw materials, and utility consumptions, overall energy efficiency, main economic factors). The decarbonized plant concepts have a 90% CO<sub>2</sub> capture rate, a value which is in line with assessment methodology of CO<sub>2</sub> capture technologies presented in relevant literature sources, e.g., International Energy Agency Greenhouse Gas Programme (IEAGHG) reports for decarbonization of iron and steel production [3] or cement production [4]. In addition to technical and environmental indicators, the economic impact of process decarbonization is also presented considering key performance indexes.

The selected industrial applications were subject to various technical investigations ranging from the conceptual design of decarbonized plants and evaluation of  $CO_2$  capture unit mass and energy integration analysis, usage of computer-aided tools for process design, and integration to the evaluation of main plant performance indexes based on industrial and simulation results. The key novelty aspect of the presented work is to provide an integrated in-depth techno-economic and environmental evaluation methodology of decarbonized industrial processes.

#### 2. Carbon Capture Technologies for Efficient Decarbonization of Industrial Applications

As decarbonization technologies, two carbon capture methods were assessed in view of integration into fossil-intensive industrial applications. The first option considers a mature technology based on chemical scrubbing employing alkanolamines [10]. This technology represents the conventional option when acid gas removal (e.g.,  $CO_2$ , sulfur compounds, etc.) is required in various chemical industrial processes (e.g., natural gas reforming for ammonia synthesis, sulfur removal from oil refinery, etc.). For this main reason, the reactive gas-liquid absorption technology was considered as potential decarbonization process for the evaluated energy-intensive industrial applications. To illustrate this decarbonization technology, Methyl-DiEthanol-Amine (MDEA) was selected as a chemical solvent. The selection of the MDEA solvent was based on the following key advantages over more conventional Mono-Ethanol-Amine (MEA): higher  $CO_2$  loadings (1 mole  $CO_2$ /mole MDEA vs. up to 0.5 mole  $CO_2$ /mole MEA), higher solution concentration (50% vs. 30%), lower degradation, corrosion and toxicity, better thermal stability, etc. [11]. The overall chemical reaction for MDEA-based decarbonization is presented below:

$$CO_2 + H_2O + MDEA \leftrightarrow MDEAH^+ + HCO_3^-$$
 (1)

For this decarbonization technology, the above global chemical reaction is used in a cycle, as presented in Figure 1. The gas to be decarbonized is treated in the absorption column being put in contact with an MDEA aqueous solution (50% wt.). The loaded (rich) solvent is pumped in a separate column where using heat, the  $CO_2$  is desorbed, and thus, the solvent regenerated. The regenerated (lean) solution is pumped back in the absorption column (some make-up being necessary to cover the solvent losses). The  $CO_2$  is treated for moisture removal and compressed to the final delivery pressure (120 bar) prior to storage/utilization. A key element of this decarbonization technology represents the heat consumption (at the bottom of the desorption column) for  $CO_2$  desorption and solvent regeneration. Currently, for the post-combustion  $CO_2$  capture configurations applied to fossil-based power generation plants (10–15 volumetric percentages of  $CO_2$  content in the gas to be treated), this heat duty is about 3 GJ/t  $CO_2$  [12]. For pre-combustion capture configurations, the heat consumption for solvent regeneration is significantly reduced to about 0.6–0.8 GJ/t due to pressure reduction [13].



Figure 1. CO<sub>2</sub> capture by chemical scrubbing via an absorption-desorption cycle.

The second evaluated decarbonization option makes use of an innovative reactive system based on calcium adsorption (Calcium Looping—CaL). Similar with chemical scrubbing option presented above, the chemical looping cycle can also be used for pre- and post-combustion decarbonization configurations. This technique uses two separate reactors for decarbonization as follow [9]:

The carbonation reactor where the gas to be decarbonized is put in contact with the calcium sorbent for  $CO_2$  capture. The reactions for pre- and post-combustion decarbonization are exhibited below:

Post – combustion capture :  $CO_2 + CaO \leftrightarrow CaCO_3$  (2)

$$Pre-combustion capture: CO + H_2O + CaO \leftrightarrow CaCO_3 + H_2$$
(3)

Calcination reactor when CaCO<sub>3</sub> is thermally disintegrated to CaO and CO<sub>2</sub> according to the next chemical reaction:

$$CaCO_3 \leftrightarrow CaO + CO_2$$
 (4)

The innovative calcium looping method was selected as decarbonization technology for the fossil-intensive industrial applications based on the following reasons: it represents a promising technology in reducing the  $CO_2$  capture energy and cost penalties, possibility to use the spent sorbent (deactivated calcium sorbent) within the main process, sorbent lower cost and large availability, etc. [14]. For this decarbonization technology, the conceptual design is presented in Figure 2. As for gas-liquid absorption decarbonization technology, the calcium looping cycle requires an additional energy input (for the calcination reactor). In the calcination reactor, some sort of fuel (e.g., natural gas, syngas,

coal, etc.) is to be oxy-combusted for providing the required energy input for the calcium carbonate decomposition. Oxygen must be used for combustion in order not to dilute the  $CO_2$  captured stream with nitrogen. Different for gas-liquid decarbonization technology, the CaL cycle is operating at significantly higher temperatures: carbonation ( $CO_2$  fixation) reactor to 500–650 °C and calcination reactor to about 850–1000 °C [15]. These operating conditions enable high-temperature heat recovery with positive consequences on the overall energy conversion yield [16].



Figure 2. CO<sub>2</sub> capture by the calcium-based sorbent looping cycle.

## 3. Conceptual Designs, Main Design Assumptions, and Process Integration Elements

To illustrate the influence of plant decarbonization over the most relevant techno-economic and environmental indexes, some key fossil-intensive industrial applications were considered as follow:

- Cases 1: Coal-based gasification power plants;
- Cases 2: Coal-based super-critical power plants;
- Cases 3: Integrated iron & steel plants;
- Cases 4: Cement production plants.

The evaluated gasification plants consider both pre- and post-combustion decarbonization scenarios is based on the Integrated Gasification Combined Cycle (IGCC) design [17]. The conventional gasification-based power plant design without carbon capture involves a partial oxidation process (with oxygen and steam) of the solid fuel to syngas (mainly a mixture of hydrogen and carbon monoxide). Further, the syngas is treated for sulfur removal in an acid gas removal unit, and the clean gas is used for power generation in a combined cycle gas turbine unit [18].

For the pre-combustion capture, the syngas is decarbonized either by gas-liquid absorption or calcium looping, and the hydrogen-rich stream is then used for power generation (in a combined cycle gas turbine unit) or hydrogen and power co-generation. For the post-combustion capture, the flue gases from the syngas-fueled gas turbine are treated for decarbonization with the same two carbon capture technologies (MDEA-based gas-liquid absorption and calcium looping). The conceptual plant layouts of decarbonized IGCC power plants are shown in Figure 3.

The evaluated coal-based super-critical power plants are based on the conventional state of the art design [19]. The combustion-based power plants involve total oxidation of solid fuel with air. The hot flue gases are then used for steam generation. The steam cycle parameters were selected in line with industrial standards: live steam at 290 bar/582 °C also having two steam reheats at 75 bar/580 °C, and 20 bar/580 °C. The cooled flue gases are treated for particulate matter, NO<sub>x</sub>, and SO<sub>x</sub> removal prior to decarbonization. The two decarbonization technologies analyzed in this paper (MDEA-based chemical scrubbing by gas-liquid absorption and calcium-based gas-solid looping cycle) were evaluated in a post-combustion capture configuration. The conceptual plant layout of the decarbonized super-critical power plant, is presented in Figure 4.



Figure 3. Decarbonized Integrated Gasification Combined Cycle (IGCC) power plant options.



Figure 4. Decarbonized super-critical power plant.

The evaluated decarbonization scenario for iron and steel production considers an integrated steel mill in accordance with the current state of the art [20]. The iron and steel production involve sinter production, iron production (a blast furnace), desulphurization plant, steel production (basic oxygen furnace), and various metallurgical steps. Within an integrated steel mill, there are numerous  $CO_2$  emission sources; this analysis considers the carbon capture for the main ones: captive power and heat (steam) plant, blast furnace and hot stoves, lime and coke production systems [21]. The two decarbonization technologies (MDEA-based chemical scrubbing by gas-liquid absorption and calcium-based gas-solid looping cycle) were evaluated in a post-combustion configuration. The conceptual plant layout of decarbonized iron and steel production system is presented in Figure 5.

The evaluated decarbonization scenario for cement production considers the current conventional design [22]. The cement production involves raw meal production, preheating, calcination (clinker production), and grinder (cement production) steps. The generated CO<sub>2</sub> within the cement production process has two main sources—one from the fuel to be combusted in the calcination step and one from the calcium carbonate decomposition [23]. The two decarbonization technologies (MDEA-based chemical scrubbing and calcium-based gas-solid looping cycle) were evaluated in a post-combustion capture configuration. The conceptual plant layout of the decarbonized cement plant is presented in Figure 6.



Figure 5. Decarbonized integrated steel mill.



Figure 6. Decarbonized cement plant.

Table 1 shows the most important design assumptions of evaluated fossil-intensive industrial applications (power generation, iron, steel, and cement production) to be decarbonized as well as the two CO<sub>2</sub> capture technologies (reactive gas-liquid absorption and calcium looping cycle). More detailed specifications are provided in the reference sources indicated in Table 1. Assumptions were furthermore used for modeling and simulation of assessed case studies. In this respect, ChemCAD software was used as a process flow modeling tool. Then the simulation results were employed to evaluate the most important plant performance indexes (e.g., fuel consumption, ancillary ene.g., and raw materials consumption, plant decarbonization rate, carbon footprint, etc.).

Unit	Design Assumptions			
Fossil fuel (coal) specifications [22]	Ultimate analysis (dry weight percentages): 72.30% carbon, 4.11% hydrogen, 1.69% nitrogen, 7.45% oxygen, 0.56% sulfur, 13.89% ash; Moisture: 8%; Lower heating value: 25.17 MJ/kg			
Gasification power plant [12]	Entrained-flow gasifier with syngas quench Separate H <sub>2</sub> S and CO <sub>2</sub> removal by absorption and adsorption systems Combined cycle power using one M701G2 gas turbine			
Super-critical power plant [17]	290 bar/582 °C and two reheats at 75 bar/580 °C and 20 bar/580 °C 95% NOx removal yield by selective catalytic reduction unit 98–99% SOx removal yield by wet desulphurization unit			
Integrated steel mill [3]	Plant capacity: 4 Mt/y hot-rolled coil (HRC) Decarbonization of power plant, hot stoves, lime and coke production Natural gas-based heat and power unit for ancillary consumptions Captive heat and power plant: subcritical boiler and combined cycle			
Cement plant [4]	Plant capacity: 1 Mt/y cement 95% NOx removal yield by selective catalytic reduction unit 98–99% SOx removal yield by wet desulphurization unit Coal-based heat and power unit for ancillary energy consumptions			
Decarbonization unit employing a chemical scrubbing system [19]	Methyl-diethanol-amine (MDEA) aqueous solution 50% wt. Absorption/desorption columns: 42–55 °C/115–125 °C 90% flue gas decarbonization rate (pre- and post-combustion) Solvent regeneration: thermal using LP steam at 140–150 °C			
Decarbonization unit employing a Ca-based sorbent system [19]	Natural limestone as calcium-based sorbent Carbonation/calcination reactors: 540–615 °C/825–975 °C 90% flue gas decarbonization rate (pre- and post-combustion) Oxy-fuel combustion system to provide heat for sorbent regeneration Power consumption for oxygen production unit: 200 kWh/t			
CO <sub>2</sub> conditioning unit [14]	Four compression stages with 120 bar final pressure at plant gate Moisture removal by gas-liquid absorption using Tri-Ethylene-Glycol (TEG) $CO_2$ composition (volume percentages): >95% $CO_2$ , <2000 ppm $CO$ , <200 ppm $H_2O$ ,<50 ppm $H_2S$ , <4% other gases			

Table 1	. Main	design	elements	of eva	aluated	decarbo	onized	industrial	processes.

The evaluated carbon capture designs were assessed in view of heat and power integration analysis for optimization of overall energy conversion yield [24]. In this respect, Pinch Analysis was used for Heat Integration of hot and cold streams within the plant. The main focus of Heat Integration analysis of the two carbon capture technologies was to enhance heat recovery potential by process-to-process heat exchange and to reduce external hot and cold utility consumptions [25]. To show the fundamental advantage in terms of high-temperature heat recovery of calcium-based gas-solid looping cycle over the chemical scrubbing option, Figure 7 presents the Hot and Cold Composite Curves for CaL cycle used for super-critical power plant decarbonization [16].



Figure 7. Calcium looping cycle Heat Integration analysis.

In contrast with the calcium looping cycle, the reactive gas-liquid absorption cycle has low-temperature hot streams (40–60  $^{\circ}$ C); therefore, the available heat cannot be used in an

energy-efficient manner (e.g., for steam generation) but only to be taken by cooling water (external cooling utility) [11]. It can be observed that the high-temperature heat recovery capacity of the CaL unit significantly improves the overall energy conversion yield. In fact, the CaL unit can be seen not only as a carbon capture system but also as an energy conversion system since additional fuel (coal) is oxy-combusted with the goal to provide the heat input for CaCO<sub>3</sub> decomposition.

## 4. Techno-Economic and Environmental Assessment Methodology

This section of the paper is dedicated to present the overall techno-economic and environmental plant performance indicators. All evaluated decarbonized energy-intensive industrial concepts were mathematically modeled and simulated using ChemCAD software. The most important simulation data (as mass and energy balances, fuel conversion yields, overall plant decarbonization rate, etc.) were benchmarked against industrial data for model validation; e.g., for the gasification [18] and combustion [19] power plant concepts, for the integrated steel plants [3], for the cement production plants [4], for the alkanolamines-based carbon capture processes [26] and for the calcium looping-based carbon capture [27] and its integration into various energy systems [28]. No significant differences between simulation results and literature data were noticed/registered.

After validation of the simulation results, the most relevant techno-economic and environmental plant performance indexes were calculated using the following equations:

*Gross/net* power conversion efficiencies show the energy conversion rates for the gasification and combustion power plants:

$$\eta_{gross/net} = \frac{Gross / net \ electricity \ output \ [MW_e]}{Thermal \ energy \ of \ used \ fuel \ (coal) \ [MW_{th}]} \times 100$$
(5)

Ancillary power consumption was calculated considering all electricity consumptions of various plant sub-systems:

Ancillary power consumption = 
$$\sum$$
 Plant units power consumption (6)

Plant decarbonization rate (noted as CO<sub>2</sub> Capture Rate—*CCR*) takes into account the percentage of feedstock carbon that was captured:

$$CCR = \frac{Sequestered \ carbon \ molar \ flow \ [kmole/h]}{Carbon \ molar \ flow \ of \ coal \ [kmole/h]} \times 100$$
(7)

Specific emission of CO<sub>2</sub> ( $SE_{CO2}$ ) quantifies the vented CO<sub>2</sub> quantity when 1 MW of power or 1 ton of steel/cement is produced:

$$SE_{CO_2} = \frac{Emitted CO_2 mass flow [kg/h]}{Net power output [MW_e] / Steel or cement output [t]} \times 100$$
(8)

Specific consumption of primary energy for  $CO_2$  avoided (*SPECCA*) takes into accounts both non-carbon capture and carbon capture power plant concepts using the following equation:

$$SPECCA = \frac{SPECCA = \frac{M_{JLHV}}{MWhe} - Heat rate_{No capture} \left[\frac{M_{JLHV}}{MWhe}\right]}{Specific emissions_{No capture} \left[\frac{k_{S}CO_{2}}{MWhe}\right] - Specific emissions_{Capture} \left[\frac{k_{S}CO_{2}}{MWhe}\right]}$$
(9)

Specific capital investment (*SCI*) calculates the capital investment required for production of 1 kW of net power or 1 ton of steel/cement:

$$SCI = \frac{Capital investment [MEuro]}{Net power output [kW_e] / Steel or cement output [t]} \cdot 100$$
(10)

Levelized costs of decarbonized power, steel, and cement were calculated according to the International Energy Agency-Greenhouse Gas R&D Program methodology [29] using the net present value method. This method was translated into in-house developed calculation routines.

 $CO_2$  avoided cost was calculated considering the levelized cost of electricity (or steel/cement) and *specific*  $CO_2$  *emissions* in both plant conditions with and without carbon capture:

$$CO_2 avoided \cos t = \frac{LCOE_{Capture} - LCOE_{No \, capture}}{Specific CO_2 \, emissions_{No \, capture} - Specific CO_2 \, emissions_{Capture}}$$
(11)

The above-mentioned performance indicators, as well as others (e.g., fuel and raw material consumptions, ancillary energy consumptions, etc.) were calculated for the evaluated fossil-intensive industrial applications. Regarding environmental impact indicators, several Life Cycle Assessment (LCA) studies were performed by the authors for gasification [30], combustion [31], and iron and steel [32] but due length constraints only the carbon footprint was presented in details. The LCA results are presented in detail for one illustrative case (i.e., super-critical combustion power plant).

## 5. Results and Discussions

# 5.1. Coal-Based Gasification Power Plants

For decarbonization of gasification-based power plants, two technical options are available. The pre-combustion route when the shifted syngas is decarbonized before combustion in a combined cycle. The post-combustion route when the syngas is used for power production in a combined cycle as in any conventional IGCC design without carbon capture, and then the combustion gases are treated for  $CO_2$  capture (see Figure 3). For gasification systems, the general opinion is that the pre-combustion configurations are more efficient than the post-combustion ones considering the partial pressure of  $CO_2$  in the gas to be decarbonized [16]. This work is considering both pre- and post-combustion capture options to illustrate, in a quantitative manner, the advantages of the pre-combustion capture option. For post-combustion capture, only the calcium looping option was considered. This consideration was based on its higher energy efficiency compared to the chemical scrubbing option. The following gasification-based power plant concepts were evaluated:

Case 1.1—Conventional gasification-based power plant without decarbonization;

Case 1.2—Decarbonized power plant based on the pre-combustion concept using reactive gas-liquid absorption (MDEA);

Case 1.3—Decarbonized power plant based on the pre-combustion concept using reactive gas-solid system (CaL);

Case 1.4—Decarbonized power plant based on the post-combustion concept using reactive gas-liquid absorption (MDEA).

The most important techno-economic and environmental performance indicators of evaluated coal-based gasification power plants are summarized in Table 2.

As shown in Table 2, the pre-combustion capture configurations have a lower decarbonization energy penalty than the post-combustion option (9.3–9.7 vs. 11.7 net energy efficiency percentage points). This energy efficiency difference of about two net percentage points between pre-combustion and post-combustion capture cases can be explained by the significantly higher partial pressure of  $CO_2$  in the gas subject to decarbonization (12–14 bar for pre-combustion cases vs. 0.13–0.16 bar for post-combustion cases). For the pre-combustion options, the MDEA concept (Case 1.2) shows higher net efficiency than the CaL concept (Case 1.3).

The specific consumption of primary energy for  $CO_2$  avoided (SPECCA indicator) shows slightly better performances for the MDEA system compared to the CaL one (either prior or after combustion). In addition, CaL design (which uses a Circulated Fluidization Bed—CFB system) is more complicated to be adjusted for operation at high pressures (about 30–40 bar) as required in the pre-combustion option [33]. Specific  $CO_2$  emissions (carbon footprint) for all decarbonized plants are significantly reduced compared to the benchmark case without carbon capture. A full Life Cycle Analysis (LCA) reveals (as illustrated below in case of super-critical combustion-based power plants) that other environmental impact indicators increase by plant decarbonization [30]. This negative element of process decarbonization is explained by increasing the raw materials consumptions, reducing energy efficiency, and introducing new plant sub-units (CO<sub>2</sub> capture and conditioning units).

Performance Index	UM	Case 1.1	Case 1.2	Case 1.3	Case 1.4
Fossil fuel (coal) consumption	t/h	151.00	166.80	222.00	228.17
Coal lower heating value (LHV)	MJ/kg		25.	17	
Power plant input thermal energy	MW	1055.74	1166.21	1552.15	1595.30
Power output (combined cycle)	MW	560.61	535.88	716.25	720.50
Power consumption	MW	76.25	108.91	156.18	175.01
Net power output	MW	484.36	426.97	560.07	545.49
Net power efficiency	%	45.87	36.61	36.08	34.19
Plant decarbonization rate	%	0.00	90.00	90.00	90.00
Specific power plant emissions	kg/MWh	760.25	85.48	83.02	88.95
SPEĈCA	MJ/kg	-	2.94	3.14	3.99
Specific capital investment	€/kW	1874.00	2620.00	2305.00	3286.00
Levelised cost of electricity	€/MWh	54.13	73.28	76.07	81.25
CO <sub>2</sub> avoided cost	€/t	-	28.38	32.40	40.39

Table 2. Gasification power plants techno-economic and environmental performance indexes.

The decarbonization process of gasification-based power generation brings significant economic penalty (23–75% increase in the specific capital investment, 35–50% increase in the electricity cost). The economic indicators show that pre-combustion capture (either gas-liquid absorption or calcium looping) is definitely better than post-combustion capture in term of specific capital investment (reduced by 20–30%), levelized cost of electricity (reduced by 6–10%) and CO<sub>2</sub> avoided cost (reduced by 19–30%). The MDEA-based decarbonization option has slightly better electricity cost, and CO<sub>2</sub> avoided cost than the calcium looping option (for the technical reasons mentioned above).

One relevant element to be mentioned here in connection with gasification systems represents the ability of this partial oxidation technology to generate, in a flexible manner, various energy carriers. For instance, after syngas decarbonization in a pre-combustion capture configuration, the hydrogen-rich gaseous stream could be employed for the generation of power, hydrogen, or other synthetic carbon-based fuels (methanol, substitute natural gas, synthetic hydrocarbons via Fischer–Tropsch process). One key advantage of these systems represents high cumulative energy efficiency. This specific design characteristic of gasification-based energy conversion systems represents an important element for the future low carbon higher efficiency systems [34].

#### 5.2. Coal-Based Super-Critical Combustion Power Plants

To quantify the techno-economic and environmental impact of the decarbonization process for the coal-based, super-critical, combustion-based, power generation using post-combustion capture systems, the next power plant concepts were used as illustrative examples in this work:

Case 2.1—Conventional combustion-based power plant without decarbonization; Case 2.2—Decarbonized power plant based on reactive gas-liquid absorption (MDEA); Case 2.3—Decarbonized power plant based on reactive gas-solid system (CaL).

The non-capture concept (Case 2.1) is based on current industrial design having 500 MW net output [19]. For the assessed decarbonized concepts (both having the same plant decarbonization degree—90%), the additional heat and power consumptions required for  $CO_2$  capture are covered by the main power block. The most important techno-economic and environmental performance indicators of evaluated coal-based super-critical power plants are summarized in Table 3.

Performance Index	UM	Case 2.1	Case 2.2	Case 2.3
Fossil fuel (coal) consumption	t/h	165.00	208.50	199.13
Coal lower heating value (LHV)	MJ/kg		25.17	
Power plant input thermal energy	MW	1153.62	1457.76	1392.24
Power output (steam turbine)	MW	528.90	569.05	596.81
Power consumption	MW	28.90	69.05	96.81
Net power output	MW	500.00	500.00	500.00
Net power efficiency	%	43.34	34.30	35.92
Plant decarbonization rate	%	0.00	90.00	90.00
Specific power plant emissions	kg/MWh	800.61	89.60	77.05
SPECCA	MJ/kg	-	3.08	2.41
Specific capital investment	€/kW	1320.00	2520.00	1875.00
Levelised cost of electricity	€/MWh	45.53	84.02	68.41
CO <sub>2</sub> avoided cost	€/t	-	49.09	31.34

Table 3. Super-critical power plants techno-economic and environmental performance indexes.

As presented in Table 3, the decarbonization penalty for super-critical power plants lays between seven to nine net energy efficiency percentage points (for the same decarbonization rate considered in both options—90%). The post-combustion calcium looping decarbonization system shows improved values in comparison to the chemical gas-liquid absorption scrubbing system. The difference in net power efficiency points for the two decarbonization systems is about 1.62. This value is justified by the high-temperature heat recovery potential of the calcium looping cycle. The specific consumption of primary energy for CO<sub>2</sub> avoided (SPECCA) also shows better value for chemical looping in comparison to chemical scrubbing (gas-liquid absorption) by about 0.67 MJ/kg. All these technical and environmental benefits of the CaL-based decarbonization process translate into improved economic performance [35]. All economic indicators are in favor of the calcium looping option in comparison to the gas-liquid absorption as assessed decarbonization technologies—specific investment cost (reduced by about 25%), levelized cost of electricity (reduced by about 18%), and  $CO_2$  avoided cost (reduced by about 36%). Also, it worth mentioning that the combustion-based power generation is cheaper than the gasification-based power generation in a non-carbon capture scenario, but when carbon capture is implemented, the economic differences are reduced significantly or even are in favor of IGCC power plants (see Tables 2 and 3).

To illustrate an in-depth environmental impact evaluation for the assessed super-critical power plants with and without carbon capture, Table 4 presents the Life Cycle Analysis (LCA) results in a cradle-to-grave approach using CML 2001 method. The full technical details of this LCA analysis are presented in the paper indicated as reference [31].

Environmental Impact Index	UM	Case 2.1	Case 2.2	Case 2.3
Global warming potential	kg CO <sub>2</sub> eq./MWh	970.37	495.93	402.20
Acidification potential	kg SO <sub>2</sub> eq./MWh	0.49	4.57	1.66
Eutrophication potential	kg PO4 <sup>-3</sup> eq./MWh	1285.44	1739.76	1121.86
Ozone depletion potential $\times 10^8$	kg R11 eq./MWh	0.59	4.07	2.63
Abiotic depletion potential	MJ/MWh	9829.28	15,231.63	13,752.06
Freshwater ecotoxicity potential	kg DCB eq./MWh	0.27	1.66	1.10
Human toxicity potential	kg DCB eq./MWh	3.41	55.27	19.84
Photochemical oxidation potential	kg Ethene eq./MWh	0.20	2.71	0.26
Terrestrial ecotoxicity potential	kg DCB eq./MWh	0.05	0.28	0.18
Marine ecotoxicity potential	kg DCB eq./MWh	6730.54	26,011.85	16,494.81

Table 4. Environmental impact indicators (Life Cycle Analysis) for super-critical power plants.

It can be noticed that the carbon footprint (noted here as Global Warming Potential—GWP) is reduced with plant decarbonization (not corresponding to 90% plant decarbonization rate but to a lower rate due to up-stream and down-stream processes). All other environmental indicators are increasing with plant decarbonization; in some cases, the increasing rate is quite high (e.g., acidification potential, indicators related to toxicity, etc.). It worth mention that for the calcium looping system, all environmental indicators have better values than for the reactive gas-liquid absorption system. The main reason for this result represents the lower environmental impact of natural-based calcium sorbent in comparison to a chemical-based solvent.

## 5.3. Integrated Steel Mills

To assess the techno-economic and environmental impact of decarbonization applied to iron and steel production, the next plant concepts were used as illustrative examples:

Case 3.1—Conventional steel mill without decarbonization;

Case 3.2—Decarbonized steel mill based on reactive gas-liquid absorption (MDEA);

Case 3.3—Decarbonized steel mill based on reactive gas-solid system (CaL).

As presented in Table 1, a conventional integrated iron and steel plant was considered in the assessments with 4 Mt/y hot-rolled coil (HRC) production capacity [3]. All assessed decarbonized steel concepts are not considering any import of heat and power (steel mill off-gases are used for this purpose). In this respect, natural gas was used as additional fuel to cover the ancillary energy consumptions [36]. The decarbonized steel mill concepts capture  $CO_2$  from the most significant plant units, e.g., captive power plant, blast furnace hot stoves, and lime coke production units. The most important performances of evaluated steel plants are exhibited in Table 5.

Table 5. Integrated steel mills techno-economic and environmental performance indexes.

Performance Index	UM	Case 3.1	Case 3.2	Case 3.3
Fossil fuel (natural gas) consumption	MW	669.80	544.00	1156.80
Power output (gas turbine)	MW	-	202.31	91.06
Power output (steam turbine)	MW	224.68	107.33	366.06
Gross power block output	MW	224.68	309.65	457.12
Power consumption	MW	9.68	1.68	132.65
Net power block output	MW	215.00	307.97	324.47
Net power block efficiency	%	32.10	56.61	28.04
Decarbonization rate (power block)	%	0.00	0.00	90.00
Specific CO <sub>2</sub> emissions (power)	kg/MWh	2455.42	370.02	242.32
Specific CO <sub>2</sub> emissions (steel)	kg/t HRC	980.48	229.50	166.10
Decarbonization rate (capture plant)	%	0.00	90.00	90.00
Overall plant specific CO <sub>2</sub> emissions	kg/t HRC	2092.50	833.55	640.00
Quantity of captured CO <sub>2</sub>	kg/t HRC	0.00	1615.80	1495.20
Specific capital investment	€/t HRC	955.00	1077.00	1015.00
Levelised cost of steel	€/t HRC	520.73	614.05	580.70
CO <sub>2</sub> avoided cost	€/t	-	73.46	68.92

As shown in Table 5, the decarbonization of main CO<sub>2</sub> emitters (captive heat and power plant, blast furnace, lime and coke production units) from an integrated iron and steel production plant cut the overall carbon footprint significantly (with a reduction of about 60 to 70%). Considering the significant amount of greenhouse gas emission of this important industrial sector, this reduction is very substantial [36]. The investigated looping cycle has some important advantages in comparison to the chemical scrubbing system, e.g., higher energy conversion yield and subsequent lower decarbonization energy and cost penalties, integration of spent calcium-based sorbent in the iron and steel production process with a positive impact on plant environmental performance [37]. As for the above-presented cases (combustion and gasification-based power generation), the decarbonization process reduces carbon footprint but increases other environmental indicators [36].

Decarbonization of iron and steel production process brings an economic penalty (6–13% increase of specific capital investment, 12–18% increase of steel production cost). The economic indicators

show that the calcium looping option has slightly better economic performance than post-combustion capture in terms of specific capital investment (reduced by about 6%), steel production cost (reduced by about 5%), and  $CO_2$  avoided cost (reduced by about 6%).

## 5.4. Cement Plants

To evaluate the impact of process decarbonization (in a post-combustion capture configuration) for cement production, the next plant concepts were considered as illustrative examples:

Case 4.1—Conventional cement production plant without decarbonization;

Case 4.2—Decarbonized cement production based on reactive gas-liquid absorption (MDEA);

Case 4.3—Decarbonized cement production based on reactive gas-solid system (CaL).

As presented in Table 1, a conventional cement plant was considered in the assessments with 1 Mt/y production capacity [4]. For decarbonized cement production cases by reactive gas-liquid and gas-solid systems, a coal-based combustion unit in conjunction with a steam cycle power block was used to cover the ancillary energy (heat and power) consumptions of the carbon capture plants [38]. The excess energy was exported as the power to the grid with a 520 kg/MWh as the carbon dioxide emission factor. The most relevant techno-economic and environmental performance indexes of the assessed cement plants are exhibited in Table 6.

Performance Index	UM	Case 4.1	Case 4.2	Case 4.3
Fossil fuel (coal) consumption	t/h	-	33.50	22.10
Coal inferior calorific value	MJ/kg		25.17	
Thermal energy (decarbonization unit)	MW	-	234.22	154.51
Power output (steam turbine)	MW	-	54.40	58.12
Gross power block output	MW	-	54.40	58.12
Power consumption	MW	16.24	34.16	42.38
Net power block output	MW	-	20.24	15.74
Net power block efficiency	%	-	8.64	10.18
Plant decarbonization rate	%	0.00	90.00	90.00
Specific plant CO <sub>2</sub> emissions (on-site)	kg/t cement	728.42	135.78	120.74
Specific plant CO <sub>2</sub> emissions (export)	kg/t cement	42.02	-79.93	-62.35
Specific plant CO <sub>2</sub> emissions (total)	kg/t cement	770.44	55.85	58.39
Quantity of captured CO <sub>2</sub>	kg/t cement	0.00	1214.17	962.20
Specific capital investment	€/t cement	263.00	557.00	458.00
Levelized cost of cement	€/t cement	65.60	127.68	106.73
CO <sub>2</sub> avoided cost	€/t	-	86.87	57.76

Table 6. Cement plants techno-economic and environmental performance indexes.

As presented in Table 6, the cement plant decarbonization cut significantly the overall carbon footprint of the process (by about 92–93%). The investigated looping cycle exhibits several benefits in comparison to the chemical scrubbing system, e.g., higher energy conversion yield and subsequent lower decarbonization energy penalty. Another important element of the calcium-based reactive gas-solid system refers to the potential to reuse the spent solid material in the cement plant (clinker production). This element brings advantages in terms of reducing carbon footprint as well as improving technical and economic performance indicators [39]. As for the above-presented cases, the decarbonization process brings a positive effect in reducing the carbon footprint, but on the other hand, increases other environmental indicators [40].

Decarbonization of the cement production process brings significant economic penalty (74–112% increase of specific capital investment, 62–95% increase of cement production cost). The economic indicators show that calcium looping option has better economic performance than post-combustion capture in term of specific capital investment (reduced by about 18%), cement production cost (reduced by about 16%), and CO<sub>2</sub> avoided cost (reduced by about 34%).

## 6. Conclusions

This techno-economic and environmental analysis assessed in a quantifiable manner the decarbonization process of some important fossil-based industrial processes (electricity generation, iron, steel and cement production). As decarbonization technologies, two reactive gas-liquid (chemical scrubbing using alkanolamines-MDEA) and gas-solid (based on calcium-based sorbents) systems were assessed. The CaL decarbonization option exhibits improved performance indicators over the chemical scrubbing for the evaluated post-combustion capture configurations (higher energy conversion yields, lower carbon footprint and specific primary energy consumption, better economic indicators, etc.). In addition, for some of the evaluated processes (steel and cement production plants), the spent solid sorbent from the looping cycle can be utilized by the integration of the whole production chain with positive techno-economic and environmental results. For the pre-combustion capture configuration (evaluated here in relation to a coal gasification plant), the chemical scrubbing by gas-liquid absorption shows higher energy efficiency and more potential for future developments than CaL decarbonization option. Regarding the environmental impact of the LCA analysis, it is worth mentioning that the carbon footprint is reduced by the process decarbonization, but other environmental indicators show significant increases. To illustrate this element, the results of an LCA analysis were presented in detail for super-critical combustion-based power plant concepts.

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