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Comparative Life Cycle Assessment (LCA) of Accelerated Carbonation Processes Using Steelmaking Slag for CO_2 Fixation

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ABSTRACT

Carbon capture, utilization, and storage (CCUS) is one of the most prominent emerging technologies for mitigating global climate change. In this study, a comparative evaluation for CO_2 fixation by carbonation of steelmaking slag was performed by life cycle assessment (LCA) using Umberto 5.5.4 software, with the Swiss Eco-invent 2.2 database. Six scenarios of carbonation for basic oxygen furnace slag (BOFS), steel converted slag (SCS), and blended hydraulic slag cement (BHC) in different types of reactors and/or method were established. The environmental impacts for each scenario are quantified using the valuation system of ReCiPe, where global warming potential (GWP), ecosystem quality potential (EQP), and human health potential (HHP) were evaluated. In addition, sensitivity analysis was carried out to evaluate the relevant uncertainties of heating efficiency on the GHG emissions in direct carbonation processes. According to the results of LCA and sensitivity analysis, the direct carbonation of steelmaking slag in a slurry reactor was found to be the most attractive method, since the GWP was the lowest among the selected scenarios. Furthermore, the best available technology (BAT) for CO_2 capture by carbonation processes of alkaline wastes was proposed according to the key performance indicators (KPIs) with respect to engineering considerations and environmental impacts. It was concluded that the accelerated carbonation of steelmaking slag should be performed by combining the slurry reactor with a rotating packed bed (RPB) to maximize carbonation conversion and minimize environmental impacts and additional CO_2 emissions.

Keywords: Technology assessment; Umberto; Environmental impacts; ReCiPe; Sensitivity analysis; Rotating packed bed.

INTRODUCTION

The increase of carbon dioxide (CO_2) concentrations in the atmosphere has spurred worldwide concerns of global climate change among industrial sectors, governmental departments, and academic institutes (Khoo and Tan, 2006; IPCC, 2007; Pan *et al.*, 2012). Thousands of action plans have been executed to pursue practical technologies and scientific solutions to overcome the challenges of global warming (IPCC, 2007; Yang *et al.*, 2008). According to the IEA report, the strategies for reducing CO_2 emissions include improving overall energy efficiency, implementing carbon capture and storage (CCS) technologies, and utilizing renewable energy and material recycling (IEA and UNIDO, 2011). It was noted that CCS could reduce CO_2 emissions by 9–50% in industrial sectors, compared to the present level, by 2050 and could mitigate cumulative global climate change by 15–55% by 2100 (Metz *et al.*, 2005; IEA and UNIDO, 2011). In addition, according to the concluding remarks of the COP 17 meeting held at Duban in 2011, the implementation of CCS technologies was regarded as an eligible clean development mechanism (CDM) project and/or activity. However, the geological storage of CO_2 demonstrated around the world still faces many uncertainties and risks such as accidental leakage of CO_2, environmental impacts, and public acceptance (Duan, 2010; Terwel *et al.*, 2011; Terwel *et al.*, 2012; Zoback and Gorelick, 2012). Therefore, carbon capture, utilization, and storage (CCUS) has recently received global attention as a viable option for reducing CO_2 emissions (Birat, 2010; Chiu and Ku, 2012; Pan *et al.*, 2012; Yu *et al.*, 2012a, b; Lee *et al.*, 2013).

Mineral sequestration is a method of carbon capture that accelerates the natural weathering of silicate minerals, allowing them to react with CO_2 to form stable products, carbonate minerals and silica for further utilization (Lackner, 2003; Uibu *et al.*, 2011; Pan *et al.*, 2012; Santos *et al.*, 2012a, b). Mineral carbonation occurs slowly under natural conditions, but this process might be accelerated if additional energy and resources were introduced. A variety of feedstock

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of industrial alkaline solid wastes including steelmaking slag, cement kiln dust, waste ash, paper mill waste and mining waste are recommended for mineral sequestration (Costa *et al.*, 2007; Li *et al.*, 2007; Pérez-López *et al.*, 2008; Huntzinger *et al.*, 2009; Bobicki *et al.*, 2012; Pan *et al.*, 2013a). Steelmaking industries, one of the largest sectors generating abundant CO_2, accounting for 6–7% worldwide, produce significant quantities of waste slag with high mass fractions of calcium, exhibiting a potential for carbonation. Therefore, CO_2 could be fixed by CaO or MgO in silicate-based materials reacted with carbon dioxide, and then used to form stable carbonate precipitations (Eloneva *et al.*, 2009; Chang *et al.*, 2011a).

Generally, carbonation processes can be categorized into two groups: direct and indirect. Direct carbonation includes gas-solid reactions and mineralization in aqueous solution, which often requires a pre-treatment process for feedstock. On the other hand, indirect carbonation is accomplished by extraction or bioleaching prior to carbonation. The challenge of mineral sequestration is to accelerate the carbonation process; while, at the same time, exploiting the heat of reaction with minimal energy and material losses (Bobicki *et al.*, 2012). Recently, scientists have attempted to turn this process into a practical technology and capture CO_2 with lower energy and chemical usage (Eloneva *et al.*, 2008; Baciocchi *et al.*, 2009; Chang *et al.*, 2011b; Eloneva *et al.*, 2012).

In addition, life cycle assessment (LCA) is a scientific and technical tool to assess the requirements and impacts of technologies, processes and products so as to determine their propensity to consume resources and contribute to pollution (Guinée *et al.*, 2011; Mckone *et al.*, 2011; Michalek *et al.*, 2011). Since CO_2 capture process would consume energy and materials, generating additional CO_2 emission into the atmosphere, the overall process should be critically evaluated by LCA (Viebahn *et al.*, 2007; Khool *et al.*, 2011; Zapp *et al.*, 2012). The development of the international standards for LCA was mainly responsible by the international organization for standardization (ISO). ISO 14000 including ISO 14040:1997, ISO 14041:1999, ISO 14042:2000, and ISO 14043:2000 is a family of standards related to environmental management, which are generally accepted by all stakeholders and the international community (Finkbeiner *et al.*, 2006). According to ISO 14040, the procedures for implementing LCA include (1) definition of goal and scope, (2) life cycle inventory (LCI), (3) life cycle impact assessment (LCIA), and (4) data interpretation. The key issues such as energy penalty, scale-up challenges, non-climate environmental impacts, uncertainty management, policy-making needs, and market effects should be considered in LCA (Pehnt and Henkel, 2009; Sathre *et al.*, 2012; Viebahn *et al.*, 2012).

As a result, the objectives of this investigation were to systematically assess carbonation processes (including autoclave reactor, slurry reactor and rotating packed bed) with respect to technology development and environmental impacts. The environmental impacts, including global warming potential (GWP), ecosystem quality potential (EQP), and human health potential (HHP), were quantified by LCA. In addition, the uncertainty of the heating efficiency was determined by a sensitivity analysis. Furthermore, the best available technology (BAT) for carbonation of steelmaking slag was proposed based on the results of technology assessment and LCA.

METHODS

Criteria for Technology Assessment

In general, CO_2 fixed by mineral carbonation can be divided into two methods, i.e., direct and indirect carbonation. Table 1 summarizes the representative techniques for accelerated carbonation of steelmaking slags from the literature. The content of CaO and SiO_2 for the above steelmaking slags ranged from 42–52% and 11–28%, respectively. Fig. 1 shows the research framework for evaluating CO_2 capture by accelerated carbonation of steelmaking slags to identify the best available technology (BAT). The carbonation efficiency, electricity consumption, global warming potential, eco-system quality potential, and human health potential are considered to be the key performance indicators (KPIs) for proposing the BAT. The KPIs should provide a measure of current performance, a clear statement of what might be achieved in terms of future performance targets and a yardstick for measurement of future progress along the way (Jefferson *et al.*, 2007). The selected KPIs must be met the following requirements including representative, reasonably simple, sensitive to change, comprehensive and data availability (Alwaer and Clements-Croome, 2010).

According to the observations reported in the literature (Bobicki *et al.*, 2012; Chang *et al.*, 2012a; Pan *et al.*, 2014), the key operating factors affecting the carbonation reaction include reaction time, liquid-to-solid (L/S) ratio, temperature, pressure, and initial pH of solution. As a result, carbonation processes should be critically assessed under different operating conditions. Since the LCA framework was utilized for evaluation of system-wide energy and environmental footprints of CCS technology deployment (Sathre *et al.*, 2012), LCA should be employed to assess the environmental impacts of various carbonation processes.

Definition of Goal and Scope in LCA

According to ISO 14040, the goal and objectives, method, comparison basis (functional unit), process (system boundary), time and system options, and types of environmental impacts should be precisely defined in this step to ensure the validity of the results. Fig. 2 shows the scope of work for evaluating the environmental impacts of carbonation processes using alkaline wastes to capture CO_2. In this study, the pretreatment of alkaline wastes and carbonation process were employed within the system boundary. The inputs include energy (e.g., electricity) and material (e.g., water, steelmaking slag, and CO_2 source); while the outputs comprise pollutant emissions and treated wastes. The environmental impacts of each input and output material and/or energy source for carbonation processes of steelmaking slag were quantified by LCA using Umberto 5.5.4 software, with the Swiss Eco-invent 2.2 database in Umberto. Umberto (ifu hamburg, Germany) is a professional

Table 1. Summary of the selected unit processes for carbonation of alkaline solid wastes.

	Types of Carbonation	Unit processes[1]	Process Description	Reference
Scenario 1	Direct carbonation (One-step)	P1: Grinding of slag P2: Pressuring of CO_2 P3: Heating of slurry C1: Carbonation in an autoclave reactor	· Slag pretreatment · High temperature (160°C) and high pressure (700 psig)	Chang *et al.*, 2011b
Scenario 2	Direct carbonation (One-step)	P1: Grinding of slag C1: Carbonation in a slurry reactor	· Slag pretreatment · Lower reaction temperature (70°C) and ambient pressure (1 bar)	Chang *et al.*, 2011a
Scenario 3	Direct carbonation (One-step)	P1: Grinding of slag P2: Slurry stirring P3: Slurry pump C1: Carbonation in a rotating packed bed (RPB)	· Slag pretreatment · Lower reaction temperature (65°C) and ambient pressure (1 bar) · Additional energy-consumption equipment	Chang *et al.*, 2012a
Scenario 4	Indirect carbonation (Two-step)	P1: Grinding of slag C1: Extraction of calcium ions from SS[2] C2: Calcium carbonate precipitation C3: Regeneration of acetic acid	· Generating a calcium acetate solution and an SiO_2-solid residue · Lower temperature (30°C) · Chemical use (e.g., acetic acid)	Eloneva *et al.*, 2008
Scenario 5	Indirect carbonation (Two-step)	P1: Grinding of slag C1: Extraction of calcium ions from SS C2: Calcium carbonate precipitation C3: Regeneration of ammonium chloride	· Calcium silicate dissolved in an aqueous solution of NH_4Cl · Small solvent loss · Lower temperature (30°C) · Chemical use (e.g., ammonium chloride)	Eloneva *et al.*, 2012
Scenario 6	Indirect carbonation (Two-step)	P1: Grinding of slag C1: Extraction of calcium ions from SS C2: Calcium carbonate precipitation C3: Regeneration of ammonium nitrate	· Solvent recycled · Lower temperature (30°C) · Chemical use (e.g., ammonium nitrate)	Eloneva *et al.*, 2012

[1]: P is referred as Physical treatment, and C is referred as Chemical treatment; [2]: SS is Steelmaking Slag.

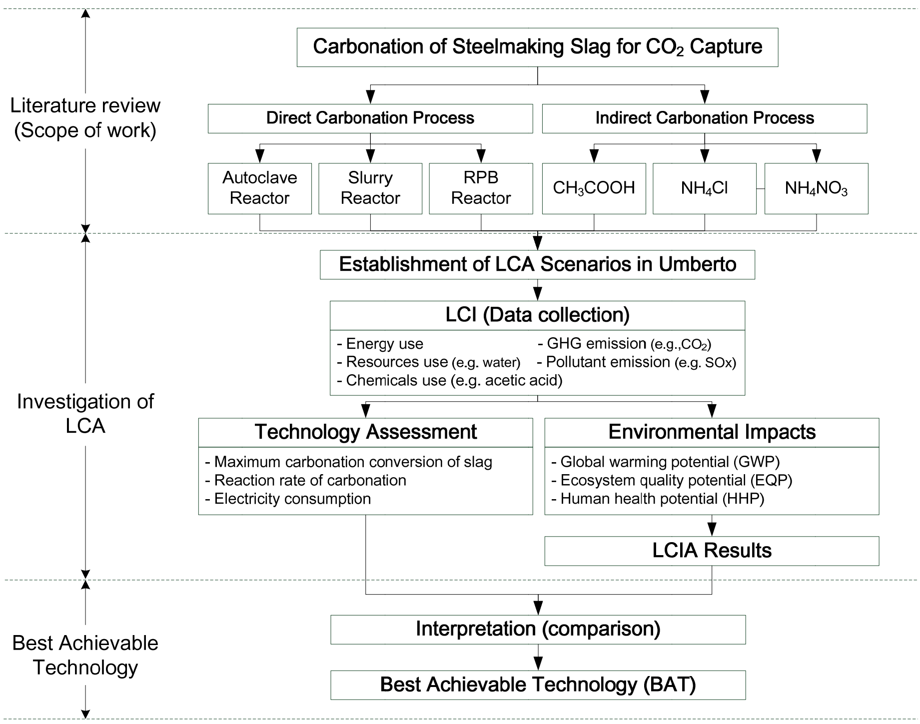

Fig. 1. Systematic comparison of carbonation processes for steelmaking slag by LCA.

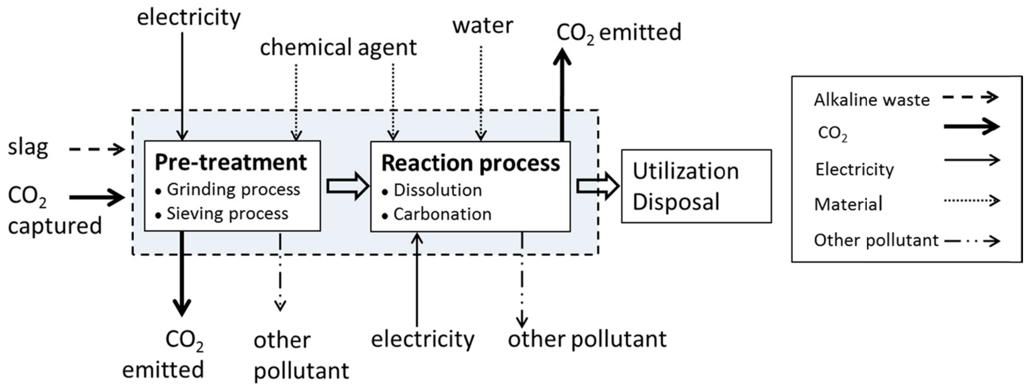

Fig. 2. System boundary of carbonation process defined for this study.

tool which is commonly utilized for computer-based material and energy flow analysis, including life cycle assessment, integrated cost accounting, and carbon footprint, to achieve successful energy management, material efficiency and process optimization (IFU and IFEU, 2005).

System Boundary and Inventory

In this study, six scenarios were established for CO_2 capture by carbonation of various steelmaking slags, including basic oxygen furnace slag (BOFS), steel converted slag (SCS), and blended hydraulic slag cement (BHC). The performance and environmental impacts of direct carbonation methods (i.e., scenarios 1 to 3) with various reactors was compared with that of indirect carbonation methods (i.e., scenarios 4 to 6) using different extraction agents. Figs. 3(a), 3(b), and 3(c) show the scenarios of direct carbonation for steelmaking slag in various types of reactors including autoclave, slurry reactor, and rotating packed bed (RPB), respectively. In cases of indirect carbonation, the dissolution of calcium ions from the solid matrix of steelmaking slag was commonly evaluated using acetic acid, ammonium nitrate, and ammonium chloride as shown in Figs. 3(d), 3(e), and 3(f), respectively.

The functional unit was set to be "1 kg CO_2 captured by carbonation process." In the Umberto program, transitions representing conversion processes are symbolized in squares, and places, i.e., the input and output of the process, are symbolized in circles. The links between the transitions and the places are established with arrows. A balance for material and energy flows could be drawn up in Umberto to calculate and analyze the investigated system boundary. The life cycle inventory (LCI) data of materials and processes within the system boundary are processed from the database in Umberto. On the other hand, the emission

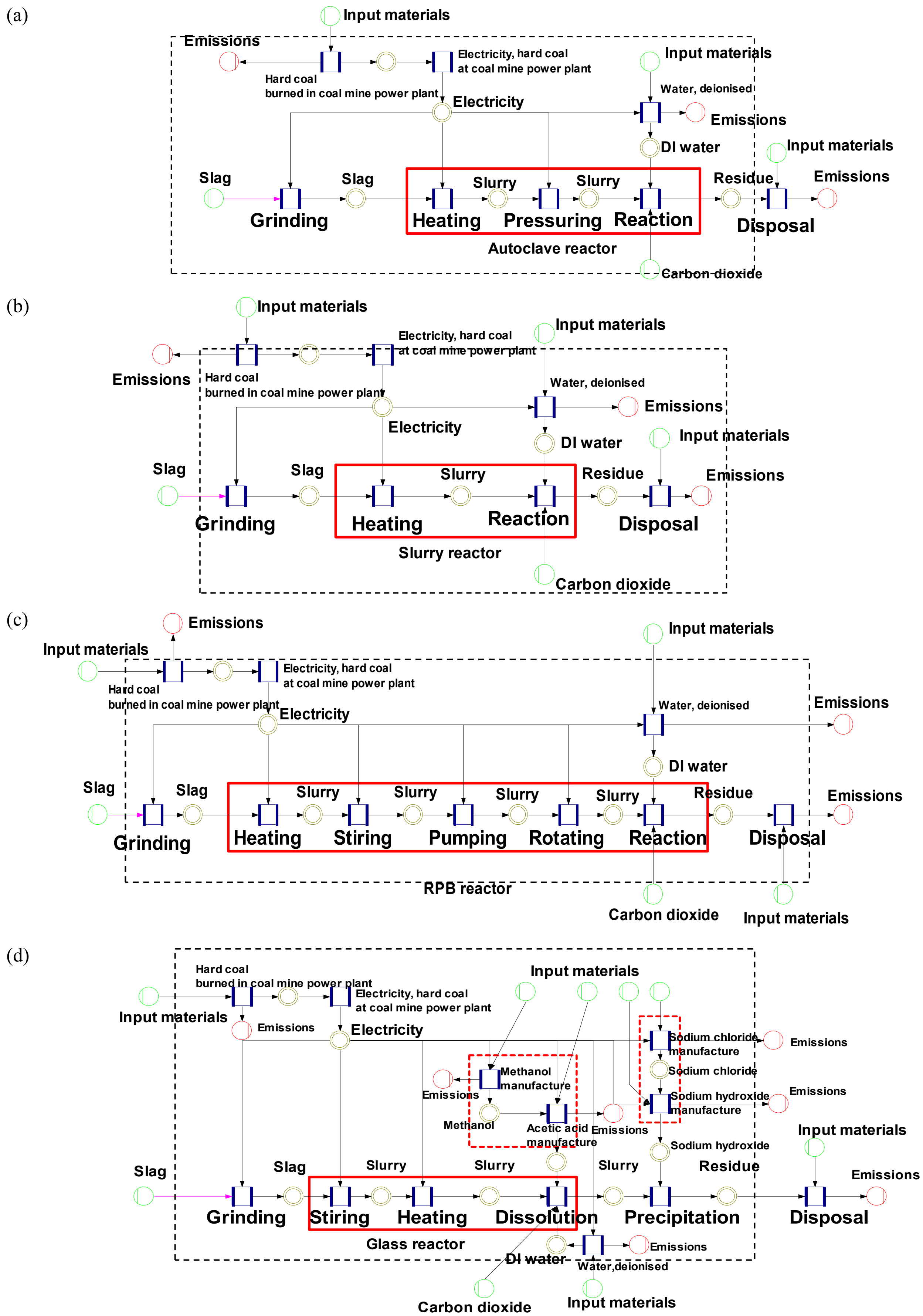

Fig. 3. Material and energy flow network of carbonation scenarios.

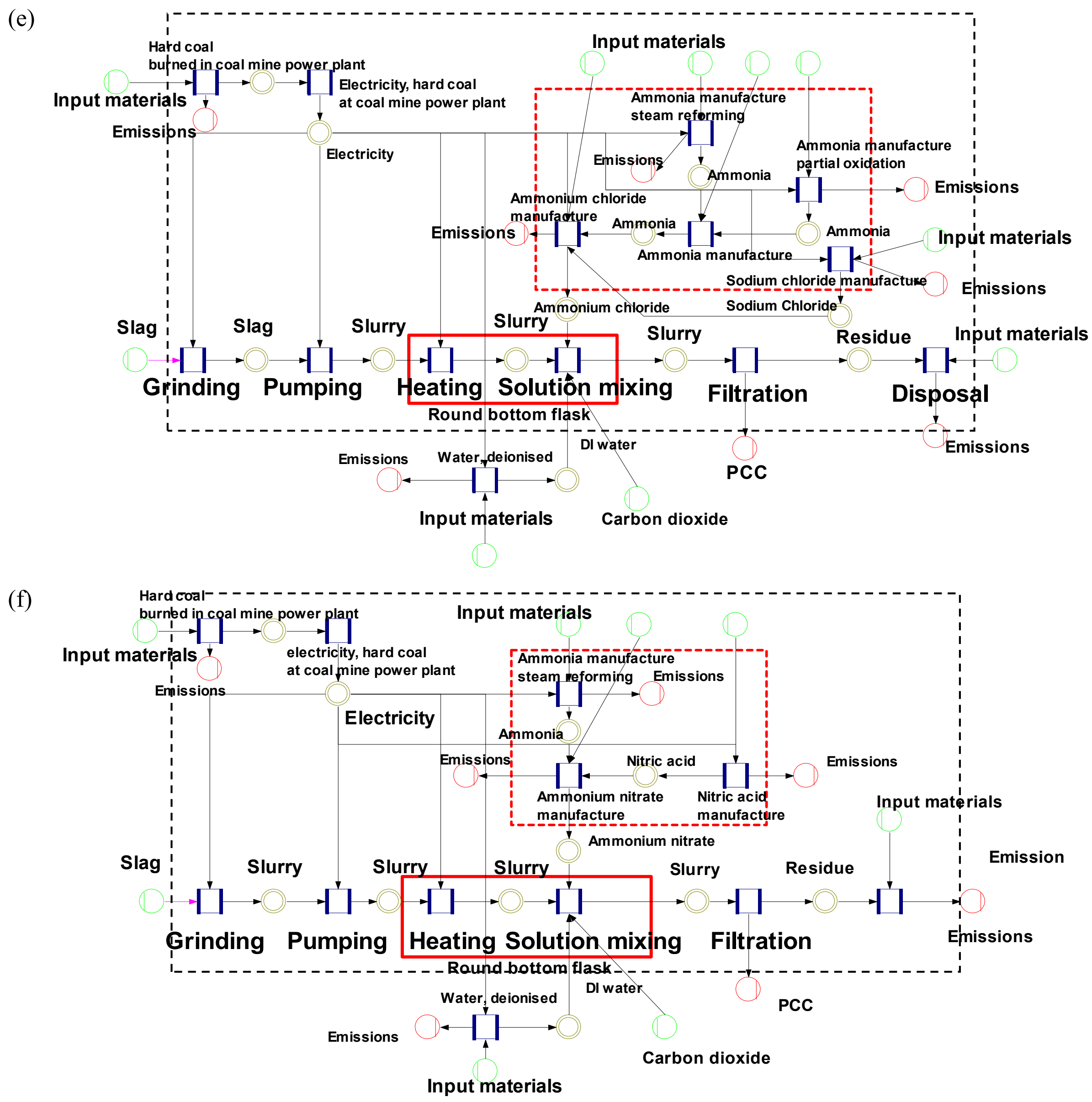

Fig. 3. *(continued).*

factors of air pollutants for CO_2, NO_x, SO_x, and PM_{10} with respect to electricity generation were assumed to be 0.612 kg/kWh, 0.446 kg/kWh, 0.493 kg/kWh, and 0.031 kg/kWh, respectively (MOEA, 2010; Tai-power Company, 2010). System input and outputs such as chemical manufacture, pollutant emissions, and energy consumption are investigated, where the residues generated from each scenario are assumed to be treated by landfill. In addition, the energy consumption of the heating process is estimated by assuming that the power for heating is equal to the heat dispersed into the surrounding environment, related to the thermal conductivity of the insulating layer.

Environmental Impact Assessment

The environmental impacts could be assessed by mid-point impact and end-point impact, of which the midpoint impact is a problem-oriented approach describing environment themes, whereas the endpoint impact is a damage-oriented approach expressing a consistent and concise view of ecosystem and human health effects. In this study, the environmental impacts for each unit operation process are evaluated and quantified using the valuation system of ReCiPe. Because ReCiPe is constructed based on both the CML (mid-point assessment approach) and Eco-indicator 99 (end-point assessment approach), it includes eighteen midpoint indicators and three endpoint indicators (De-Schryver *et al.*, 2009; Goedkoop *et al.*, 2009).

The midpoint indicators in ReCiPe include decreased ozone potential, hazardous waste dose, absorbed dose, ozone concentration, PM_{10} concentration, infra-red forcing, hazardous waste concentration for terrestrial eco-toxicity, base saturation, occupied area, transformed area, hazardous waste concentration for marine eco-toxicity, algae growth for marine eutrophication, algae growth for fresh water eutrophication, hazardous waste concentration for fresh water eco-toxicity, energy content, decreased concentration, and water use. The above indicators can be classified into three damage categories (also referred to as the mid-point indicators), i.e., human health potential, ecosystem quality potential, and ecosystem resources potential. Since the large number of midpoint indicators with a very abstract meaning is difficult to elucidate and interpret, the endpoint having three indicators with a more understandable meaning could be utilized for interpretation. In addition, the global warming potentials (GWP), reflecting the potential influence of greenhouse gases produced per kWh of electricity, over a time period of 100 years (i.e., GWP 100) was selected in accordance with the Kyoto Protocol.

RESULTS AND DISCUSSION

Technology Assessment with Respect to Engineering and Environmental Considerations

The carbonation process is an acid-base reaction, in which the carbonate ions (acid) are neutralized by the alkaline materials (base). In this study, two types of carbonation methods, i.e., direct and indirect carbonation were evaluated using the LCA. In the case of the direct carbonation process, the steelmaking slags react with the dissolved CO_2 in aqueous slurry to form the carbonate precipitations. Generally, the process chemistry of direct carbonation can be expressed as Eq. (1):

$$(Mg, Ca)_xSi_yO_{(x+2y+z)}H_{2z}(s) + xCO_2(g) \rightarrow x(Mg, Ca)CO_3(s) + {}_ySiO_2 + zH_2O(l) \quad (1)$$

Enhancement of mass transfer, suitability of mineral feedstocks, and innovation of reactor design are crucial to carbonation reaction. On the other hand, the indirect carbonation reaction can be divided into several steps, e.g., extraction of calcium ions from solid matrix into liquid solution, followed by further reaction of the calcium-rich solution with CO_2 to form the pure carbonate precipitation. The end product of indirect carbonation is precipitated calcium carbonate (PCC) with higher purity and customized morphology, which could be used as a filler and coating pigment in papers (Pan *et al.*, 2012). The process chemistry for extraction of calcium ions using acetic acid (scenario 4) can be expressed as Eqs. (2) to (4):

$$CaSiO_3 + 2CH_3COOH \rightarrow Ca^{2+} + 2CH_3COO^- + H_2O + SiO_2 \quad (2)$$

$$Ca(CH_3COO)_2 + 2NaOH \rightarrow 2CH_3COONa + Ca(OH)_2 \quad (3)$$

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \quad (4)$$

In addition, it was noted that the ammonium salt solvents could be recycled from the extraction processes, which could be considered the most promising routes for selective extraction of calcium ions from steelmaking slag. Eqs. (5) and (6) present the process chemistry of extraction using ammonium nitrate (scenario 5) and ammonium chloride (scenario 6), respectively, as follows:

$$CaSiO_3 + 2NH_4 \rightarrow Ca^{2+} + 2Cl^- + 2NH_3 + SiO_2 + H_2O \quad (5)$$

$$Ca^{2+} + 2Cl^- + 2NH_3 + CO_2 + H_2O \rightarrow CaCO_3 + 2NH_4^+ \quad (6)$$

After extraction of calcium ions, the mother liquid (i.e., rich in calcium ions) was further carbonated with the CO_2 source. Table 2 shows the required amounts of material and energy in each scenario for capturing 1 kg of CO_2 based on the maximum conversion of steelmaking slag as reported in the literature.

Since the carbonation in an autoclave reactor (scenario 1) was operated at a relatively higher pressure and temperature, scenario 1 could be the most energy-intensive one. Although the carbonation of steelmaking slags in an RPB reactor (scenario 3) exhibits the highest carbonation conversion, i.e., 93.5%, scenario 3 would consume additional energy for process operation. In comparison with the performance of an autoclave reactor (scenario 1) and a slurry reactor (scenario 2), the RPB reactor (scenario 3) exhibits a relatively higher conversion of BOFS with a relatively shorter reaction time. In contrast, indirect carbonation, i.e., scenarios 4 to 6, requires less energy; however, it needs to utilize extra chemicals such as acetic acid, which might cause adverse effects on ecosystem quality and human health. These conflicting interests in scenarios 1 to 6 suggest that the proposed BAT should be critically assessed by LCA under various operating conditions to meet the requirements for minimizing energy consumption and maximizing overall CO_2 capture capacity.

Life Cycle Impact Assessment (LCIA)

Mid-point Assessment: Global Warming Potential (GWP)

Global warming potential (GWP), causing adverse effects on ecosystem quality and human health, can be evaluated as the midpoint level in terms of CO_2 equivalence (CO_2-eq). Fig. 4 shows the results of GWP for different scenarios, which indicates that, in general, indirect carbonation would lead to lower impacts on GWP than direct carbonation. Carbonation of steelmaking slag using the autoclave reactor (scenario 1) exhibits the highest additional CO_2 emissions of all scenarios due to consuming the greatest amount of electricity for heating and pressurizing. On the other hand, indirect carbonation using ammonia chloride and ammonia nitrite as an extractant (in scenario 5 and 6, respectively) exhibits less GWP. Energy consumption for the apparatus would generate additional CO_2 emissions thereby causing a significant increase in GWP.

In addition, Fig. 5 presents the contribution of various processes to GWP in each scenario, which indicates that the heating process would cause a significant increase in GWP. In the case of direct carbonation, the electricity used for the heating process is about 99.2%, 98.6%, and 47.2% in total electricity input for scenarios 1, 2, and 3, respectively. In contrast, the electricity used for the heating process in scenarios 4, 5, and 6 is about 20.1%, 18.7% and 18.6%, respectively. It was noted that increasing the reaction temperature would accelerate the carbonation rate and improve the carbonation conversion of steelmaking slags;

Table 2. Experiment information and main input data for different carbonation processes based on functional unit.

Item		Unit	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5	Scenario 6
Experiment Information	Reactor type	-	Autoclave	Slurry reactor	RPB	Glass reactor	Round bottom flask	Round bottom flask
	Pressure	psig	700	14.7	14.7	14.7	14.7	14.7
	Temperature	°C	160	70	65	30	30	30
	Conversion	%	60.0	56.6	93.5	25.5	59.0	59.0
	Reaction time	h	2	2	0.5	2	2	2
	Particulate size	μm	44	44	22.9	125-300	500	500
Solid Wastes	Name	-	BHC	BOFS	BOFS	SCS	SCS	SCS
	CaO	%	52.82	51.11	42.43	45.90	45.90	45.90
	SiO_2	%	27.30	11.15	12.00	13.90	13.90	13.90
Inventory data	Electricity	kWh	107.65	13.27	23.29	15.89	11.44	11.44
	Solid waste	kg	4.03	4.41	3.12	10.87	4.70	4.70
	Pure water	kg	40.3	44.1	64.2	54.4	94.0	94.0
	Other Chemicals	kg	-	-	-	6.39 CH_3COOH	0.05 NH_4Cl	0.07 NH_4NO_3

* Note: RPB: Rotating Packed Bed reactor; BHC: Blended Hydraulic Slag Cement; BOFS: Basic Oxygen Furnace Slag; SCS: Steel Converted Slag.

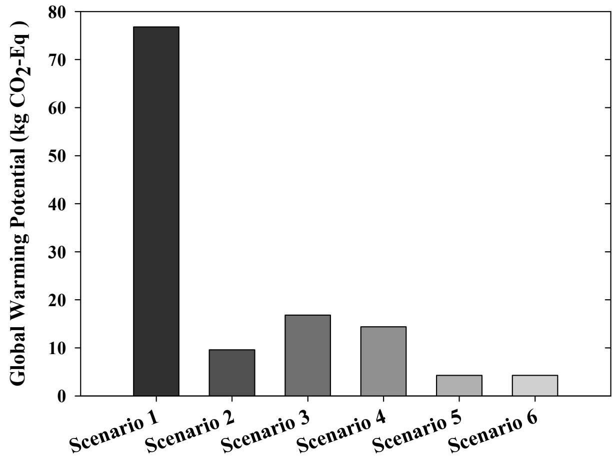

Fig. 4. Life cycle assessment results in global warming potential (GWP) for different carbonation scenarios.

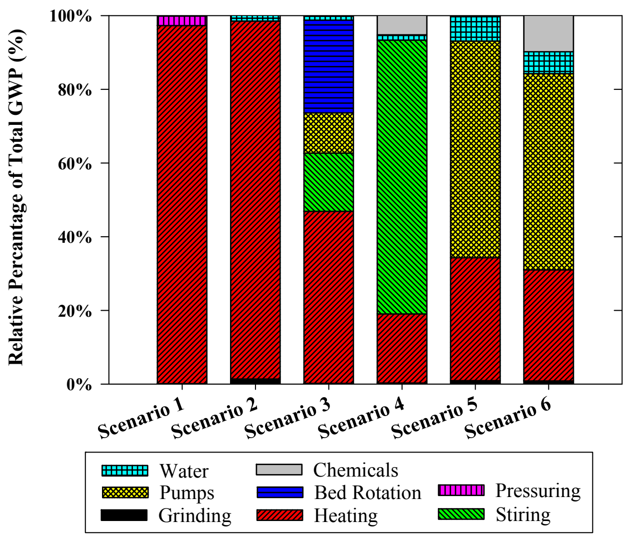

Fig. 5. Relative percentage of GWP among different transitions for each scenario.

however, it also would generate additional CO_2 emission and reduce the overall CO_2 capture capacity (Chang *et al.*, 2012b; 2013a). On the other hand, in the case of indirect carbonation, the stirring process and pumps are found to account for more than 50% of total energy use. Since the extractants were regenerated and reused in scenarios 5 and 6, the GWP proportion of chemicals accounts for only 0.3% and 9.8%, respectively. It was thus concluded that the energy consumption of the heating process for maintaining the optimum reaction temperature should be assessed systematically from engineering and environmental aspects as follows.

End-point Assessment: Ecosystem Quality Potential (EQP) and Human Health Potential (HHP)

With regard to the end-point assessment (i.e., damage assessment), the normalization factors and appropriate weights of damage were applied to convert the various impact categories into standardized importance measures. Different types of impacts within the ecosystem quality potential category are analyzed and quantified to the same dimensional unit namely PDF (Potentially Disappeared Fraction). Fig. 6 presents the results of the end-point assessment in ecosystem quality for different scenarios, in terms of PDF. The results indicated that the climate change impacts contributed more than 90% of the total damage in ecosystem quality, except for scenario 1 (approx. 70%). In addition, acidification and eutrophication are considered as the major impacts in ecosystem quality, corresponding to 3.7–4.1%. In the case of the indirect carbonation process, the impacts for chemical manufactures are relatively small compared to the generation of electricity. Compared to the GWP, the impacts of eco-toxicity contributed only a partial fraction of the PDF score. It was noted that a higher carbonation conversion of solid wastes did not exhibit a better environmental performance and/or benefits.

On the other hand, the various impacts also are normalized into human health potential in the dimension unit of DALY (Disability Adjusted Life Year). Fig. 7 presents the results of the end-point assessment in human health potential for different scenarios in terms of DALY. The formation potential of particulate matter (PM) was found to have the most significant impacts on human health, followed by the climate change potential and human toxicity potential. Because of the amount of electricity used to maintain the high-temperature and -pressure operation, the greatest human health impact was observed in scenario 1. In addition, scenario 5 was more environmentally benign due to the lower electricity consumption. This suggests that energy consumption should be regarded as the key factor affecting the direct carbonation processes (scenarios 1 to 3) according to the results of LCIA.

Determination of Best Available Technology for Carbonation

According to the investigation reported by IPCC (2007), accelerated carbonation requires a great deal of energy, resulting in a relatively higher cost. As a result, the challenges for CO_2 capture by carbonation process are to enhance the reaction rate by maximizing the CO_2 capture capacity and

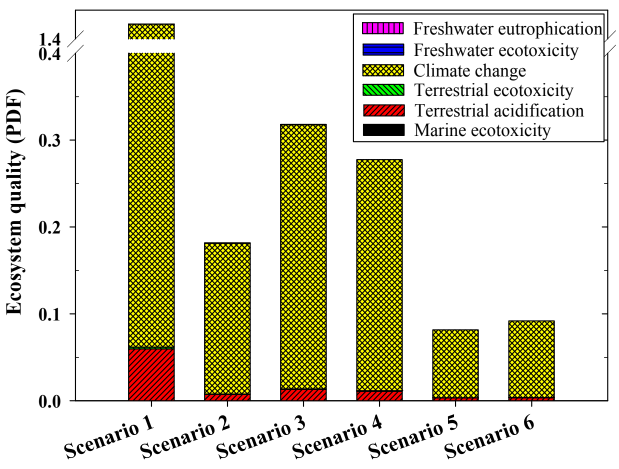

Fig. 6. Life cycle assessment results in ecosystem quality potential for different scenarios (in terms of PDF).

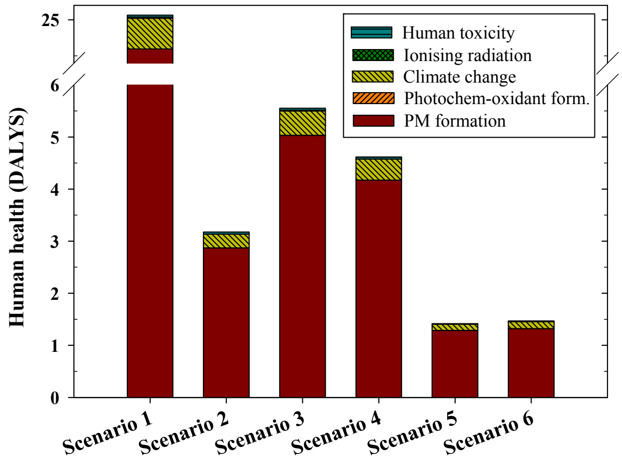

Fig. 7. Life cycle assessment results in human health potential for different scenarios (in terms of DALY).

minimizing the energy demand. Since alkaline solid wastes are generated near the source of CO_2, both environmental and economic benefits could be obtained by sequestering the portion of CO_2 emitted from the industries (Bobicki *et al.*, 2012; Pan *et al.*, 2013b). In this study, the BAT for CO_2 capture by carbonation processes of alkaline wastes was assessed by the KPIs, i.e., carbonation efficiency, electricity consumption, GWP, EQP, and HHP. In addition, the relevant uncertainties of heating efficiency on the GHG emissions also were evaluated by a sensitivity analysis for enhancing the overall thermal efficiency.

A sensitivity analysis may be undertaken to determine the effects of assessment parameters such as quantifiable data components (e.g., generation rate and composition of wastes) on the results (Lo *et al.*, 2005; Cleary, 2009). According to the results of LCIA for steelmaking slag, the pretreatment process, e.g., grinding process, exhibits less GWP, EQP, and HHP than the reaction process, e.g., heating process. Therefore, the uncertainty of heating efficiency on GWP evaluated by sensitivity analysis was presented in Table 3, which indicates that the GWP decreases as the heating efficiency increases. The effect of heating efficiency is more significant for direct carbonation processes than indirect carbonation processes. The results indicate that direct carbonation in the autoclave reactor (scenario 1) exhibited the greatest reduction in GWP with the heating efficiency increased because the reaction temperature in scenario 1 is the highest (i.e., 160°C). In addition, the GWP for indirect carbonation using NH_4Cl (scenario 5) and NH_4NO_3 (scenario 6) shows less dependence on the heating efficiency because the original energy consumption for indirect carbonation was less (ambient temperature) than thsat in direct carbonation. On the other hand, if the heating efficiency is increased by 90%, the GWP of indirect carbonation using CH_3COOH (scenario 4) would become the highest. Meanwhile, the GWP for direct carbonation in the slurry reactor (scenario 2) was the lowest among all of the scenarios. This suggests that direct carbonation in the slurry reactor should be the most attractive method, since the GWP was the lowest among the selected scenarios.

Table 3. Sensitivity analysis for heating efficiency on global warming potential (GWP) for different scenarios.

GWP ($kg/1 kg CO_2$)	Heating efficiency increased by					
	30%	50%	60%	65%	70%	90%
Scenario 1	54.41	39.49	32.03	28.30	24.57	9.64
Scenario 2	6.79	4.93	4.00	3.53	3.07	1.21
Scenario 3	14.46	12.89	12.11	11.71	11.32	9.75
Scenario 4	13.55	13.02	12.75	12.61	12.48	11.94
Scenario 5	3.86	3.57	3.43	3.36	3.28	3.00
Scenario 6	3.89	3.63	3.50	3.43	3.37	3.11

Table 4. Summary of technical assessment on the selected six scenarios according to the results of LCIA and sensitivity analysis.

	Carbonation Efficiency	Life Cycle Impact Assessment			Interpretation	Concluding remarks
		GWP	EQP	HHP		
Scenario 1	Median	High	High	High	· Heating process required the most energy (97.2%)	· Highest environmental impacts due to its high operating temperature and pressure
Scenario 2	Median	Median	Low	Low	· Heating process required the most energy (97.1%) · Lower carbonation conversion of BOFS (57%)	· The most viable process due to its relatively lower environmental impacts
Scenario 3	High	Median	Median	Median	· Heating and rotating processes are the highest energy-consumed (71.8%) · Higher carbonation conversion (93.5%)	· Higher carbonation conversion could be achieved in a short reaction time, with median environmental impacts.
Scenario 4	Low	Median	Median	Median	· Stirring process required the most energy (74.3%) · Chemicals are contributed in 5.2% GWP	· Median environmental impacts with a low carbonation conversion.
Scenario 5	Low	Low	Low	Low	· Pumps is the most energy (58.8%) · Water use is contributed in 6.6% GWP	· Lower environmental impacts with lower carbonation efficiency due to the challenge of reactor design
Scenario 6	Low	Low	Low	Low	· Pumps is the most energy (53.2%) · Water use is contributed in 6.0% GWP · Chemicals are contributed in 9.8% GWP	· Lower environmental impacts with lower carbonation efficiency due to the challenge of reactor design · Potential economic value of end product (pure-$CaCO_3$)

Table 4 summarizes the results of LCIA and sensitivity analysis, where the BAT for CO_2 capture by carbonation processes of alkaline wastes was proposed accordingly. Carbonation could be accelerated by using the RPB reactor and/or operating at a higher temperature, which requires additional electricity, raising concerns about energy issues and environmental impacts. From the viewpoint of carbonation efficiency, in most cases of direct carbonation, the reaction initially occurred rapidly, and then remained approximately constant afterward. Therefore, we could divide the entire reaction into two zones: positive CO_2 capture (Zone 1) and negative CO_2 capture (Zone 2), where the energy consumption for equipment would diminish the amount of CO_2 capture in zone 1. Therefore, the heating process would be considered for maximizing the overall thermal efficiency in cases of both direct and indirect carbonation. This suggests that additional energy consumption also should be minimized by utilizing waste heat from the industrial process.

According to the summary of LCIA results and sensitivity analysis presented in Table 4, the direct carbonation in a slurry reactor would be a viable method due to its lower environmental impacts for achieving the same level carbonation conversion. In addition, the carbonation of steelmaking slag using an RPB exhibited a higher carbonation rate under the ambient operating conditions (Chang *et al.*, 2012a, 2013b; Pan *et al.*, 2013a). Therefore, combining the slurry reactor with an RPB reactor might be a feasible process for accelerated carbonation of steelmaking slag to maximize carbonation conversion and minimize environmental impacts and additional CO_2 emissions. In that case, particle size of alkaline solid wastes, rotating speed, L/S ratio, and reaction temperature would be the key factors to optimize the overall carbonation process, which should be systematically evaluated through response surface methodology (RSM) in our future research work. In addition, since all the selected scenarios in this study were evaluated on the basis of laboratory scale, the feasibility of scaling up and the scaling factors for different types of reactors should be taken into consideration. Meanwhile, establishment of innovative technology development in reactor design and development of integrated networks for waste-to-resource supply chain for system optimization need to be further investigated prior to the deployment of carbonation processes for CO_2 capture in large scale.

CONCLUSIONS AND RECOMMENDATIONS

Since steelmaking slag was generated in potentially huge amounts near the point sources of CO_2 emissions in industries, it would be suitable for use as the feedstock for carbonation reaction. In this study, six scenarios of direct and indirect carbonation using various reactors such as an autoclave reactor, a slurry reactor, and a rotating packed bed (RPB) were selected to evaluate the environmental impacts by life cycle assessment (LCA). Employing LCA for carbonation processes would enable systematic evaluation of the energy and environmental footprints of CCS technology deployment. According to the inventory analysis, the carbonation of steelmaking slags in an RPB reactor (scenario 3) exhibited the highest carbonation conversion, 93.5%, and energy consumption for equipment such as rotating beds and pumps. On the other hand, indirect carbonation using ammonium chloride and ammonium nitrite as an extractant (in scenarios 5 and 6, respectively) would require less energy, thereby resulting in less GWP. However, extra chemicals such as acetic acid would be required, which could contribute to environmental and human health impacts.

In contrast, the HHP was found to be greatest in scenario 1 because of the large amounts of electricity used to maintain the high-temperature and -pressure operation. Energy consumption is considered the most influential factor in direct carbonation processes (scenarios 1 to 3). In addition, direct carbonation in the slurry reactor would be the most attractive method according to the results of sensitivity analysis. Furthermore, the carbonation of steelmaking slag has been proven to possess a rapid carbonation rate using an RPB. It was thus concluded that carbonation should be performed by combining the slurry reactor with an RPB reactor to maximize the carbonation conversion of steelmaking slag and minimize the environmental impacts and additional CO_2 emissions. Moreover, particle size of alkaline solid wastes, rotating speed, L/S ratio, and reaction temperature are the key factors affecting the overall carbonation process. The optimum operation guidelines should be systematically developed throughout the RSM and LCA in our future research work.

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NOMENCLATURE

BAT	Best available technology
BHC	Blended hydraulic slag cement
BOFS	Basic oxygen furnace slag
CCS	Carbon capture and storage
CDM	Clean development mechanism
DALY	Disability Adjusted Life Year
EQP	Ecosystem quality potential
GWP	Global warming potential
HHP	Human health potential
ISO	International organization for standardization
KPI	Key performance indicator
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact analysis
PCC	Precipitated calcium carbonate
PDF	Potentially Disappeared Fraction
PM	Particular matter
RSM	Response surface methodology
RPB	Rotating packed bed
SCS	Steel converted slag

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