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Engineering, environmental and economic performance evaluation of high-gravity carbonation process for carbon capture and utilization

Shu-Yuan Pan [a], Ana Maria Lorente Lafuente [b,c], Pen-Chi Chiang [a,d,*]

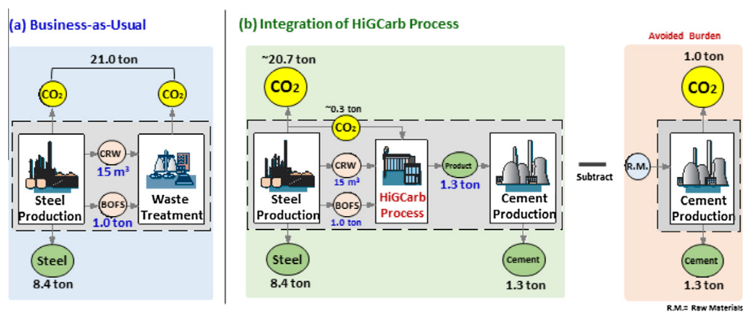
[a] Graduate Institute of Environmental Engineering, National Taiwan University, 71 Chou-Shan Road, Taipei City, Taiwan 10673, Taiwan, ROC
[b] Institute of Technical Thermodynamics, RWTH Aachen University, Schinkelstraße 8, 52062 Aachen, Germany
[c] Institute for Advanced Sustainability Studies (IASS) Potsdam, Berliner Strasse 130, D-14467 Potsdam, Germany
[d] Carbon Cycle Research Center, National Taiwan University, 71 Chou-Shan Road, Taipei City, Taiwan 10673, Taiwan, ROC

HIGHLIGHTS

- Energy consumption and CO_2 reduction potential were evaluated based on field test.
- Environmental impacts in terms of mid- and end-points were quantified using ReCiPe.
- Revenue including profits and costs was estimated at different electricity prices.
- HiGCarb can reduce up to 6.5% CO_2 emissions within the steel and cement industries.
- Best operation modulus of HiGCarb was identified using 15 KPI by 3E triangle model.

GRAPHICAL ABSTRACT

A 3E triangle model was used to evaluate tradeoffs in CO_2 capture performance by the developed HiGCarb process while considering the larger life-cycle environmental impacts due to energy use and material consumption as well as the economic implications of the revenue gained and the operating costs.



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ABSTRACT

Multi-waste treatment of slag and wastewater can be combined with CO_2 capture in the steelmaking industry by the high-gravity carbonation (i.e., HiGCarb) process using a rotating packed bed. In this study, the HiGCarb process is comprehensively evaluated by an engineering, environmental and economic (3E) triangle model. The feedstock CO_2 for the HiGCarb process can be obtained directly from the industrial stacks, eliminating the need for additional CO_2 concentration and transportation. The reacted steelmaking slag, i.e., basic oxygen furnace slag (BOFS), is suited as cement substitution material, avoiding environmental burden from the cement industry, also a CO_2-intensive emission source. Significant environmental benefits can be realized by establishing the waste-to-resource supply chain between the steelmaking and cement industries. The life-cycle assessment shows a net CO_2 capture amount by the HiGCarb process of 282 kg-CO_2/t-BOFS, accompanied by a CO_2 avoidance of 997 kg-CO_2/t-BOFS due to the product utilization. Moreover, the amount of revenue gained was estimated to be 20.2–23.2 USD/t-BOFS treated by the HiGCarb process. According to the 3E triangle model, the HiGCarb process is shown to be environmentally promising and economically feasible due to its high overall engineering performance, which makes it suitable as a potential CO_2 sink in industry.

* Corresponding author at: Graduate Institute of Environmental Engineering, National Taiwan University, 71 Chou-Shan Road, Taipei City, Taiwan 10673, Taiwan, ROC. Tel.: +886 2 23622510; fax: +886 2 23661642.
E-mail address: pcchiang@ntu.edu.tw (P.-C. Chiang).

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1. Introduction

Accelerated mineralization, so-called accelerated carbonation, refers to the reaction of CO_2 with alkaline divalent cations from natural ores [1] or alkaline solid wastes, such as steel slag [2] and fly ash [3], to produce carbonate minerals. The process chemistry of accelerated mineralization, in the presence of water, can be briefly expressed as Eq. (1):

$$MO_{(s)} + H_2O_{(l)} + CO_{2(g)} \rightarrow MCO_{3(s)} + H_2O_{(l)} \tag{1}$$

where M is a divalent cation (e.g., Ca^{2+} or Mg^{2+}), and MCO_3 is a carbonate product (e.g., $CaCO_3$ or $MgCO_3$). Since the carbonate product is stable at atmospheric conditions, accelerated mineralization has been considered as one of the permanent CO_2 sequestration technologies in a scalable manner, especially when alkaline solid wastes are used as the feedstock of the mineralization process [4–6]. Kirchofer et al. [7,8] estimated a direct mitigation potential of 1.8–23.7% of the total CO_2 emissions in the U.S., depending on the availability of alkalinity sources and the performance of the mineral carbonation. In addition, indirect CO_2 emissions from the cement industry can be avoided since the reacted product (e.g., carbonated solid wastes) can be used as supplementary cementitious materials in cement and concrete [7,9]. Compared to the total global CO_2 emissions, although the CO_2 sequestration potential by accelerated mineralization using the solid wastes is low, it offers unique application for hazardous waste remediation and provides a means for industries to produce valuable by-products from the waste while reducing the CO_2 emissions [10].

For mineralization using solid wastes, extensive efforts have been underway to evaluate the effect of various key operating factors on carbonation conversion and to characterize the carbonate product for potential utilization [11–13]. The carbonation performance, i.e., rates of metal ion leaching and CO_2 dissolution, can also be promoted by introducing wastewater from the steelmaking industries [14–16]. Since accelerated mineralization has been shown to be a diffusion-controlled reaction [9,17–19], the mass transfer rate of the CO_2 dissolution and metal ion diffusion has increased using enhanced leaching [20], autoclave [21], rotating packed bed (RPB) [14,15,22], and ultrasound [23,24].

Among the aforementioned methods, accelerated mineralization using the RPB, also referred to as the high-gravity carbonation (HiGCarb) process, is an attractive approach. The HiGCarb process achieves a high CO_2 capture efficiency (i.e., >98%) with a relatively short reaction time at ambient temperature and pressure [22]. Using the centrifugal force provided by the RPB reactor, the mass transfer rate of carbonation in the HiGCarb process was significantly greater than that using a fix packed bed. The height of a transfer unit in a liquid-film limited system (i.e., carbonation) can be reduced to 4–66 cm using the HiGCarb process [25]. In addition, the environmental impacts of the HiGCarb process were lower compared to using conventional processes, such as autoclave reactor [26].

The life-cycle assessment (LCA) approach has been used as the most suitable tool for environmental assessment of CO_2 utilization technologies and processes along their entire life cycles [27–30]. Several studies on the LCA of mineral carbonation using different processes have been published [26,31–34]. These studies employed inventory data from lab-scale experiments in contrast to our work which is based on a real industry installation. For this reason, stages of the life-cycle such as raw material production and end-product utilization were not included in the literature. Furthermore, several critical issues for the HiGCarb process such as energy consumption, net CO_2 emission reduction, indirect CO_2 emission avoidance, and cost-benefit analysis of the HiGCarb process have not been comprehensively addressed yet.

For these reasons, in this study, the HiGCarb process is systematically assessed from the perspectives of engineering, environment and economy (referred to as 3E in this paper) using a triangle model. Since the complex relationships among 3E aspects can be easily visualized on a ternary plot among different scenarios, the triangle graphical presentation can be used for evaluating key factors that are related but also complementary [35,36]. The energy consumption, net CO_2 capture amount, and environmental impacts by the HiGCarb process were evaluated by means of LCA. In addition, the revenue gained of HiGCarb was estimated by considering operating cost and process profits (such as carbon credits and product sales). Furthermore, according to the results of the comprehensive performance evaluation through the 3E triangle model, operating guidelines for the HiGCarb process were proposed.

2. Methods

2.1. Scopes and definitions of business-as-usual and HiGCarb process

To critically evaluate the benefits of integrating the HiGCarb process in the steelmaking industry, the performance before (i.e., business-as-usual case) and after integration of HiGCarb process was evaluated. Table S1 (see Appendix A) presents a comparison of business-as-usual and integration of the HiGCarb process in the steelmaking industry from the 3E aspects. In the business-as-usual case, the alkaline cold-rolling mill wastewater is neutralized and adjusted by chemical agents at a wastewater treatment plant, while the fresh basic oxygen furnace slag (BOFS) is disposed at a landfill plant. In addition, the CO_2 emitted from the processes is not captured or fixed.

In the HiGCarb process [16], the CO_2 emitted from the steelmaking industry can be directly used to neutralize alkaline cold-rolling mill wastewater. At the same time, the contents of free-CaO and $Ca(OH)_2$ in BOFS can be eliminated, which upgrades the physico-chemical properties of BOFS to be utilized as cement replacement materials [6]. In this study, the CO_2 used for the HiGCarb process was captured from the hot-stove gas at the No. 3 Blast Furnace Plant in the China Steel Corp. (Kaohsiung, Taiwan). The average CO_2 concentration of the hot-stove gas was around 28–32%. The fresh BOFS and alkaline cold rolling wastewater were used directly from the steelmaking manufacturing process. The packed-bed section of the RPB had an arithmetic mean diameter of 46.5 cm, and the axial height of the packed bed was 19.9 cm, where the stainless steel wire with a mesh size of 1 cm $\times$ 1 cm was packed. The mean thickness of the stainless steel wire was 0.3 mm. The base-case gas-(Q_G) and slurry-(Q_S) flow rates were 0.38 m^3/min and 0.33–0.56 $m^{3/}$h, respectively. For these conditions, the capture scale of the HiGCarb process was 75–170 kg CO_2 per day.

2.2. Establishment of 3E triangle model

In this study, the 3E performance of the HiGCarb process was conducted in a triangle model to provide a holistic assessment. The triangle model has been extensively utilized for assessments of new energy technologies or sustainability trends [37,38]. In this case, the 3E perspectives exhibit the equal importance (i.e., each weighting one-third) for performance evaluation. The 3E performance of each scenario results directly from the amounts of the materials and energy used such as electricity, and the performance of CO_2 capture within a certain operating period.

As shown in Fig. 1, the 3E triangle model considers the aspects of life-cycle environmental impact (LCEI) on the X axis, engineering performance (EP) on the Y axis, and life-cycle cost (LCC) on the Z

axis. Each axis, ranging from 0 to 1 in the counter-clockwise direction, is divided into five levels: very low (0–0.2), low (0.2–0.4), medium (0.4–0.6), high (0.6–0.8), and very high (0.8–1.0). Based on the above categories, the areas within the triangle can be subdivided into five zones: A (excellent), B (good), C (fair), D (poor), and E (worse). A point located in "zone A" represents an operation that offer very high performance with a very low environmental impact at a very low cost.

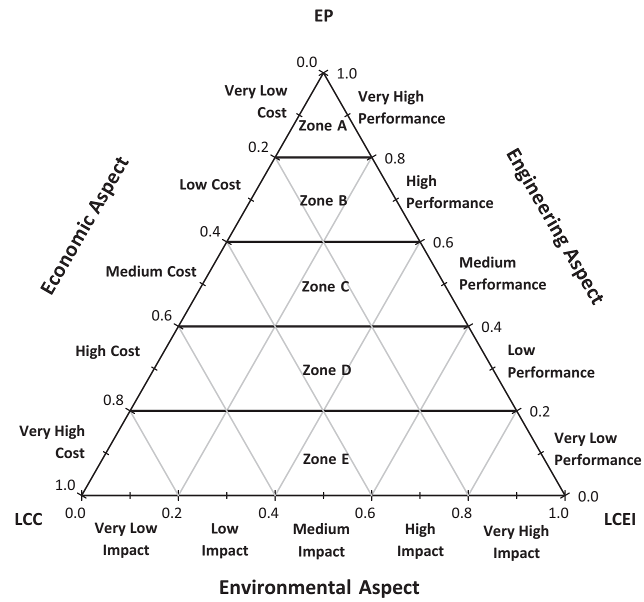

Fig. 1. 3E (Engineering–Environment–Economy) triangle model for evaluating HiGCarb process. LCEI (*X* axis): life-cycle environmental impact; EP (*Y* axis): engineering performance; LCC (*Z* axis): life-cycle cost. Zone A: excellent; Zone B: good; Zone C: fair; Zone D: poor; and Zone E: worse.

2.3. Key performance indicators for 3E triangle model

A total of 15 key performance indicators (KPIs) were selected for evaluating the entire HiGCarb process using a triangle model, as presented in Table 1. To calculate the life-cycle environmental impact (LCEI), eight environmental indicators were selected from ReCiPe midpoint/endpoint assessment due to their expected relevance for the HiGCarb process. The rest of impact categories in ReCiPe were excluded for the 3E analysis, as they did not exhibit significant difference among the scenarios. Engineering performance (EP) was calculated using three technology indicators (engineering aspect). EP_1, EP_2 and EP_3 represent the HiGCarb capacities for CO_2 removal, wastewater neutralization, and carbonated product, respectively.

To consider the process profits and operating cost, four different economic cost (EC) indicators, including the profit of direct carbon credit by the HiGCarb process ($P_{cc,\ dir}$), profit of indirect CO_2 avoidance credit by product use ($P_{cc,\ ind}$), profit of BOFS treatment avoidance (P_{ta}), profit of end-product sale (P_{ep}), and operating cost (C_{op}) were determined. The amount of revenue gained (*RG*, in terms of USD/t-BOFS) can be estimated by Eq. (2):

$$RG = (P_{cc,dir} + P_{cc,ind}) + P_{ta} + P_{ep} - C_{op} \tag{2}$$

For operating cost (EC_1), different electricity prices for industry were used by multiplying with energy consumption of process (see details in Results and Discussion). For EC_2, the price of stabilized BOFS was about 6.0 USD/ton [39], which could be considered as the profit of carbonated BOFS product sales. Since the physico-chemical properties of BOFS can be upgraded after carbonation by the HiGCarb process, it was noted that further treatment of BOFS (e.g., grinding or stabilizing processes) within the steelmaking industry can be avoided if the carbonated BOFS was utilized as supplementary cementitious materials [16]. The current treatment fee of BOFS was about 10 USD/ton [40], which can be saved in the case of the HiGCarb process (as EC_3). For EC_4, the price of carbon credit in the emission reduction unit (ERU) market in 2014 was approximately 8.1 USD/t-CO_2 [41].

The above 15 KPIs can be arranged as a data matrix for LCEI, EP, and EC indicators, as shown in Eqs. (3)–(5):

$$\mathrm{LCEI}_{yi} = (e_{yi}) = \begin{bmatrix} e_{11} & \cdots & e_{1m} \\ \vdots & \ddots & \vdots \\ e_{s1} & \cdots & e_{sm} \end{bmatrix} \tag{3}$$

$$\mathrm{EP}_{yj} = (p_{yj}) = \begin{bmatrix} p_{11} & \cdots & p_{1m} \\ \vdots & \ddots & \vdots \\ p_{s1} & \cdots & p_{sm} \end{bmatrix} \tag{4}$$

$$\mathrm{EC}_{yk} = (c_{yk}) = \begin{bmatrix} c_{11} & \cdots & c_{1m} \\ \vdots & \ddots & \vdots \\ c_{s1} & \cdots & c_{sm} \end{bmatrix} \tag{5}$$

where LCEI_{yi}, EP_{yj}, and EC_{yk} are the original data matrix for LCEI, EP, and EC indicators, respectively. "y" is the yth studied object, which is nine different scenarios with various levels of CO_2 capture performance; and i, j, and k are the i^{th} selected LCEI indicator, the j^{th} selected EP indicator, and the k^{th} selected EC indicator, respectively.

Since the KPIs include multiple dimensions and the value range of KPI data varies widely, feature rescaling is adopted to make the features independent of each other and to scale the range in [0, 1]. Two types of feature rescaling methods were applied for two different types of situations, respectively. For those of which the maximal value in a certain row represents the highest level of dimensionless feature, i.e., "1", a direct rescaling was used according to Eq. (6); for instance, a high operating cost (EC_1) represents a high impact on economic aspect. Conversely, for those of which the maximal value possesses the lowest level of dimensionless feature, i.e., "0", an inverse rescaling was used as shown in Eq. (7); for instance, a high end-product sale profit (EC_2) represents a low economic impact.

$$X'_{yj} = (x'_{yj}) = \left(\frac{x_{yj} - \min\limits_{y=1}(x_{yj})}{\max\limits_{y=1}(x_{yj}) - \min\limits_{y=1}(x_{yj})} \right) = \begin{bmatrix} x'_{11} & \cdots & x'_{1m} \\ \vdots & \ddots & \vdots \\ x'_{s1} & \cdots & x'_{sm} \end{bmatrix}$$
$$(y = 1, 2, \ldots, s; \quad j = 1, 2, \ldots, n) \tag{6}$$

$$\mathrm{T}'_{yi} = (t'_{yi}) = \left(\frac{\max\limits_{y=1}(t_{yi}) - t_{yi}}{\max\limits_{y=1}(t_{yi}) - \min\limits_{y=1}(t_{yi})} \right) = \begin{bmatrix} t'_{11} & \cdots & t'_{1m} \\ \vdots & \ddots & \vdots \\ t'_{s1} & \cdots & t'_{sm} \end{bmatrix}$$
$$(y = 1, 2, \ldots, s; \quad i = 1, 2, \ldots, m) \tag{7}$$

where X'_{yi} and T'_{yj} are the standardized data matrix for direct and inverse rescaling indicators, respectively.

The synthetic KPI indexes for LCEI, EP and LCC can be calculated by Eq. (8):

$$\mathrm{KPI}_y = \sum_{i=1}^{m} (\mathrm{KPI}_{yi'} \cdot \mathrm{W}_{yi}) \tag{8}$$

where the W_i is the weighting factor of each KPI. The weighting factors (W_i) of each KPI were determined by an ad hoc committee

Table 1
Key performance indicators (KPIs) for evaluation of HiGCarb process, and their corresponding weighting factors determined by Delphi method.

Aspects	Key performance indicators (KPI)[b]		Units	W_i[c]	Remarks
Engineering	EP_1	CO_2 capture scale by HiGCarb	t CO_2/t-BOFS	0.70	Related to technology risk
	EP_2	Wastewater neutralization by HiGCarb	m^3/t-BOFS	0.15	Related to commercialization risk
	EP_3	End-product production by HiGCarb	t/t-BOFS	0.15	Related to commercialization risk
Environmental[a]	$LCEI_1$	Global warming potential (GWP100) reduction	kg CO_2-Eq/t-BOFS	0.30	Related to ecosystem risk
	$LCEI_2$	Freshwater ecotoxicity reduction	kg 1,4-DCB/t-BOFS	0.10	Related to ecosystem risk
	$LCEI_3$	Particulate matter formation reduction	kg PM_{10}-eq/t-BOFS	0.10	Related to human health risk
	$LCEI_4$	Marine eutrophication potential reduction	kg N-eq/t-BOFS	0.10	Related to ecosystem risk
	$LCEI_5$	Urban land occupation avoidance	m^2a/t-BOFS	0.10	Related to ecosystem risk
	$LCEI_6$	Ecosystem quality improvement	points/t-BOFS	0.10	Related to ecosystem risk
	$LCEI_7$	Human health protection	points/t-BOFS	0.10	Related to human health risk
	$LCEI_8$	Resource depletion reduction	points/t-BOFS	0.10	Related to ecosystem risk
Economic	EC_1	Operating costs for capturing	US$/t-BOFS	0.30	Related to economic risk
	EC_2	End-product sale profit	US$/t-BOFS	0.20	Related to economic risk
	EC_3	Waste treatment free avoided	US$/t-BOFS	0.15	Related to economic risk
	EC_4	Carbon credit profit	US$/t-BOFS	0.35	Related to regulation risk

[a] Functional unit = one ton of BOFS input for HiGCarb process.
[b] LCEI: life-cycle environmental impact; EP: engineering performance; EC: economic cost.
[c] W_i = Weighting factor.

(expert consulting) using the Delphi method [42], a widely used and accepted method for gathering data from respondents within their domain of expertise [43,44]. The ad hoc committee comprised 30 highly informed academic (70%) and industrial (20%) experts from diverse backgrounds as well as government officials (10%) from the region. The Delphi study was conducted over a period of two month and comprised three rounds, during which the participating experts were consulted through sequential online questionnaires.

2.4. Scenario set-up and data inventory

In this study, nine scenarios were compared based on the CO_2 removal ratio (η) from the flue gas via the HiGCarb process, which was experimentally determined by Eq. (9):

$$\eta\ (\%) = \frac{(\rho_{CO_2,i} Q_{g,i} C_{g,i} - \rho_{CO_2,o} Q_{g,o} C_{g,o})}{\rho_{CO_2,i} Q_{g,i} C_{g,i}} \times 100 \tag{9}$$

where $\rho_{CO_2,i}$ and $\rho_{CO_2,o}$ (g/L) are the CO_2 mass density at the temperature of inflow and exhaust gas streams, respectively; $Q_{g,i}$ (L/min) and $Q_{g,o}$ (L/min) were the volumetric flow rate of the inlet gas and exhaust gas, respectively; and $C_{g,i}$ (%) and $C_{g,o}$ (%) were the CO_2 volume concentration in the inlet gas and exhaust gas, respectively. Table 2 presents the inventory data of the nine scenarios, where the CO_2 removal ratio (η) was categorized into low (<70%, L), medium (70–90%, M), and high (>90%, H) levels because of different operating conditions.

The power consumption of the stirring process, blowers, air compressors, pumps, and RPB reactor was determined by multiplying the operating voltage by the operating current of the existing equipment, as presented in Table S2 (see Appendix A). The energy consumption of BOFS grinding (E_G) was estimated by Bond equation [45], as shown in Eq. (10), which has been extensively used for estimating crushing energy in the literature [16,46,47].

$$E_G = \varpi_i \left(\frac{10}{\sqrt{D_{P80}}} - \frac{10}{\sqrt{D_{F80}}} \right) \tag{10}$$

where the D_{F80} (μm) and D_{P80} (μm) are the 80% passing size of feed (~3000 μm) and product (~125 μm) BOFS, respectively. The ϖ_i (kW h/ton) is the work index of ground BOFS, which can be found in the literature [16]. It was noted that, within the conventional grinding range of 25,000–20 μm, the Bond equation should give the most accurate estimation of grinding energy requirement [48].

2.5. Life-cycle environmental impact assessment

Fig. 2 shows the LCA system boundaries of the business-as-usual case and the integration of the HiGCarb process in industry. In the business-as-usual case, three existing waste sources were separately operated or treated: (1) CO_2 emissions from hot-stove stack without CO_2 capture, (2) wastewater treatment and discharge, and (3) BOFS stabilization and disposal. In contrast, in the proposed HiGCarb process, the CO_2 emitted from the hot-stove stack was directly reacted with the slurry of BOFS and wastewater in an RPB reactor. The main unit operation processes for the CO_2 capture stage by the HiGCarb process include BOFS grinding, stirring machines, blowers, air compressors, pumps, RPB reactor, CO_2 emission from the stack, and electricity generation. In addition, the stages of both raw material extraction (i.e., RPB reactor manufacturing) and product use (i.e., substitution in CEM I/42.5 Portland cement) were included. The detailed boundaries of the RPB reactor manufacturing and cement substitution processes employed in the LCA study can be found in Figs. S1 and S2 (see Appendix A), respectively.

To compare the LCA results with the business-as-usual case, the functional equivalent (unit) was one ton of fresh BOFS produced from steelmaking industry or delivered to the carbonation process. The functional unit choice allows to analyze several issues such as net CO_2 fixation amounts per unit weight of BOFS. The life-span of the HiGCarb facilities was assumed to be 20 years. The environmental impacts of the entire HiGCarb process were quantified by Umberto 5.6 using the ReCiPe Midpont (E) and Endpoint (E,A) methodology [49], which follows the LCA method described in the ISO 14040:2006 and ISO 14044:2006 International Standards [50,51]. For power generation, the air-pollutant emission coefficients of Taiwan in 2013 were used in the LCA, i.e., 0.522 kg CO_2-eq, 0.302 kg SO_x, 0.327 kg NO_x, and 0.027 kg PM_{10} per kW h [52], which were specified as CO_2, SO_2, NO_x, and PM_{10} in the Umberto model.

3. Results and discussion

3.1. Analysis of energy uses for HiGCarb process

Fig. 3 shows the energy consumption of the main unit processes in the nine HiGCarb scenarios. It should be noted that the results, herein, are referred to a different functional unit, i.e., per t-CO_2, since energy consumption is one of the major concerns to the cost effectiveness of a CO_2 capture process. The total energy

Table 2
Operating information and life-cycle data inventory including main material inputs and energy consumption for nine scenarios[a].

ID	CO_2 removal ratio	Performance and operation conditions[b]				Inventory (per t-CO_2 captured)			Engineering performance (EP)		
		η	ω	Q_G	Q_S	Material inputs (t)		Total energy (kW h)[d]	EP_1	EP_2	EP_3
		(%)	(rpm)	(m^3/min)	(m^3/h)	CRW[c]	BOFS		t CO_2/t-BOFS	m^3/t-BOFS	t/t-BOFS
L1	Low level (<70%)	42.3	158	0.38	0.33	105.2	7.01	441.4	0.143	15.0	1.142
L2		51.4	158	0.38	0.50	140.2	7.01	432.7	0.143	20.0	1.144
L3		65.1	200	0.38	0.40	81.3	6.11	356.1	0.164	13.3	1.164
M1	Medium level (70–90%)	71.3	350	0.38	0.40	78.7	3.93	263.7	0.254	20.0	1.256
M2		77.0	500	0.38	0.50	91.0	4.55	290.6	0.220	20.0	1.218
M3		86.4	450	0.38	0.33	52.2	3.48	226.4	0.287	15.0	1.287
H1	High level (>90%)	95.8	350	0.38	0.40	55.8	4.19	247.7	0.238	13.3	1.240
H2		98.3	400	0.38	0.33	47.5	3.16	204.7	0.316	15.0	1.318
H3		99.5	200	0.38	0.56	86.3	6.49	354.6	0.154	13.3	1.155

[a] Data inventory was obtained from the experiment in field test.
[b] η is CO_2 removal ratio as determined by Eq. (9), ω is rotating speed, Q_G is gas flow rate, Q_S is slurry flow rate, and L/S is liquid-to-solid ratio.
[c] CRW: cold rolling wastewater.
[d] Assumed the scale factor = 0.8.

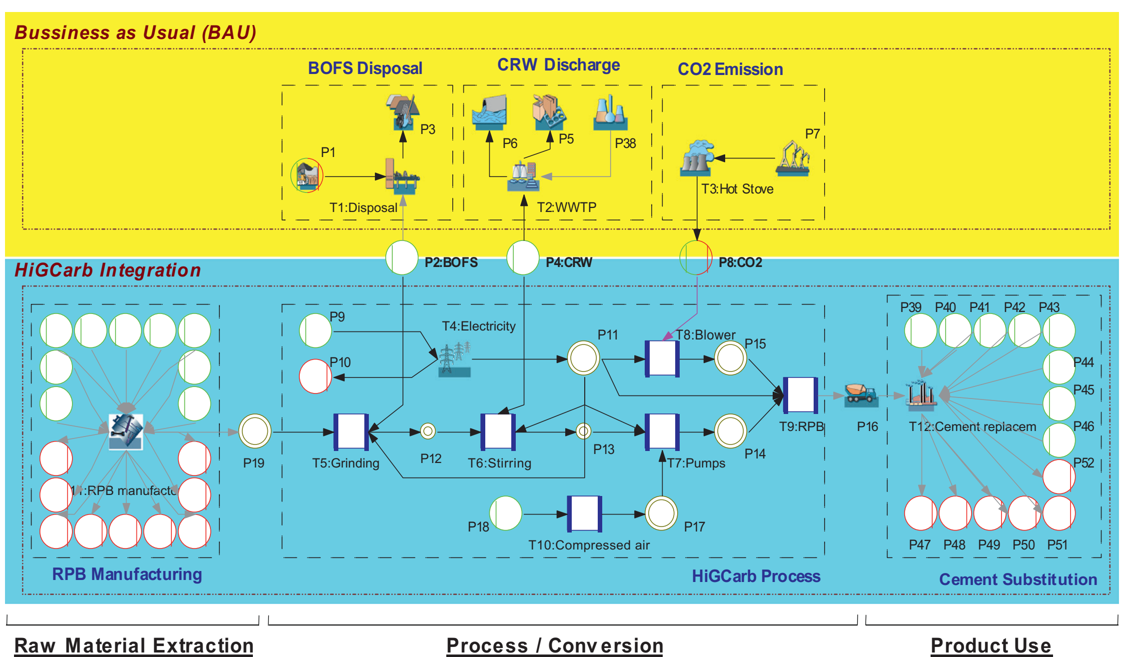

Fig. 2. System boundaries of business-as-usual (top part of the figure), and high-gravity carbonation (HiGCarb, bottom) process which includes manufacturing of the rotating packed bed (RPB) and substitution for cement.

consumption of the HiGCarb process was estimated to range from 205 to 440 kW h/t-CO_2, with a capture scale of 75–170 kg CO_2 per day. The pre-processing of material, i.e., BOFS grinding, was found to be the most energy-intensive process, contributing 65–79% of total energy consumption. However, the BOFS grinding process was required for effective and efficient carbonation reaction, as well as for further utilization of carbonated BOFS as supplementary cementitious materials in Portland cement. The energy consumption of the RPB reactor for generating a high-gravity field by centrifugal force inside the packed bed was found to be the second highest, i.e., 11–21% of total energy consumption. Since the accelerated carbonation occurred at ambient temperature and pressure in an RPB, no additional energy consumption was required to force the reaction in contrast to autoclave or slurry reactors [17,21,53,54].

In general, scenarios with low CO_2 removal ratio, e.g., L1, L2, and L3, would require a longer operating time to achieve the same CO_2 capture scale than scenarios with higher CO_2 removal ratio [22], thereby resulting in a relatively higher energy consumption for equipment operation. As shown in Fig. 3, scenario H2 exhibited the lowest energy consumption for the HiGCarb process, i.e., ~205 kW h if related to capturing one ton of CO_2. It is worthy to mention that the HiGCarb process met the criteria suggested by the U.S. Department of Energy that a cost-effective CO_2 capture facility should achieve a CO_2 removal ratio of 90%, while maintaining <35% impact on the cost of electricity [55], corresponding to a maximal energy consumption of 420 kW h/t-CO_2 [56].

3.2. Evaluation of engineering performance

Table 2 presents the operating conditions, inventory data and engineering performance for the nine scenarios. The highest specific capture capacity of BOFS (EP_1) was found in scenario H2, corresponding to 316 kg CO_2/t-BOFS. It was also noted that the specific

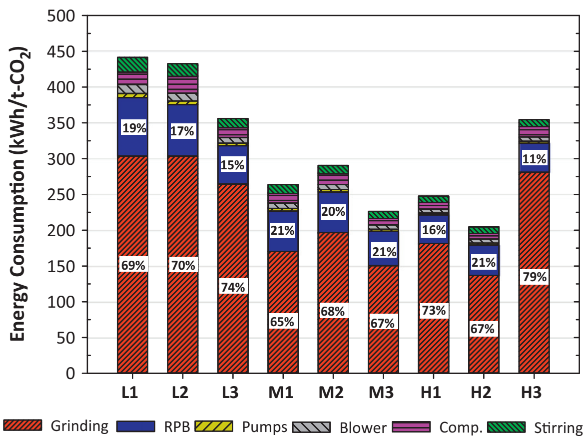

Fig. 3. Energy consumption of main unit processes in HiGCarb process for different scenarios, with their corresponding contribution in percentage. Number within each bar stack represents contribution percentage.

capture capacity of BOFS depends on the operating conditions, and was not directly correlated to CO_2 removal ratio. For example, although CO_2 removal ratio in scenario H3 was the highest (99.5%), the EP_1 of scenario H3 was the second lowest (~154 kg CO_2/t-BOFS) of the nine scenarios. However, in scenario H3, a relatively greater amount of BOFS (i.e., low *L/S* ratio) and higher slurry flow rate were used, the CO_2 capture scale of the entire HiGCarb system (EP_2) was promoted, i.e., 56.8 t-CO_2/yr, to be the top three high in the nine scenarios. In other words, the EP_2 should be mainly relevant to the amount of BOFS introduced into HiGCarb system per unit time.

For the amount of wastewater neutralization (EP_3), scenario H3 exhibited the highest treatment capacity (~4906 m^3/yr), followed by both scenarios L2 and M2 (4380 m^3/yr). The EP_3 was not related to CO_2 removal ratio but replied on the slurry flow rate and *L/S* ratio. For the amount of carbonated product (EP4), scenario 3 also exhibited the highest capacity (~425.7 t/yr), followed by scenario H1 (~326.3 t/yr) and scenario L3 (~306.6 t/yr). It was noted that the reacted product can be used as supplementary cementitious materials in blended cement, where several mortar properties, such as early-stage compressive strength and soundness can be further enhanced [6].

3.3. Quantification of environmental impacts

The environmental impacts of business-as-usual (without the HiGCarb process) and the HiGCarb process were compared by means of LCA, including manufacturing and operation of the RPB reactor and end-product use as cement substitution materials. The environmental impacts of RPB reactor manufacturing and cement substitution processes can be found in Table S3 (see Appendix A). For the assumed life time of the system of 20 years, the impact of the RPB manufacturing process and its maintenance could be neglected because the magnitude of environmental impacts by RPB production is 10^4–10^5 times less than that of operating processes.

Fig. 4 shows the global warming potential (GWP) of each scenario as determined by LCA. The GWP were calculated by considering all the CO_2-equivelent emissions of each piece of equipment from the entire life cycle of the HiGCarb process. The CO_2 capture capacity per ton of BOFS by carbonation reaction ranged from 140 kg to 320 kg according to the direct measurement of CO_2 reduction amounts in the flue gas. However, the CO_2 capture amounts could be offset by the increase in the energy consumption due to both the manufacturing and operation of additional equipment, causing additional CO_2 emissions.

On the other hand, the reaction product (i.e., carbonated BOFS) from the HiGCarb process can be used as a substitution material in blended cement, thereby resulting in additional avoidance of CO_2. It is worth mentioning that cement production is a CO_2-intensive process, in which roughly 0.73–0.99 tons of CO_2 are generated for one ton of cement production [57]. To account for the environmental impact of cement substitution stage, the avoided burden method [50,51] has been applied. As shown in Fig. 4, a significant amount, approximately 0.87–1.00 ton, of CO_2 emission can be indirectly avoided by utilization of the carbonated BOFS as a supplementary cementitious material. The highest GWP reduction was found to be ~1279 kg CO_2-eq per t-BOFS in scenario H2, where the additional CO_2 emissions from the HiGCarb process due to electricity consumption (i.e., 33.8 kg CO_2-eq) were lower than that of being captured by the HiGCarb process (i.e., 316.7 kg CO_2-eq). This suggests that the HiGCarb process was able to serve as a "real" CO_2 capture technique from the viewpoint of LCA.

Fig. 5 illustrates the results of the endpoint assessment on ecosystem quality, human health, and resource depletion for different scenarios by the ReCiPe methodology. The endpoint impacts of business-as-usual (as presented in the red bars) also differ due to the various initial material flows, i.e., wastewater discharge and CO_2 emission, for the nine scenarios. It was observed that the particulate formation (PM) potential of the HiGCarb process (as indicated by yellow[1] bars) was significantly higher than that of the business-as-usual case (as indicated by red[1] bars) because the HiGCarb process would consume additional electricity, resulting in a greater human health impact for all scenarios. Nevertheless, the adverse impact on human health due to process electricity usage can be compensated by the utilization of carbonated BOFS as supplementary cementitious materials (as indicated by green[1] bars). For this reason, in the case of CO_2 removal ratio higher than 75%, the net endpoint impact (as indicated by white bars) can eventually be reduced by up to 12.4 points (e.g., scenario M2) over that of business-as-usual case. However, the net endpoint impact was still relatively higher in scenarios of low CO_2 removal ratio (e.g., L1 and L2). This suggests that the proposed HiGCarb process can reduce not only GHG emission but also environmental impacts on ecosystem quality, human health, and resource depletion according to the LCA results.

3.4. Estimation of process profit and operating cost

Fig. 6 shows the effect of CO_2 removal ratio on operating costs and revenue gained (*RG*). Three different levels of average electricity price for industrial use in 2013 were selected for evaluation, i.e., 0.091 USD/kW h (represents a low industrial electricity price such as Taiwan, denoted as Case A), 0.168 USD/kW h (represents a medium industrial electricity price such as Germany, denoted as Case B), and 0.319 USD/kW h (represents a high industrial electricity price such as Italy, denoted as Case C). The results indicated that the operating cost of the HiGCarb process was 5.4–5.9 USD for processing one ton of BOFS, in the case of the low electricity price. In Case C, the operating cost of the HiGCarb process would increase to 19.0–20.8 USD per one ton of BOFS input.

The profit returns from direct and indirect carbon credit were estimated to be 8.3–10.1 USD per ton of BOFS input to the HiGCarb process. Furthermore, no additional CO_2 storage cost is needed for the HiGCarb process, since the CO_2-based mineral product can be directly used as substitution materials for CEM I/42.5 Portland cement in the cement industry. Therefore, the total profits returned including carbon credit and BOFS-related returns were around 25.8–29.0 USD per ton of BOFS input to the HiGCarb pro-

[1] For interpretation of color in Fig. 5, the reader is referred to the web version of this article.

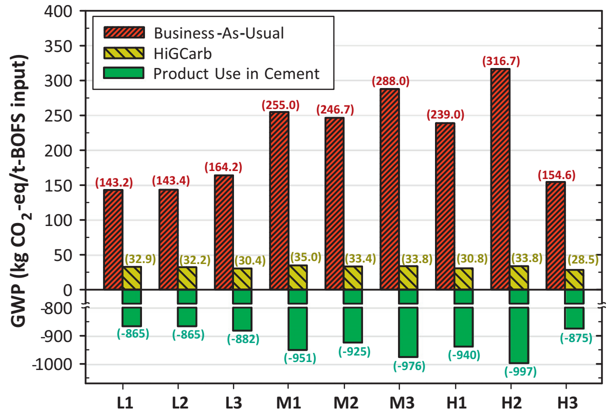

Fig. 4. Direct (CO_2 capture) and indirect (CO_2 avoidance) reduction on global warming potential (GWP) in each scenario for one ton BOFS input to HiGCarb process. HiGCarb: carbon capture and utilization by mineralization using an RPB (following ReCiPe midpoint methodology).

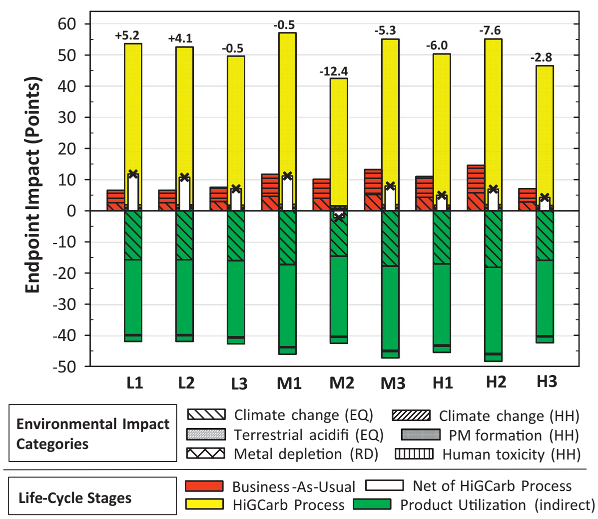

Fig. 5. Endpoint assessment including ecosystem quality (EQ), human health (HH), and resource depletion (RD) on different scenarios. HiGCarb: carbon capture and utilization by mineralization using an RPB. The numbers (Points) on each bar represent the difference between business-as-usual and HiGCarb process (following ReCiPe endpoint methodology).

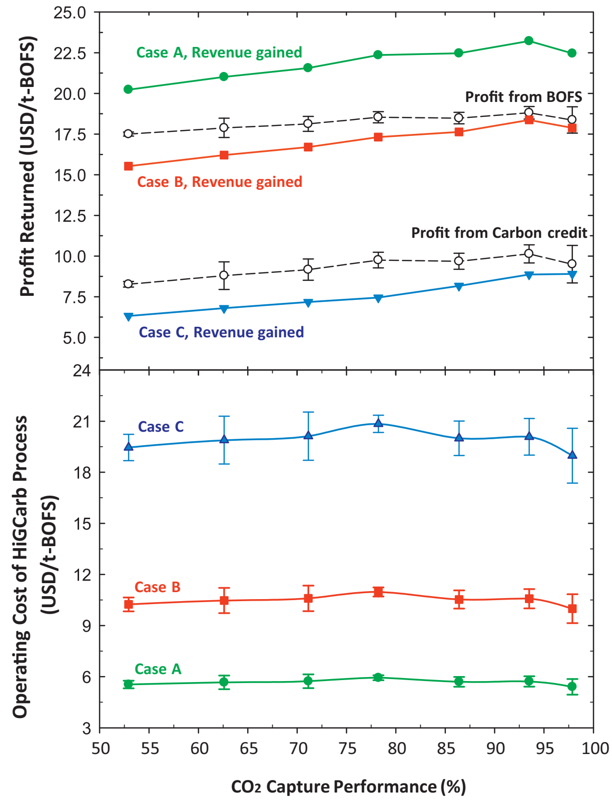

Fig. 6. Effect of CO_2 removal ratio on operating costs and revenue gained. Case A: electricity price = 0.091 USD/kW h (represents the one with low electricity price); Case B: electricity price = 0.168 USD/kW h (represents the one with medium electricity price); and Case C: electricity price = 0.319 USD/kW h (represents the one with high electricity price). *RG was calculated by Eq. (2).

cess. According to the above analysis, the revenue was estimated to be 20.2–23.2 USD per ton of BOFS input (in Case A) by Eq. (2), where the highest revenue was gained with a CO_2 removal ratio greater than 93%.

3.5. Determination of best operating model using 3E triangle model

A 3E triangle model provides a holistic assessment from the view point of environmental, engineering, and economic aspects for the HiGCarb process using a graphical presentation. The total scores for the 3E aspects were summed up after multiplying each KPI by its corresponding weighting factors (W_i) determined by the ad hoc committee using the Delphi method. As shown in Fig. 7, the results indicate that scenario H2 exhibited a superior engineering performance (as indicated by 1) with a relatively lower environmental impact (as indicated by 2) and a relatively lower cost (as indicated by 3). Although the CO_2 removal ratio of scenario H3 was the highest (i.e., 99%), the high quantity of BOFS input eventually resulted in medium environmental impact and economic cost. On the other hand, the effects of CO_2 removal ratio (η) on economic costs was not significant because the LCC were typically located at levels between 0.25 and 0.40. A poor engineering performance (scenarios L1, L2 and L3) is typically accompanied by medium capture costs but also relatively higher environment impacts. In other words, an increase in CO_2 removal ratio should be able to simultaneously reduce the environmental impacts, which make integration of the HiGCarb process into the steelmaking industry more environmentally friendly.

The annual production of BOFS in China Steel Corp. was approximately 1.2 Mt, which should be treated and/or utilized [58]. By applying the HiGCarb process under scenario H2, the annual direct CO_2 reduction using BOFS is estimated to be 0.33 Mt, corresponding to a reduction potential of 1.5% in total CO_2 emission from the studied steelmaking industry. In addition, the annual indirect CO_2 reduction due to substitution of carbonated BOFS for cementitious materials is roughly 1.05 Mt, accounting for ~5% in total CO_2 emission from the steelmaking industry. Therefore, a CO_2 reduction potential of up to 6.5% in total CO_2 emission from the steelmaking industry could be achieved. For the rest of the CO_2 emission from the steelmaking industry, several post-combustion CO_2 capture technologies, such as aqueous amine absorption [59,60] and sorbent-based adsorption [61], could be combined with the HiGCarb process as a portfolio solution. In each circumstance, the captured CO_2 would need to be further sequestered in geological formations or be utilized, which should also be critically assessed via the proposed 3E triangle model.

In this study, a waste-to-resource supply chain between the steelmaking and cement industries could be established via the HiGCarb process to reduce the overall CO_2 emissions. In the steelmaking industry, the waste treatment could be avoided, at the same time produce a valuable product as green cement. To minimize energy

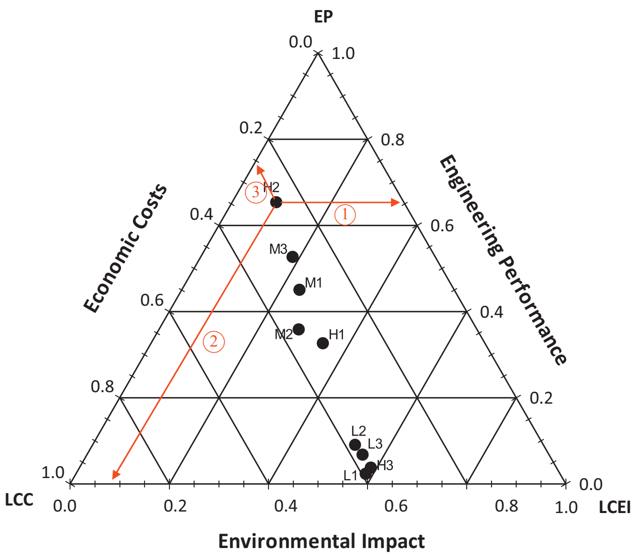

Fig. 7. Performance evaluation of HiGCarb process for different scenarios using 3E triangle model.

and material used for carbonation, the key strategies are to accelerate the reaction and exploit the heat of reaction. It is noted that both the BOFS grinding and RPB rotation are the major energy consuming processes in the HiGCarb system. If the residual heat in the flue gas could be utilized and maintained at 60–80 °C [9], the capture performance of the HiGCarb system should be significantly enhanced. In addition, other air pollutants in flue gas, such as particulate matter (PM) and sulfur dioxide (SO_2), might slightly reduce. As a result, the additional environmental benefits could be gained due to co-removal of PM and SO_2 using the HiGCarb process, which will be addressed in our future research.

4. Conclusions

To develop and implement the waste-to-resource supply chain between the steelmaking and cement industries, a comprehensive performance evaluation on the high-gravity carbonation (HiGCarb) process was carried out by the 3E triangle model, i.e., from the engineering, environmental and economic point of view. The capture capacity of the HiGCarb process was 75–170 kg CO_2 per day, associated with an energy consumption ranging from 205 to 440 kW h/t-CO_2. Compared to the business-as usual case, the environmental impacts can be substantially reduced by the HiGCarb process, e.g., a decrease in global warming potential and endpoint categories up to ~1279 kg CO_2-eq/t-BOFS and 12.4 points/t-BOFS, respectively. Furthermore, the carbon credit profits were estimated to be 8.3–10.1 USD/t-BOFS input to the HiGCarb process, assuming a price of one ton CO_2 at 8.1 USD on the ERU system in 2014. Based on the 3E triangle model, the optimal operating modulus should be the H2 scenario (see details in Table 2), where a CO_2 reduction potential of up to 6.5% in total CO_2 emission from the steelmaking industry could be achieved. An integrated approach to the proper utilization of alkaline wastes (i.e., wastewater and steelmaking slags) that permanently fixes CO_2 from the steelmaking industry while producing valuable supplementary cementitious materials from the cement industry can be achieved via the HiGCarb process.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.apenergy.2016.02.103.

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