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Methodology to Calculate the CO₂ Emission Reduction at the Coal-Fired Power Plant: CO₂ Capture and Utilization Applying Technology of Mineral Carbonation

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Abstract: This study introduces a novel methodology to calculate the carbon dioxide (CO₂) emission reduction related to residual emissions, calculating the CO₂ emission reduction through a 2 MW (40 tCO₂/day) carbon capture and utilization (CCU) plant installed at a 500 MW coal-fired power plant in operation, to evaluate the accuracy, maintainability, and reliability of the quantified reduction. By applying the developed methodology to calculate the CO₂ emission reduction, the established amount of CO₂ reduction in the mineral carbonation was evaluated through recorded measurement and monitoring data of the 2 MW CCU plant at the operating coal-fired plant. To validate the reduction, the accuracy, reproducibility, consistency, and maintainability of the reduction should be secured, and based on these qualifications, it is necessary to evaluate the contribution rate of nationally determined contributions (NDCs) in each country. This fundamental study establishes the concept of CCU CO₂ reduction and quantifies the reduction to obtain the validation of each country for the reduction. The established concept of the CCU in this study can also be applied to other CCU systems to calculate the reduction, thereby providing an opportunity for CCU technology to contribute to the NDCs in each country and invigorate the technology.

Keywords: emission reduction calculation; carbon capture and utilization; mineral carbonation; carbon dioxide; power plant

1. Introduction

Under the Paris Agreement agreed upon in 2015, all countries must submit their nationally determined contributions (NDC), a voluntary reduction goal, to the United Nations (UN), and undergo a performance evaluation every five years for the submitted NDC [1]. Thus, to reduce their GHG emissions, countries are developing greenhouse gas (GHG) reduction technologies in various fields. South Korea is developing diverse policies and technologies to achieve its set goal, which is to reduce GHG by 37%, compared to business as usual (BAU), by 2030 [2].

The development of GHG reduction technologies can be categorized into technology to reduce CO₂ emission, the main emission source, and technology to reduce non-CO₂ GHG emissions. Among the technologies, active research efforts have been made on carbon capture and utilization or storage (CCUS), as it is one of the technologies to reduce CO₂ emissions. Korea has arranged a CCUS study to reduce CO₂ by 10.3 million tons by 2030 for CCUS technology commercialization [2].

Carbon capture and storage (CCS) is considered the key technology for mitigating emissions from fossil-fuel power plants while these are still operational [3]. Much research has been carried out on how to store the carbon using various methods, such as geological sequestration [4,5], biological fixation [6,7], and ocean disposal [8,9]. However, CCS technologies has some barriers, such as the fact that it is an unprofitable activity that requires large capital investment [10], and in some countries CCS is not optional as their geological capacity is limited or, in some cases, only available offshore, which increases transportation and injection costs [11]. Owing to these downsides of CCS, carbon capture and utilization (CCU) has gained better evaluation for its cost-effectiveness and practical measuring and monitoring of GHG emission reduction compared to CCS [12,13].

CO₂ is utilized in a variety of ways: chemicals or fuels through electro-catalytic conversion [14], plastics based on CO₂-based polymers [15], urea [16], mineral carbonation, etc. The mineral carbonation technology, CCU technology, converts CO₂ to solid inorganic carbonate through a chemical reaction, and uses it in construction materials and coal mine landfills, or safely disposes of the CO₂ without potential CO₂ leakage, which may be threatening to human and environmental safety [17,18].

Although CCU has different GHG emission reduction calculation methods for each of its various technologies, even the UNFCCC, which provides the most effective methodology for GHG emission reduction, is not able to provide an adequate methodology [19].

In this study, we establish a novel methodology to calculate the CO₂ emission reduction related to residual emissions, where convention methods, e.g., UNFCCC CDM or ISO 14064-2, does not consider the residuals. The novel methodology has evaluated accuracy, maintainability, and reliability of quantified CO₂ emission reduction based on the CO₂ emission reduction in 2 MW class (40 tCO₂/day) installed at a currently operating 500 MW coal-fired power plant. This novel method could be the opportunity to quantify the CO₂ reduction in CCU and increase the reliability of CCU technology to gain more attention, and ultimately contribute to sustainable development worldwide.

2. Mineral Carbonation CCU Technology

2.1. Overview of Object Technology

By reacting CO₂ with the reagent CaO, the CO₂ concentration in power generation emission decreases from 10–15% to under 1% and allows the resulting permanently captured CO₂ to be utilized as construction materials in building and civil construction. This study is based on the operating data of a 40 tCO₂/day class (12,000 tCO₂/y, 2 MW) CCU pilot plant installed at a coal-fired power plant in Korea.

Figure 1 shows that the CCU process of mineral carbonation technology consists of a two-tiered reaction tower. In the reaction tower, the reagent, CaO, reacts with water, resulting in an aqueous solution of Ca(OH)₂. Then, CO₂ from the flue gas is injected by the blower to the Ca(OH)₂ solution, resulting in a carbonation reaction.

Figure 1. Principle of direct CO₂ capture and mineral carbonation technologies.

The characteristics of this process involve three phases (gas (CO₂), liquid (water), and solid (CaO)) in the reaction, and the slow melting speed of solid reactant. The reaction equations are as follows [20,21]

After carbonation, the CaCO₃ slurry produced is dehydrated and dried, to be utilized in the final powder form.

2.2. Reaction Mechanism by Mineral Carbonation

To confirm the introduced reaction mechanism from Section 2.1, chemical analysis was conducted on the materials before and after the reaction with CO₂, CaO, and H₂O in the 40 tCO₂/day class (2 MW) CCU pilot plant.

2.2.1. Results of the Chemical Analysis of Pre-Reaction Liquid Reagent

For chemical component analysis of the pre-reaction liquid agents, produced before the reaction of CaO, H₂O, and CO₂ in the CCU facility, a glass disc was made by fusing the agent with sodium tetraborate (Na₂B₄O₇) or lithium tetraborate (Li₂B₄O₇), after the loss of ignition (Ig.loss) was measured. Moreover, after measuring the loss of ignition of the concentration-known standard material, glass disc was made with the same method. The calibration curve of the fluorescence X-ray intensity was drawn according to the content changes in the corresponding components (silicon dioxide, aluminum oxide, calcium oxide, etc.), and the fluorescence X-ray intensity of these components was measured and quantified using wavelength dispersive X-ray fluorescence spectrometry (WD-XRF) and inductively coupled plasma-optical emission spectrometry (ICP-OES).

The pre-reaction liquid agent composition was based on three reagents, mainly CaO of 63–68%, and partially SiO₂, Al₂O₃, Fe₂O₃, MgO, etc. Table 1 shows that the loss of ignition was confirmed to be 27–28%.

Table 1. Chemical analysis results of pre-reaction liquid reagent. Samples were four different glass discs made by fusing the agent with Na₂B₄O₇ or Li₂B₄O₇ for four different times.

Analysis Results (Mass %)											
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	Ig.Loss	Total
Sample 1	0.65	0.62	0.24	–	68.1	1.83	–	–	0.10	28.46	100.00
Sample 2	0.96	0.42	0.27	–	67.74	1.81	0.21	0.00	0.11	28.48	100.00
Sample 3	1.13	0.46	0.33	–	66.54	2.22	1.45	0.11	0.41	27.35	100.00
Sample 4	1.01	0.46	0.31	–	63.88	2.15	3.42	0.09	0.29	28.39	100.00

2.2.2. Results of the Chemical Analysis of the Post-Reaction Product

The produced product from the reaction of liquid reagent and CO₂ in CCU facility was chemically analyzed as per the Section 2.2.1 analysis. After the reaction with CO₂, the product starts to react. Ten samples were obtained from sampling every 2 h during the reaction of the product. The analysis results of the product showed that CaO of 51–53% was the principal component; SiO₂, Al₂O₃, Fe₂O₃, MgO, etc. were confirmed to be partially included and the Ig.loss was determined to be 41–43% (Table 2).

Table 2. Chemical analysis results of the post-reaction product.

	Analysis Results (Mass %)						Total
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Ig.loss	
1st	2.29	0.81	0.4	51.4	2.03	42.7	99.63
2nd	2.08	0.58	0.29	52.1	1.86	42.7	99.61
3rd	2.14	0.59	0.31	52.7	1.88	42.1	99.72
4th	2.27	0.91	0.34	52	1.9	42.2	99.62
5th	2.46	0.89	0.36	51.9	1.86	42.2	99.67
6th	2.6	0.91	0.37	51.7	1.95	42.1	99.63
7th	2.36	0.87	0.33	52	1.99	42	99.55
8th	1.79	0.73	0.28	53	1.96	42	99.76
9th	2.2	1.07	0.33	52.5	2.01	41.6	99.71
10th	1.55	0.59	0.28	53.4	2.51	41.4	99.73

To verify the reaction mechanism according to the reaction progress of the final product, the X-ray diffraction (XRD) chart was used to check the mineral phases' changes. Figure 2 shows the analysis results, which confirm that the pre-reaction liquid reagent was mainly lime (Ca(OH)_2) (Figure 2a); but through the reaction with CO_2 , lime was converted to calcite (CaCO_3) and aragonite (CaCO_3) (Figure 2b,c).

Figure 2. XRD results according to the reaction progress. (a) pre-reaction liquid reagent Sample 1. (b) post-reaction product 2nd sampled data and (c) 8th sampled data.

A thermogravimetric analyzer (TGA) was also employed to analyze the reaction mechanism according to the progression, which enables the detection of mass changes as the temperature changes, as it increases along with the constant heating rate. As the result shown in Table 3, the first mass loss of water evaporation occurred at 40–600 °C and the desorption of CO_2 was confirmed at 600–900 °C resulting in a mass loss of 37–39%, which confirmed the chemical conversion of CO_2 to the final product of CaCO_3 [22].

Table 3. Thermogravimetric analyzer (TGA) analysis according to the reaction time. Unit for the results are mass %.

	Step 1 (40–600 °C)	Step 2 (600–900 °C)	TGA (Total)
1st	4.02	38.83	42.85
2nd	3.83	39.03	42.86
3rd	3.90	38.78	42.68
4th	3.80	38.87	42.67
5th	3.86	38.78	42.64
6th	4.04	38.52	42.56
7th	4.38	37.66	42.04
8th	4.87	37.53	42.40
9th	4.74	37.43	42.17
10th	6.22	35.73	41.95

The TGA results show that as the reaction progresses (Figure 3a–c), the CO₂ contents increase, maintaining constant mass percent for the 2nd–6th steps (Figure 3b), and as the CaO concentration decreases, the CO₂ content gradually decreases for the 7th–10th steps (Figure 3c).

Figure 3. TGA results according to sampling time. (a) TGA analysis results of pre-reaction liquids agent. (b) TGA analysis at 2nd step of product reaction (blue line is weight, red line is temperature) (c) TGA analysis at 8th step of product reaction. (Blue line is weight, red line is temperature).

2.2.3. Analysis Results of the Reaction Mechanism

Chemical analysis was performed according to the reaction progress of the three input reagents, CaO, H₂O, and CO₂, in the CCU facility. The XRD and TGA analysis results showed that through the reaction, the input reagents were converted into CaCO₃, which confirmed that this technique is able to permanently capture CO₂ by converting CO₂ into CaCO₃ which has a lower energy level (Figure 4) [23].

Figure 4. Energy level of carbon.

3. Establishment of a Method to Calculate the CO₂ Emission Reduction

3.1. Concept of CO₂ Emission Reduction

The UNFCCC's Clean Development Mechanism (CDM) and ISO 14064-2 concepts were introduced to evaluate the CO₂ reductions. In CDM, the reduced amount of net CO₂ is calculated based on continuous measurement and sampling, by considering the amount of CO₂ emission and CO₂ residual emissions before and after the reduction facility was introduced. Therefore, in this study, the CO₂ emissions from a coal-fired power plant, which is the baseline scenario before the installation of a reduction facility, are treated with desulfurization and a dust collection facility. The baseline CO₂ emission was calculated in terms of emission into the atmosphere without CO₂ capture. After the facility installation, CO₂ emission was continuously measured to calculate the project CO₂ emissions, while CO₂ residual emissions were calculated by analyzing the captured CO₂ amount in the product, to calculate the net CO₂ reductions.

3.2. Method to Calculate the CO₂ Emission Reduction

3.2.1. Method to Calculate the Baseline CO₂ Emission

The baseline CO₂ emissions calculation method excludes the calculated CO₂ in outlet gas from the calculated CO₂ in inlet gas. The amount of inlet CO₂ gas was calculated by multiplying the inflow rate of flue gas, infused into the CO₂ capture and reaction facility, and the CO₂ concentration. The CO₂ in outlet gas was calculated by multiplying the CO₂ concentration value and flow rate of the flue gas emitted from the facility. Flow rates and concentrations were measured continuously during the monitoring period (Equation (4) and Table 4).

$$BE_M = \sum_{m=1}^M (Q_{in,m,gas} \times C_{in,m,CO_2} \times \gamma - Q_{out,m,gas} \times C_{out,m,CO_2} \times \gamma) \quad (4)$$

Table 4. Explanation of symbols used in the Equation (4).

Symbol	Definition	Unit
BE_M	Baseline emissions during the monitoring period	tCO ₂ -eq/MP
$Q_{in,m,gas}$	Accumulated measured value of flue gas flow rate at the inlet of the CO ₂ capture and reaction facility during time interval m	Nm ³ /MP
C_{in,m,CO_2}	Mean measured value of concentration of CO ₂ in the gas at the inlet of the CO ₂ capture and reaction facility during time interval m	v/v%
$Q_{out,m,gas}$	Accumulated measured value of flue gas flow rate at the outlet of the CO ₂ capture and reaction facility during time interval m	Nm ³ /MP
C_{out,m,CO_2}	Mean measured value of concentration of CO ₂ in the flue gas at the outlet of the CO ₂ capture and reaction facility during time interval m	v/v%
γ	The conversion factor of Nm ³ and kg of CO ₂ at 0 °C, 1 atm (44 kgCO ₂ /22.4 Nm ³ = 1.964 kgCO ₂ /Nm ³)	tCO ₂ /Nm ³
M	Total number of time interval m in monitoring period (MP)	
m	time interval	

3.2.2. Method to Calculate the Project CO₂ Emission

Project CO₂ emission is the emission of greenhouse gases caused by the additional energy use, energy penalty, in project activity. Furthermore, when power generation is 100% renewable, the CO₂eq emissions of electricity use is zero. Therefore, this value should reflect the proper emission factor according to the contribution of renewables to electricity production during the monitoring and project period, e.g., national electric power emission factor of Korea.

In this study, the calculated greenhouse gas emission from additional electricity consumption was applied to calculate the project CO₂ emission (Equation (5) and Table 5).

$$PE_M = EC_{EL,M} \times EF_{grid} \quad (5)$$

Table 5. Explanation of symbols used in the Equation (5).

Symbol	Definition	Unit
PE_M	Amount of project emission during the monitoring period	tCO ₂ -eq/MP
$EC_{EL,M}$	Usage amount of electricity during the monitoring period	MWh/MP
EF_{grid}	Electricity emission factor	tCO ₂ -eq/MWh

3.2.3. Method to Calculate the Residual Emissions

The residual emission amount is the amount of residual emissions of CO₂ by the production of the product, which implies that the captured CO₂ from the flue gas is not captured by the product, but dissolved, and remains in the business boundary by water or other substance. The residual emissions factor, RE_{ef} , was developed to calculate the residual emission amount during the sampling period. The RE_{ef} was developed by applying the difference between analyzed amount during sampling period of the utilized CO₂ at the CCU facility and the fixed CO₂ at the actual product, by the CO₂ content analysis of samples (Equation (6) and Table 6).

$$RE_{ef} = 1 - \frac{P_m \times (1 - M_c) \times C_{sample,CO_2}}{\sum_{m=1}^S (Q_{in,m,gas} \times C_{in,m,CO_2} \times \gamma - Q_{out,m,gas} \times C_{out,m,CO_2} \times \gamma)} \quad (6)$$

Table 6. Explanation of symbols used in the Equation (6).

Symbol	Definition	Unit
RE _{ef}	Residual emissions factor during the sampling period	–
Q _{in,m,gas}	Accumulated measured value of flue gas flow rate at the inlet of the CO ₂ capture and reaction facility time interval <i>m</i>	Nm ³ /SP
C _{in,m,CO2}	Mean measured value of concentration of CO ₂ in the gas at the inlet of the CO ₂ capture and reaction facility during time interval <i>m</i>	v/v%
Q _{out,m,gas}	Accumulated measured value of flue gas flow rate at the outlet of the CO ₂ capture and reaction facility during time interval <i>m</i>	Nm ³ /SP
C _{out,m,CO2}	Mean measured value of concentration of CO ₂ in the flue gas at the outlet of the CO ₂ capture and reaction facility during time interval <i>m</i>	v/v%
P _m	Produced amount of product during the sampling period	ton/SP
M _c	Water content of the final product	mass%
C _{sample,CO2}	The CO ₂ concentration of the dried product	mass% (content)
Y	The conversion factor of m ³ and kg of CO ₂ at 0 °C, 1 atm (44 kg CO ₂ /22.4 m ³ = 1.964 kg CO ₂ /Nm ³)	tCO ₂ /Nm ³
S	Total number of time intervals <i>m</i> in sampling period (SP)	
m	Time interval	

The residual emissions amount (RE_M) was calculated by applying the developed RE_{ef} (Equation (7) and Table 7).

$$RE_M = BE_M \times RE_{ef} \quad (7)$$

Table 7. Explanation of symbols used in the Equation (7).

Symbol	Definition	Unit
RE _M	Residual emissions during the monitoring period	tCO ₂ -eq/MP
BE _M	Baseline emissions during the monitoring period	tCO ₂ -eq/MP
RE _{ef}	Residual emissions factor during the sampling period	–

3.2.4. Method to Calculate the CO₂ Emission Reduction

The CO₂ reduction amount was calculated by excluding the sum of project CO₂ emissions and residual amount from the baseline CO₂ emissions amount (Equation (8) and Table 8).

$$ER_M = BE_M - PE_M - RE_M \quad (8)$$

Table 8. Explanation of symbols used in the Equation (8).

Symbol	Definition	Unit
ER _M	Emissions reductions amount of CO ₂ during the monitoring period.	tCO ₂ -eq/MP
BE _M	Baseline emissions during the monitoring period	tCO ₂ -eq/MP
PE _M	Amount of project emission during the monitoring period	tCO ₂ -eq/MP
RE _M	Residual emissions during the monitoring period	tCO ₂ -eq/MP

4. Results and Discussion

To evaluate the CO₂ reduction in applying technology, the monitoring measurement points were defined (yellow points on Figure 5), and the normal operating data collected during 22 h of monitoring period.

Figure 5. Monitoring measurement point of direct CO₂ capture and mass-utilization storage technology.

4.1. The Baseline CO₂ Emission

The measured CO₂ from the measuring instrument was used to determine the baseline CO₂ emissions, the results of which are shown in Table 9.

Table 9. Results of baseline CO₂ emission.

Measured Time	Inflow CO ₂ (ton/h)	Outflow CO ₂ (ton/h)	Baseline CO ₂ Emission (tCO ₂ /h)	Capturing Efficiency (%)
05/29/14.00	1.49	0.15	1.35	89.93
05/29/15.00	1.62	0.15	1.47	90.74
05/29/16.00	1.54	0.15	1.39	90.25
05/29/17.00	1.56	0.15	1.41	90.38
05/29/18.00	1.62	0.14	1.48	91.35
05/29/19.00	1.53	0.17	1.36	88.88
05/29/20.00	1.46	0.12	1.34	91.78
05/29/21.00	1.51	0.15	1.36	90.06
05/29/22.00	1.46	0.13	1.33	91.09
05/29/23.00	1.24	0.16	1.08	87.09
05/30/00.00	1.20	0.15	1.04	87.50
05/30/01.00	1.20	0.18	1.02	85.00
05/30/02.00	1.15	0.17	0.98	85.21
05/30/03.00	1.11	0.18	0.93	83.78
05/30/04.00	1.03	0.16	0.87	84.46
05/30/05.00	0.97	0.15	0.83	84.53
05/30/06.00	1.03	0.20	0.83	80.58
05/30/07.00	1.13	0.26	0.87	76.99
05/30/09.00	0.92	0.20	0.73	78.26
05/30/10.00	0.90	0.21	0.69	76.66
05/30/11.00	0.94	0.23	0.72	75.53
05/30/12.00	0.93	0.23	0.70	75.26
Total	27.5	3.8	23.78	85.71

4.2. The Project CO₂ Emission

The electricity was the energy source during the monitoring period; therefore, the additional electric power consumption was measured through the watt-hour meter to determine the project CO₂

emissions. The electric power emission factor of Korea, which also considers the renewable energy, was applied to the measured electricity consumption [24]. The measured electricity consumption is 8.91 MWh, and the CO₂ emission is 4.155 tCO₂ during the monitoring period, 22 h.

4.3. Residual Emissions

The residual emissions factor (RE_{ef}) during the sampling period has to be calculated to estimate the residual emissions amount (RE_M). The TGA results and the sampling data of product mass were utilized to calculate the residual emissions factor (RE_{ef}). The CO₂ content of the sampled product are shown in column Step 2 of Table 3 in Table 10, and the product mass from the sampling data were gained from a measuring instrument (Product Mass in Table 10). Based on the Equation (6), using the analyzed data, the residual factor (RE_{ef}) during the sampling period was 0.1297, hence the residual emission amount is 3.085 tCO₂ during 22 h of the monitoring period (Equation (7)).

Table 10. Data for residual emission calculation.

Measured Time	Step 2 of Table 3 (%)	Product Mass (ton/h)	Amount of CO ₂ Included in Final Product (tCO ₂ /h)
05/29/14.00–15.00	38.83	3.06	1.19
05/29/16.00–17.00	39.03	3.16	1.23
05/29/18.00–18.00	38.78	3.36	1.30
05/29/20.00–21.00	38.87	3.04	1.18
05/29/22.00–23.00	38.78	3.02	1.17
05/30/00.00–01.00	38.52	2.37	0.91
05/30/02.00–03.00	37.66	2.23	0.84
05/30/04.00–05.00	37.53	1.99	0.75
05/30/06.00–07.00	37.43	1.89	0.71
05/30/11.00–12.00	35.73	1.63	0.58

4.4. CO₂ Emission Reduction

Following the emission results from Sections 4.1–4.3, the CO₂ reduction was confirmed to be 16.54 tCO₂ during 22 h of the monitoring period. Assuming that this facility operates for 24 h/day for 300 days, it was analyzed to reduce by 5413 tCO₂ annually.

4.5. Conservative Approach Method to CO₂ Emission Reduction

The technology in this paper utilizes the produced CaCO₃ through reaction with CaO and CO₂ with coal-fired power plant flue gas as construction materials. To calculate the actual amount of captured CO₂, the CO₂ was quantified in the reactor by the measuring instrument and the unreacted CO₂ residual emissions were quantified by analyzing the CO₂ amount in the final product. Our calculation method applied the amount of CO₂ reduction to the calculation method, which is a continuous measurement of CCU applied inlet/outlet flowrate of CO₂. Table 11 shows that our calculation method calculated 30% less reduction than the conventional CCU reduction, where residual emissions are not considered, thereby avoiding the risk of overestimating the emission reduction amount.

Table 11. Comparison of annual CO₂ emission reduction.

	Our Study	Conventional Method
CO ₂ Emission Reduction (tCO ₂ /yr)	5413	7782

As is mentioned in Section 3.2.3, the residual emission factor in this study signifies the ratio of consumed unreacted CO₂, which was determined by the difference between the amount of the actually captured CO₂ and the amount of CO₂ in the final product. Therefore, the developed residual emissions factor in this paper provides a more accurate calculation method that avoids overestimation for mineral carbonation CCU technology in various intensive greenhouse gas-emitting plants.

4.6. Maximization of the Final Product Application

The final product in this study, CaCO_3 , is expected to be used as a construction material by mixing with cement to strengthen the ground, increase the strength of structures, and produce precast pavers. Accordingly, in the case of Korea, when this technology is commercially distributed, it is suggested that it is possible to achieve a total CO_2 reduction of 6.1 million tons by using 3.8 million tons of civil engineering materials, and by using 2.3 million tons of construction material (Table 12).

Table 12. Application fields in Korea for the final product technology.

Application Fields		Final Product (ton/yr)	Estimated CO_2 Emission Reduction (tCO_2/y)
Civil engineering materials	Top layer mixing treatment	2,100,000	800,436
	Deep mixing treatment	3,500,000	1,334,060
	Soil cement underground continuous method	1,000,000	381,160
	Fluidized bed filling and mixing	1,000,000	381,160
	Other civil materials	2,500,000	952,900
	Subtotal	10,100,000	3,849,716
Construction materials	Cement raw materials	1,700,000	647,972
	Blast-furnace slag raw materials	480,000	182,956
	Ready-mixed (remicon) concrete binder	2,450,000	933,842
	Other construction materials (secondary products, etc.)	1,500,000	571,740
	Subtotal	6,130,000	2,336,511
Total		16,230,000	6,186,227

The Table 12 evaluation basis is as follows: (1) the CO_2 content of the product is 38.12% using Table 10; (2) Korea's soft ground surface development, 20 million m^3/y , solidifying agent 350 kg/m^3 , 30% replacement by CO_2 capture cement; (3) Korea's soft ground surface in-depth development, 6.6 million m^3/y , development depth 30 m, solidifying agent 300 kg/m^3 , 30% replacement by CO_2 captured solidifying agent; (4) Korea's general/special cement 450 million tons/y (the internal data in Korea cement association) 35%, 10% replacement by CO_2 captured cement; (5) Korea's blast furnace slag 96 million tons/y, 5% replacement by CO_2 captured blast-furnace slag; and (6) Korea's remicon 100 million m^3/y , 24 MPa (350 kg/m^3 binder) 70%, approximately 10% replacement by CO_2 captured material binder (35 kg/m^3).

5. Conclusions

This study has established a method to evaluate the CO_2 reduction by mineral carbonation CCU facility, and an evaluation was made on the CO_2 reductions. The evaluation result was achieved by adopting the monitoring data and recorded measurements from a 2 MW class (40 tCO_2/day) continuous capture process installed at a running 500 MW coal-fired power plant. The evaluation results show that the facility can annually reduce 5413 tCO_2 .

Korea has set the goal of reducing 10.3 million tons of CO_2 by 2030 through CCUS technology, and policies, such as the Green New Deal and Korea GHG emission trading system, are being implemented to achieve this goal. However, in order to validate the amount of GHG reduction and meet the national reduction target, a standard that can quantify the GHG reduction is needed. Particularly when converting to another material through a reaction with CO_2 , such as mineral carbonation technology among CCU technologies, residual emission occurs according to the conversion rate, and correction for the reduction amount is required through the measurement. Therefore, in this study, a methodology for calculating the reduction amount is presented by developing and applying a residual emission factor through continuous measurement data acquired from the operating mineral carbonization CCU facility, and analysis data through sampling.

Currently, the proposed CCU facility is a small scale that captures and utilizes 3.43% of CO_2 , which is a small part of a 500 MW thermal power plant. When the CCU facility is applied with the scaled-up technology, further studies can help to improve this study: securing storage space for raw materials and products; studies on monitoring factors such as pH; operational life time analysis of the

facility through track record for continuous operation; a method of calculating the reduction amount from the LCA viewpoint according to the use of the ingredient CaO, etc.

Nevertheless, countries must achieve their reduction targets by 2030, and CCU technology is regarded as an excellent method for achieving the reduction targets. In order to validate the CO₂ emission reduction, the accuracy, reproducibility, consistency and maintainability of the reduction should be secured, and it is necessary to evaluate the contribution rate of NDCs in each country. Therefore, through this research, the concept of CO₂ reduction amount of CCU was established and quantified to conduct the elemental study, to obtain validation of the reduction in each country. We hope that the established concept in this study could be applied to the calculation of other CCU technologies, providing an opportunity for CCU technology to contribute to NDCs and the activation of CCU technology.

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References

1. UNFCCC. Adoption of the Paris Agreement. In *Conference of the Parties, UNFCCC*; United Nations Framework Convention on Climate Change [UNFCCC]: Paris, France, 2015.
2. Ministry of Environment. *Roadmap for Achieving the National Greenhouse Gas Reduction Target (Plan)*; The Korean Government: Sejong, Korea, 2018.
3. Snæbjörnsdóttir, S.Ó.; Sigfússon, B.; Marieni, C.; Goldberg, D.; Gislason, S.R.; Oelkers, E.H. Carbon dioxide storage through mineral carbonation. *Nat. Rev. Earth Environ.* **2020**, *1*, 90–102. [[CrossRef](#)]
4. Hadi Mosleh, M.; Sedighi, M.; Babaei, M.; Turner, M. 16-Geological sequestration of carbon dioxide. In *Managing Global Warming*; Letcher, T.M., Ed.; Academic Press: New York, NY, USA, 2019; pp. 487–500.
5. Nguyen, D.N. Carbon Dioxide Geological Sequestration: Technical and Economic Reviews. In *SPE/EPA/DOE Exploration and Production Environmental Conference*; Society of Petroleum Engineers: San Antonio, TX, USA, 2003; p. 6.
6. Maheshwari, N.; Krishna, P.K.; Thakur, I.S.; Srivastava, S. Biological fixation of carbon dioxide and biodiesel production using microalgae isolated from sewage waste water. *Environ. Sci. Pollut. Res.* **2020**, *27*, 27319–27329. [[CrossRef](#)] [[PubMed](#)]
7. Schwander, T.; Schada von Borzyskowski, L.; Burgener, S.; Cortina, N.S.; Erb, T.J. A synthetic pathway for the fixation of carbon dioxide in vitro. *Science* **2016**, *354*, 900–904. [[CrossRef](#)] [[PubMed](#)]
8. Strand, S.E.; Benford, G. Ocean Sequestration of Crop Residue Carbon: Recycling Fossil Fuel Carbon Back to Deep Sediments. *Environ. Sci. Technol.* **2009**, *43*, 1000–1007. [[CrossRef](#)] [[PubMed](#)]
9. Wilson, T.R.S. The deep ocean disposal of carbon dioxide. *Energy Convers. Manag.* **1992**, *33*, 627–633. [[CrossRef](#)]
10. Peter Styring, D.J.; Heleen de Coninck, H.R.; Armstrong, K. *Carbon Capture and Utilisation in the Green Economy*; CO2 Chem: York, UK, 2011.
11. Khoo, H.H.; Bu, J.; Wong, R.L.; Kuan, S.Y.; Sharratt, P.N. Carbon capture and utilization: Preliminary life cycle CO₂, energy, and cost results of potential mineral carbonation. *Energy Procedia* **2011**, *4*, 2494–2501. [[CrossRef](#)]
12. Cuéllar-Franca, R.M.; Azapagic, A. Carbon capture, storage and utilisation technologies: A critical analysis and comparison of their life cycle environmental impacts. *J. CO2 Util.* **2015**, *9*, 82–102. [[CrossRef](#)]
13. Pieri, T.; Nikitas, A.; Castillo-Castillo, A.; Angelis-Dimakis, A. Holistic Assessment of Carbon Capture and Utilization Value Chains. *Environments* **2018**, *5*, 108. [[CrossRef](#)]

14. Kondratenko, E.V.; Mul, G.; Baltrusaitis, J.; Larrazábal, G.O.; Pérez-Ramírez, J. Status and perspectives of CO₂ conversion into fuels and chemicals by catalytic, photocatalytic and electrocatalytic processes. *Energy Environ. Sci.* **2013**, *6*, 3112–3135. [[CrossRef](#)]
15. Yadav, N.; Seidi, F.; Crespy, D.; D'Elia, V. Polymers Based on Cyclic Carbonates as Trait d'Union between Polymer Chemistry and Sustainable CO₂ Utilization. *ChemSusChem* **2019**, *12*, 724–754. [[CrossRef](#)] [[PubMed](#)]
16. Styring, P.; Driver, J.G.; Owen, R.; Makenyire, T.; McGregor, J.; Lake, J.A. Blue Urea: Fertilizer with reduced environmental impact. *Front. Energy Res.* **2019**, *7*, 88.
17. Brinckerhof, P. *Accerlerating the Uptake of CCS: Industrial Use of Captured Carbon Dioxide*; Global CCS Institute: Docklands, Australia, 2011.
18. Irlam, L. *Global Costs of Carbon Capture and Storage*; Global CCS Institute: Docklands, Australia, 2017; Volume 2017.
19. UNFCCC. *CDM Methodology*, 11th ed.; UNFCCC: Bonn, Germany, 2019.
20. Jin, B.; Lant, P. Flow regime, hydrodynamics, floc size distribution and sludge properties in activated sludge bubble column, air-lift and aerated stirred reactors. *Chem. Eng. Sci.* **2004**, *59*, 2379–2388. [[CrossRef](#)]
21. Pangarkar, V. *Design of Multiphase Reactors*, 1st ed.; John Wiley & Sons: Hoboken, NJ, USA, 2014.
22. Rodriguez, C.; Chun, J.; Schweiger, M.; Hrma, P. Evolved Gas Analysis for High-Alumina High Level Waste Feed. *Adv. Mater. Sci. Environ. Energy Technol. III* **2014**, *250*, 195–203.
23. Romanosky, R. CO₂ Mineral Sequestration. In Proceedings of the Advanced Research Power Program, National Energy Technology Laboratory, Mineral Carbonation Workshop, Pittsburg, PA, USA, 8 August 2001; Volume 2001.
24. GIR. *2018 Approved National Gas Emission, Absorption Coefficient*; Greenhouse Gas Inventroy and Research Center: Seoul, Korea, 2018.

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