Chapter 19
System Optimization

Abstract Since the accelerated mineralization process would consume additional energy and generate more CO_2 emissions, it should be critically assessed from the view point of 3E (Engineering, Environmental, and Economic) aspects. From the process design point of view, dissolution of reactive species (e.g., calcium ions) and the water solubility of CO_2 exhibit contradictory performances in the limiting step. Therefore, the optimum operating conditions of the accelerated carbonation process should be proposed to compromise the above conflicting phenomena. In this chapter, two different approaches, i.e., (1) mathematical programming and (2) graphical presentation, are illustrated for evaluating the engineering performance of process. To provide a holistic assessment, the 3E triangle analysis is discussed with a case study of high-gravity carbonation using steel slag. Furthermore, in this chapter, the pilot studies and demonstration projects of accelerated carbonation around the world are reviewed. The performance of accelerated carbonation is illustrated from the engineering (CO_2 capture scale and efficiency, and product utilization), economic (energy consumption and operating cost), and environmental (impacts and benefits) aspects.

19.1 Mathematical Programming Approach

19.1.1 Principles

A nonlinear program can be formulated to estimate the maximum (or minimum) objective function, e.g., carbonation conversion of alkaline solid waste. For example, the effect of different operating factors, such as reaction temperature (noted as A) and liquid-to-solid ratio (noted as B), on the carbonation conversion of alkaline solid waste (noted as δ) can be expressed as follows:

$$\delta = f(A, B) \quad (19.1)$$

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Since the δ is a function of various operating factors with interaction terms, the optimality of δ can be determined by a nonlinear program with several equations for the operating factors.

According to the Weierstrass theorem [1, 2], δ has a global optimality in set S only if

- δ is continuous on feasible S.
- S is closed and bounded.

Considering the objective of maximizing δ (A, B) subject to equality constraint h_i (A, B) converting from inequality constants s_{i}^2, it is convenient to write these conditions in terms of a Lagrange function defined as L (A, B, u) in Eq. (19.2):

$$L(A,B,u,s)=\delta(A,B)+\sum_{i=1}u_ih_i(A,B,s) \tag{19.2}$$

where u_i is the Lagrange multipliers, which can be either positive, negative, or zero. In Eq. (19.2), the maximum value of $\delta(A, B)$ should be equal to maximum $L(A, B, u, s)$, if the constraints were satisfied. To meet the necessary conditions of the Lagrange Multiplier Theorem ($L'(x^*) = 0$ and $L''(x^*) < 0$), the gradients of $L(A, B, u, s)$ function can be determined as follows:

$$L_A(x)|_{x=x*}=\frac{\partial\delta(x^*)}{\partial A}+\sum_{i=1}u_i^*\frac{\partial h_i(x^*)}{\partial A}=0 \tag{19.3}$$

$$\nabla L_B(x)|_{x=x*}=\frac{\partial\delta(x^*)}{\partial B}+\sum_{i=1}u_i^*\frac{\partial h_i(x^*)}{\partial B}=0 \tag{19.4}$$

$$\nabla L_u(x)|_{x=x*}=h_i(x^*)=0 \tag{19.5}$$

$$\nabla L_s(x)|_{x=x*}=u_i^*\frac{\partial h_i(x^*)}{\partial s}=0 \tag{19.6}$$

where x^* is the optimal solution for the carbonation of alkaline solid waste. Any point that does not satisfy the conditions of the Lagrange multiplier theorem cannot be a maximum point.

In addition, the obtained results should meet the sufficient conditions of optimality by taking the Hessian matrix of Eq. (19.2), as shown in Eq. (19.7):

$$H(x)|_{x=x*}=\begin{bmatrix}\frac{\partial^2 L}{\partial A^2} & \frac{\partial^2 L}{\partial A\partial B}\\ \frac{\partial^2 L}{\partial B\partial A} & \frac{\partial^2 L}{\partial B^2}\end{bmatrix} \tag{19.7}$$

19.1.2 Application: Case Study of Carbonation in a Slurry Reactor

From the statistical point of view, the response surface methodology (RSM), as illustrated in Chap. 9, can be used to evaluate the effect of the relating operational parameters, including reaction time, temperature, and L/S ratio on the carbonation conversion of alkaline solid wastes. For example, the carbonation conversion of steel slag (i.e., basic oxygen furnace slag, BOFS) in a slurry reactor can be expressed as Eq. (19.8) [3]:

$$\delta(\%) = 45.56 + 8.97A + 6.45B - 2.55AB - 5.36A^2 - 0.91B^2 \tag{19.8}$$

where A is the reaction temperature and B is the L/S ratio. By solving Eqs. (19.3)–(19.6), eight optimal candidate points may be theoretically obtained as a solution. In the case of $h_1 = 0$, for instance, the conditions can be determined by Eqs. (19.9)–(19.11):

$$\nabla L_{A,B}(\mathrm{x})\big|_{x=x*} = \begin{bmatrix} \frac{\partial L}{\partial A} \\ \frac{\partial L}{\partial B} \end{bmatrix}_{x=x*} = \begin{bmatrix} -10.72A - 2.55B + 8.97 \\ -2.55A - 1.82B + 6.45 + u_1 \end{bmatrix}_{x=x*} = \begin{bmatrix} 0 \\ 0 \end{bmatrix} \tag{19.9}$$

$$\nabla L_u(x)\big|_{x=x*} = \frac{\partial \mathrm{L}}{\partial u_1} = B - 1 + s_1^2 = 0 \tag{19.10}$$

$$\nabla L_s(x)\big|_{x=x*} = \frac{\partial L}{\partial s_1} = 2u_1 s_2 = 0 \tag{19.11}$$

In this case, the temperature and L/S ratio can be determined to be 62 °C (i.e., $A = 0.6$) and 20 mL g^{-1} (i.e., $B = 1.0$), respectively. The s_1 and u_1 values were 0 and −3.1, respectively. Moreover, the obtained results should meet the sufficient conditions of optimality by taking the Hessian matrix of Eq. (19.2), as shown in Eq. (19.12):

$$H(x)\big|_{x=x*} = \begin{bmatrix} \frac{\partial^2 L}{\partial A^2} & \frac{\partial^2 L}{\partial A \partial B} \\ \frac{\partial^2 L}{\partial B \partial A} & \frac{\partial^2 L}{\partial B^2} \end{bmatrix} = \begin{bmatrix} -1.82 & -2.55 \\ -2.55 & -10.72 \end{bmatrix} < 0 \tag{19.12}$$

Since the function of carbonation conversion should be negative definite, the determined condition of Eq. (19.9) results in the maximal value. As shown in Fig. 19.1, it was predicted that the maximum carbonation conversion of BOFS in a slurry reactor for 120 min should be 53.0%. It was noted that the carbonation reaction of steel slag can be enhanced if the alkaline wastewater was introduced as a liquid agent due to the presence of sodium and chloride ions in the wastewater [4, 5].

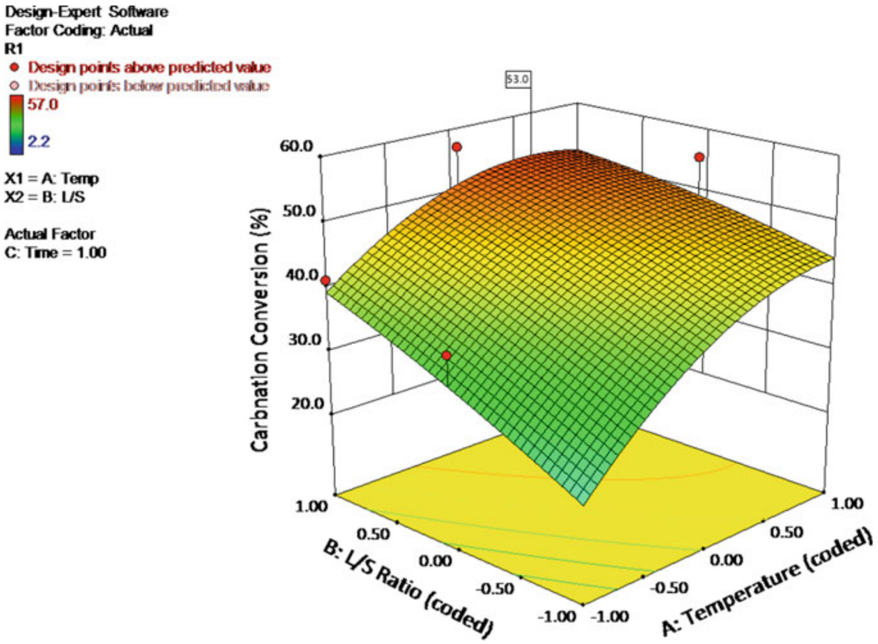

Fig. 19.1 3D response surface plot of various operating factors and conversion of steel slag in a slurry reactor

19.2 Graphical Presentation for Optimization

19.2.1 Maximum Achievable Capture Capacity (MACC)

The maximum achievable capture capacity (MACC) of solid wastes after accelerated carbonation can be obtained by considering both the carbonation conversion and energy consumption. It is noted that the carbonation kinetics can be expressed by an “exponential growth to maximum” model due to the formation of product layer, during the carbonation reaction, as reported in the above-mentioned results. However, the overall energy consumption of batch carbonation process was found to increase linearly with the increase in reaction time. Therefore, the MACC of solid waste can be systematically determined and graphically presented in Fig. 19.2 by balancing “exponential growth of capture capacity (positive capture)” and “linear increase of energy consumption (negative capture).”

The major unit operation processes for accelerated carbonation typically include grinding, transportation, stirring, pumps, blowers, reactor, and liquid–solid separation. The power consumption for most of the unit operations can be determined by multiplying the operating voltage to the operating amplitude of the existing equipment, while the power consumption (W) for grinding (crushing) can be

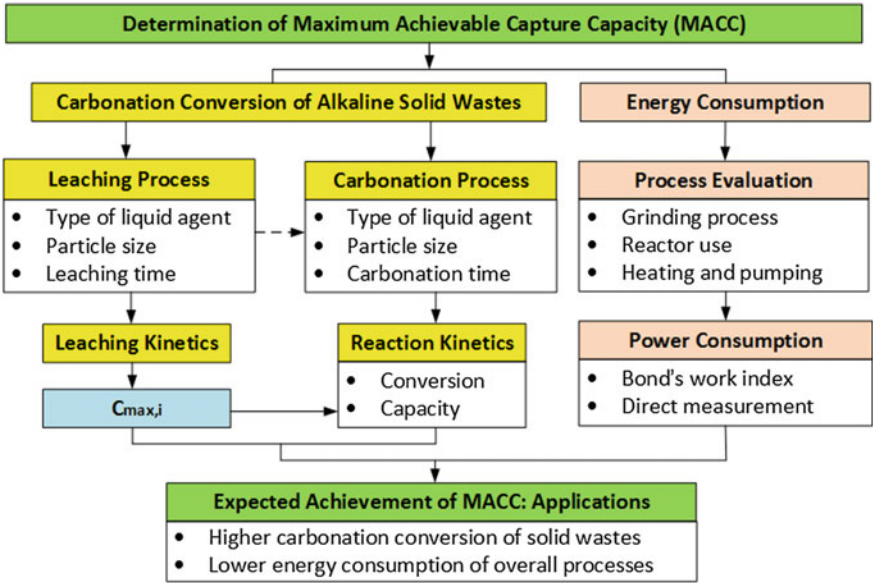

Fig. 19.2 Systematic approach to determining maximum achievable capture capacity (MACC) via leaching and carbonation processes

calculated by Bond's equation as shown in Eq. (19.13) [6]. The Bond's equation has been widely used in the literature [7, 8]:

$$W = W_i \left(\frac{10}{\sqrt{W_{P80}}} - \frac{10}{\sqrt{W_{F80}}} \right) \tag{19.13}$$

where W (kWh/ton) is the power consumption, W_{F80} (μm) and W_{P80} (μm) are the 80% passing size of the feed and the product, respectively, and W_i (kWh/ton) refers to the work index of ground material. For example, the work index for the slag obtained from the ball-mill test is approximately 30.4 kWh/ton [8].

Since the energy consumption of processes increases as the operating time increases, the overall MACC of solid wastes should be achieved at the "maximum point" by considering both the carbonation rate and energy consumption. As shown in Fig. 19.3, for example, in the case of steel slag using alkaline wastewater in a high-gravity carbonation, the operating time for reaching the MACC should be 8.5 min (from Point 1 to Point 2, and then we can obtain the Point A). In that case, the required amount of steel slag for capturing 1 ton of CO_2 by the carbonation process was estimated to be 5.13 ton (from Point 3 to Point 5, and then we can get the Point B), under which the MACC was approximately 0.20 ton CO_2 per ton steel slag. The carbonation of steel slag coupled with alkaline wastewater (i.e., cold-rolling wastewater) with a particle size less than 125 μm exhibits a relatively

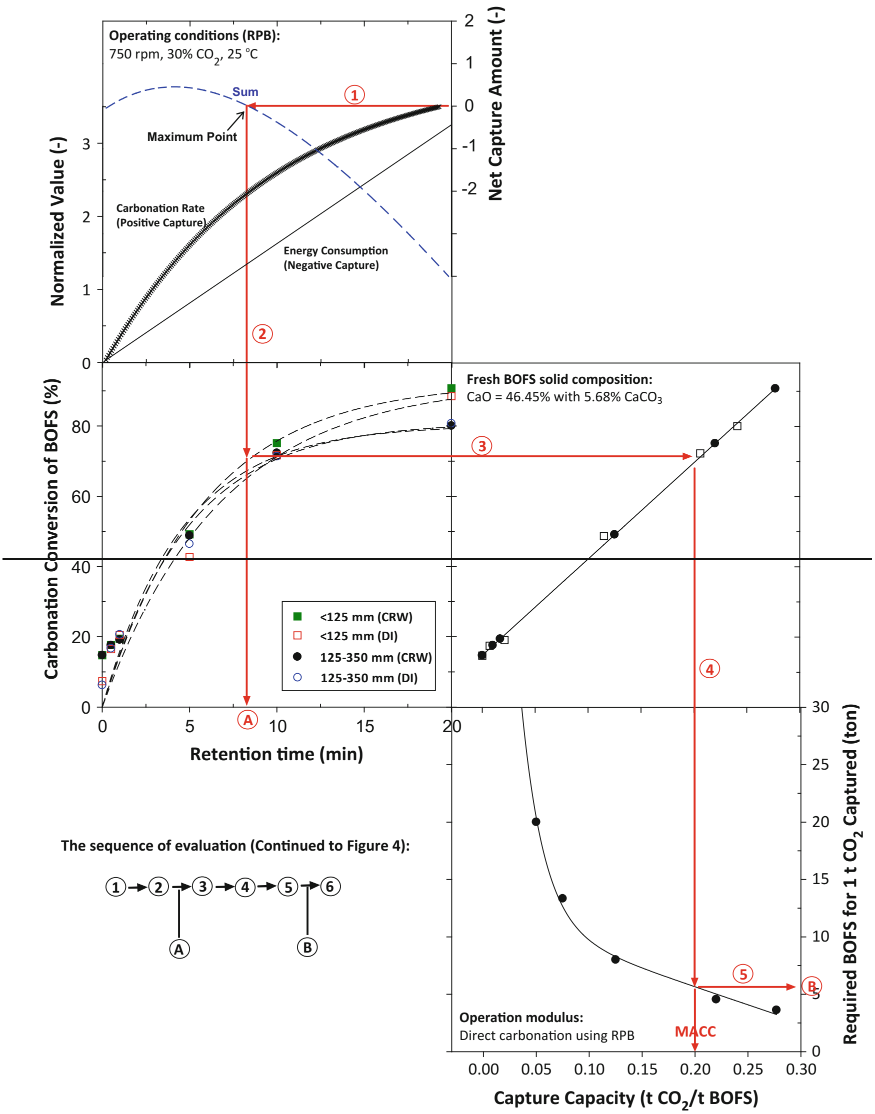

Fig. 19.3 Maximum achievable capture capacity (MACC) for steel slag in high-gravity carbonation, as indicated by *red line*. *CRMW* cold-rolling mill wastewater; *DI* deionized water. Reprinted with the permission from Ref. [4]. Copyright 2013 American Chemical Society

higher performance to achieve the lower energy consumption with higher CO_2 capture capacity.

Accordingly, the energy consumption of the carbonation process using direct carbonation of steel slag with various particle sizes under different reaction times can be determined from Fig. 19.4. As mentioned before, the capture capacity of

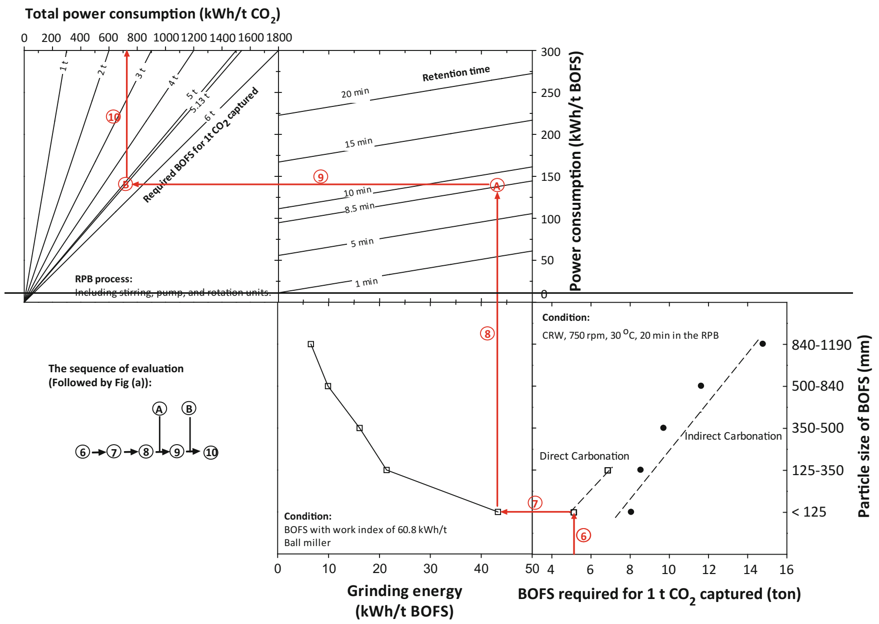

Fig. 19.4 Estimation of energy consumption for accelerated carbonation of steel slag coupled with alkaline wastewater in high-gravity carbonation, as indicated by *red line*. Reprinted with the permission from Ref. [4]. Copyright 2013 American Chemical Society

steel slag at its maximum reaction rate (operated for 8.5 min using alkaline wastewater) was estimated to be 0.195 ton CO_2 per ton steel slag (from Point 6 to Point 8, and then combined with the results of points A and B). The rotation of the packed bed is found to be the most energy-intensive process (i.e., 47.5% of total). Meanwhile, the fraction of energy consumption for the pumps and the grinding process to the total energy consumption is 18.7% and 24.3%, respectively. Although the carbonation process might require additional electricity, it could effectively neutralize the alkaline wastewater (down to a pH value of 6.3) and improve the properties of steel slag for further utilization, since the free CaO and $Ca(OH)_2$ in fresh steel slag could be totally eliminated after carbonation. The treatment cost for waste stabilization is expected to decrease because the carbonation process does not need to introduce additional chemicals or steam; it only needs to utilize waste CO_2 as a reaction agent.

19.2.2 Balancing Mass Transfer Rate and Energy Consumption

To optimize the mass transfer rate in the high-gravity carbonation process, a statistical K_Ga model should be developed according to the experimental data. The important factors including rotation speed (coded as A), gas flow rate (B), slurry flow rate (C), and L/S ratio (D) would affect the carbonation conversion of steelmaking slag in carbonation process. The analysis of the fitted response surface is generally equivalent to the analysis of the actual system if the fitted surface is a satisfactory estimation of the true response function. For instance, the developed model associated with rotation speed (A), gas flow rate (B), slurry flow rate (C), and L/S ratio (D) on overall gas-phase mass transfer coefficient (K_Ga) is presented in Eq. (19.14).

$$K_Ga\,(\text{coded}) = -0.13 + 1.32^*A - 0.71^*B - 0.16^*C - 0.14^*D + 2.04^*AB - 0.13^*A^2 + 0.11^*C^2 \tag{19.14}$$

In addition, a correlation between CO_2 removal efficiency (η) and K_Ga value can be expressed, based on the definition of K_Ga in two-film theory, as Eq. (19.15):

$$\eta(\%) = \eta_{\max}[1 - \exp(-t' \times K_Ga)] \tag{19.15}$$

where the $\eta_{\max}$ value represents the maximum CO_2 removal efficiency of the carbonation process, which has a theoretically maximum value of 100%. The exponential coefficient (t') is the characteristic time of carbonation.

In the case of high-gravity carbonation, the estimated t' value was 7.10 ± 0.45 (s), with a determination coefficient (r^2) of 0.955 [9]. It is noted that the average residence time of liquid flow in a high-gravity reactor should be approximately 0.2–0.8 s [10]. Although the carbonation reaction rate is generally fast enough compared to the retention time of gas in a high-gravity reactor, an appropriate level of gas-to-liquid (G/L) ratio should be maintained for the operation of high-gravity carbonation. It is noted that an increase in gas flow rate, corresponding to a greater G/L ratio, would reduce the retention time of gas in the packed bed zone.

To optimize the gas-phase mass transfer rate and energy consumption, the favorable operating modulus could be systematically determined via graphical presentation, as shown in Fig. 19.5. The energy consumptions could be expressed in terms of kWh per ton CO_2 capture by the process. In the case of the high-gravity carbonation process, a centrifugal acceleration should be maintained at 475 m/s^2 for a relatively lower energy consumption ($L1 \rightarrow L2$) and greater K_Ga value ($L3 \rightarrow L4$). The favorable G/L ratio should range between 40 and 55 for high-gravity carbonation (determined by both $L5 \rightarrow L6$ and $R1 \rightarrow R2 \rightarrow R3$). A further increase in G/L ratio up to 80 would lead to a low K_Ga value and high energy consumption for rotation and pumps, resulting in a poor CO_2 removal efficiency and capacity.

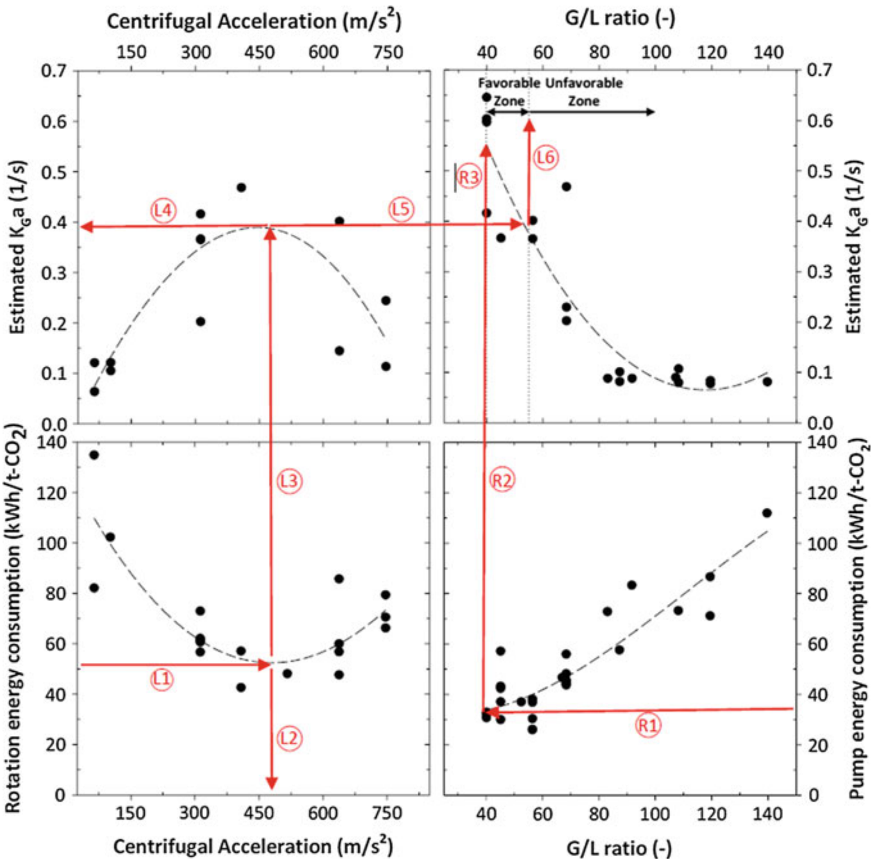

Fig. 19.5 Determination of optimal K_Ga value associated with favorable centrifugal acceleration (i.e., rotation speed) and G/L ratio via graphical presentation for high-gravity carbonation process (as indicated by *red line*). Reprinted by permission from Macmillan Publishers Ltd.: Ref. [9], copyright 2015

19.3 Comprehensive Performance Evaluation via 3E Triangle Model: A Case Study

Accelerated carbonation technologies are a feasible approach to integrating alkaline solid waste treatment with CO_2 fixation. In this section, a case study of the high-gravity carbonation (HiGCarb) process is assessed from the perspectives of engineering, environment, and economy (3E) using a 3E triangle model. The principles, key performance indicators, and data analyses and interpretation for the 3E triangle model are illustrated in Chap. 9. Several methods, such as response surface methodology, life cycle assessment and cost benefit analysis (as discussed in Chap. 9), can be utilized in the 3E triangle analysis for system optimization.

19.3.1 Scope and Scenario Setup

The performance of different carbonation processes should be critically evaluated from the perspectives of process design, energy consumption, and environmental benefits. Figure 19.6 shows the evaluation framework for the performance before (i.e., business-as-usual case) and after integration of HiGCarb process. To critically evaluate the benefits of integrating the HiGCarb process in the steelmaking industry, a systematically evaluation from the 3E aspects is quite important.

Table 19.1 presents a comparison of business-as-usual (BAU) and integration of the HiGCarb process in the steel industry from the 3E aspects. In the BAU case, three existing waste sources are separately operated or treated: (1) CO_2 emissions from hot-stove stack without CO_2 capture or fixation; (2) cold-rolling mill wastewater (CRMW) is neutralized by chemical agents at a wastewater treatment plant discharge; and (3) basic oxygen furnace slag (BOFS) stabilization and disposal at a landfill plant. In contrast, in the integration of HiGCarb process case, the CO_2 emitted from the steel industry is directly used to neutralize alkaline CRMW. At the same time, the contents of free-CaO and $Ca(OH)_2$ in BOFS can be

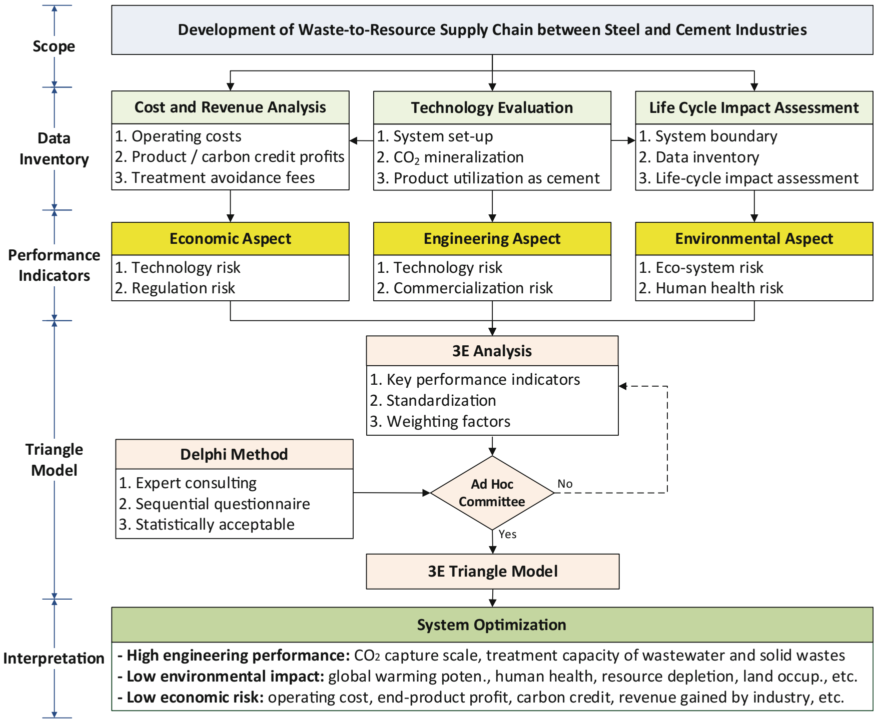

Fig. 19.6 Research framework of 3E assessment for high-gravity carbonation (HiGCarb) process using a triangle model to obtain an optimal operating modulus

Table 19.1 Remarks of business-as-usual (BAU) case and high-gravity carbonation (HiGCarb) process in steel industry

Aspects	System boundary	
	Business-as-usual (BAU)	Integration of HiGCarb process
Engineering	• No need to modify spec or features of process	• Manufacture of RPB reactor • Integration of RPB into industry • Additional energy consumption for HiGCarb process operation
Environmental	• CO_2 emission • CRMW discharge • BOFS landfill	• Reduction in CO_2 emission • Reduction in the use of chemicals for waste treatment • Increase in air pollutant emissions due to electricity use • Reduce cement production in cement industry (carbonated product utilization)
Economic	• Pay for BOFS treatment fee • Pay for CRMW treatment fee • Pay for CO_2 emission fee	• Capital and O&M costs for RPB reactor • Gain direct/indirect carbon credits • Gain profits from product sales • Avoidance of BOFS treatment fee • Avoidance of CRMW treatment fee

eliminated. As a result, the physico-chemical properties of BOFS can be utilized as cement replacement materials.

Furthermore, the carbonated product can be used as supplementary cementitious materials (SCM) in a blended cement mortar or concrete block. As cement manufacturing is an energy- and material-intensive process, with high annual production, significant indirect environmental benefits by the accelerated carbonation process can be realized.

19.3.2 Key Performance Indicators and Data Inventory

A 3E triangle model can provide a holistic assessment from the viewpoint of 3E aspects using a graphical presentation. As exemplified in Table 19.2, a total of 15 key performance indicators (KPIs) are presented for evaluating the HiGCarb process used in a 3E triangle model. Engineering performance (EP) is calculated using three technology indicators (engineering aspect), where EP_1, $EP_{2,}$ and EP_3 represent the HiGCarb capacities for CO_2 removal, wastewater neutralization, and carbonated product, respectively. To calculate the life cycle environmental impact (LCEI), eight environmental indicators are selected from ReCiPe midpoint and endpoint assessment because of their expected relevance for the HiGCarb process. The rest of impact categories in ReCiPe were excluded for the 3E analysis since they did not exhibit significant difference among the scenarios. Economic cost (EC) is determined using four economic indicators, including operating costs for capturing, end product sale profit, waste treatment free avoided, and carbon credit profit.

Table 19.2 Key performance indicators (KPIs) for the evaluation of accelerated carbonation process

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

[a] Functional unit = per ton of BOFS input for HiGCarb process. [b] *LCEI* life cycle environmental impact; *EP* engineering performance; *EC* economic cost

The weighting factors (W_i) of each KPI can be determined via Delphi method. The Delphi study is typically conducted over a period of few months and comprised at least two rounds. The participating experts, so-called ad hoc committee, could be consulted through roundtable discussion or sequential (online) questionnaires. The ad hoc committee should comprise highly informed academic and industrial experts from diverse backgrounds as well as government officials from the region.

Table 19.3 presents the examples of operating information and life cycle data inventory, including main material inputs and energy consumption, for nine scenarios. In this analysis, the CO_2 used for the HiGCarb process was introduced from the hot-stove gas at China Steel Corporation (Kaohsiung, Taiwan), where the average CO_2 concentration was 28–32%. Both the BOFS and alkaline CRMW were used directly from the manufacturing process. The 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 ranged between 75 and 170 kg CO_2 per day.

In this case study, data inventory is obtained from the experiment in field tests, and the scale factor is assumed to be 0.8. Due to different operating conditions, a total of nine scenarios are established based on various levels of CO_2 removal ratio:

- Low level: <70% (noted as L)
- Medium level: 70–90% (noted as M)
- High level: >90% (noted as H)

The choice of functional unit reveals several issues such as net CO_2 fixation amounts per unit weight of BOFS within a certain operating period. The 3E performance can be performed directly from the amounts of the materials and energy used such as electricity. In the 3E triangle model, the total scores for the 3E aspects are summed up after multiplying each KPI by its corresponding weighting factors (W_i). The W_i can be determined by the ad hoc committee using the Delphi method.

19.3.2.1 Engineering Consideration

The mass transfer rate of carbonation in the HiGCarb process could be significantly greater than that using a fix packed bed. As a result, the HiGCarb process can offer a high CO_2 capture efficiency of greater than 98% with a relatively short reaction time at ambient temperature and pressure [11]. The CO_2 removal ratio (η, %) from the flue gas via the carbonation process can be experimentally determined by Eq. (19.16):

$$\eta = \frac{\left(\rho_{\mathrm{CO_2},i}\, Q_{\mathrm{g,i}}\, C_{\mathrm{g,i}} - \rho_{\mathrm{CO_2,o}}\, Q_{\mathrm{g,o}}\, C_{\mathrm{g,o}}\right)}{\rho_{\mathrm{CO_2,i}}\, Q_{\mathrm{g,i}}\, C_{\mathrm{g,i}}} \times \% \tag{19.16}$$

Table 19.3 Data inventory including main material inputs and energy consumption for nine scenarios

ID	CO_2 removal ratio	Performance and operation conditions[a]				Inventory (per t-CO_2 captured)			Engineering performance (EP)		
		η (%)	ω (rpm)	Q_G (m^3/min)	Q_S (m^3/h)	CRMW inputs (t)[b]	BOFS inputs (t)	total energy (kWh)	EP_1 (t CO_2/t-BOFS)	EP_2 (m^3/t-BOFS)	EP_3 (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] η is CO_2 removal ratio as determined by Eq. (19.16), ω is rotating speed, Q_G is gas flow rate, Q_s is slurry flow rate, and L/S is liquid-to-solid ratio. [b] *CRMW* cold-rolling wastewater

where $\rho_{CO_{2,i}}$ and $\rho_{CO_{2,o}}$ (g/L) are the CO_2 mass densities at the temperature of inflow and outflow gas streams, respectively. $Q_{g,i}$ (L/min) and $Q_{g,o}$ (L/min) are the volumetric flow rate of the inlet and outlet gas streams, respectively. $C_{g,i}$ (%) and $C_{g,o}$ (%) are the CO_2 volume concentration in the inlet and exhaust gas, respectively.

The CO_2 capture process may involve energy-intensive units, such as stirring, heating, blowers, air compressors, pumps, liquid–solid separation, and material grinding. The power consumption of process could be directly determined by multiplying the operating voltage by the operating current of the existing equipment. Moreover, the energy consumption of material grinding (E_G) can be estimated by Bond equation [6], as shown in Eq. (19.17). It is noted that the Bond equation should give the most accurate estimation of grinding energy requirement within the conventional grinding range of 25,000 to 20 μm [12].

$$E_G = w_i\left(\frac{10}{\sqrt{D_{P80}}} - \frac{10}{\sqrt{D_{F80}}}\right) \tag{19.17}$$

where D_{F80} (μm) and D_{P80} (μm) are the 80% passing size of feed and product, respectively. The w_i (kWh/ton) is the work index of ground material, which expresses the resistance of the material to crushing and grinding. It is noted that the work index is subject to variations because of variations in the inherent properties of materials, variations in the grinding environment, and variations in the mechanism of energy transfer from the grinding equipment to its charge [13].

19.3.2.2 Environmental Consideration

Figure 19.7 shows the LCA system boundaries of the BAU case and the integration of the HiGCarb process into industry. The environmental impacts of the BAU (without the HiGCarb process) and the HiGCarb process are compared by means of LCA, including manufacturing and operation of the reactor and end product use as green materials. The functional equivalent (unit) is assumed to be one ton of fresh BOFS produced from steel industry or delivered to the carbonation process.

In the HiGCarb process, the unit operation processes include slag grinding, stirring machines, blowers, air compressors, pumps, RPB reactor, and electricity generation. Also, for the boundary system in the LCA, 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) are included. The life time of the system is assumed to be 20 years. As suggested by the LCA method described in the ISO 14040:2006 and ISO 14044:2006 [15, 16], the environmental impacts of the process could be quantified by Umberto 5.6 using the ReCiPe Midpont (E) and Endpoint (E, A) methodology [17].

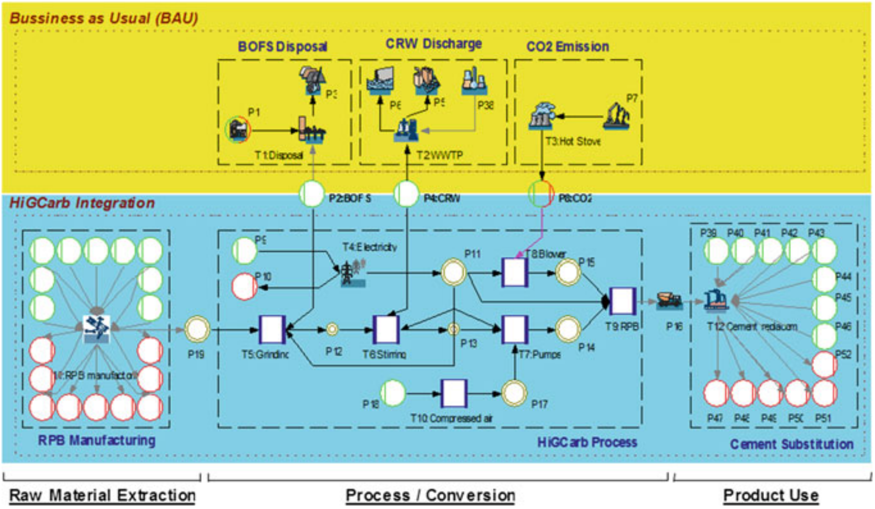

Fig. 19.7 System boundaries of business-as-usual (*top part* of the figure) and HiGCarb (*bottom*) process, which includes manufacturing of the reactor and substitution for cement. Reprinted by permission from Macmillan Publishers Ltd: Ref. [14], copyright 2016

19.3.2.3 Economic Consideration

The amount of revenue gained (RG), in terms of USD/t-BOFS, could be calculated by Eq. (19.18):

$$\mathrm{RG} = \left(P_{\mathrm{cc,dir}} + P_{\mathrm{cc,ind}}\right) + P_{\mathrm{ta}} + P_{\mathrm{ep}} - C_{\mathrm{op}} \tag{19.18}$$

where $P_{\mathrm{cc,dir}}$ is the profit of direct carbon credit by the HiGCarb process, $P_{\mathrm{cc,ind}}$ is the profit of indirect CO_2 avoidance credit by-product use, P_{ta} is the profit of BOFS treatment avoidance, P_{ep} is the profit of end product sale, and C_{op} is the operating cost.

The price of stabilized BOFS was approximately US$6.0/ton [18], which could be considered as the profit of carbonated BOFS product sales. Because the physico-chemical properties of BOFS can be upgraded after the HiGCarb process, no additional treatment of carbonated BOFS such as grinding and stabilizing processes within the steelmaking industry is required [4]. The treatment fee of BOFS was approximately US$10/ton [19], which can be saved in the case of HiGCarb process. In addition, the price of carbon credit in the emission reduction unit (ERU) market was approximately US$8.1/t-$CO_2$ in 2014 [20].

19.3.3 Performance in 3E Perspectives

19.3.3.1 Engineering Performance

Energy consumption is one of the major concerns to the engineering performance and cost effectiveness of a CO_2 capture process. It is noted that the accelerated carbonation by the HiGCarb is conducted at ambient temperature and pressure. Therefore, no additional energy is required to maintain the reaction temperature and pressure, in contrast to autoclave or slurry reactors [21–24]. Figure 19.8 shows the energy consumption of unit processes among the nine scenarios, in terms of a functional unit of "per t-CO_2". The results indicated that with a capture scale of 75–170 kg CO_2 per day, the total energy consumption of the HiGCarb process ranged from 205 to 440 kWh/t CO_2. The preprocessing of material such as feedstock grinding was the most energy-intensive process, contributing 65–79% of total energy consumption. Although the grinding process was energy-intensive, it was required for effective carbonation reaction, as well as for subsequent utilization of carbonated BOFS as SCM in Portland cement. In addition, the energy consumption of the rotating packed bed reactor was the second highest, corresponding to 11–21% of total energy consumption. Furthermore, scenarios with low CO_2 removal ratios (such as L1, L2, and L3) required a longer operating time to achieve the same CO_2 capture scale than other scenarios with high CO_2 removal ratios. Scenario H2 exhibited the lowest energy consumption of about 205 kWh for capturing one ton of CO_2. In this case, it met the criteria suggested by the US Department of Energy (US-DOE): A cost-effective CO_2 capture facility should achieve a CO_2 removal ratio of 90%, while maintaining less than 35% impact on the cost of electricity [25]. In other words, this criterion corresponds to a maximal energy consumption of 420 kWh/t-CO_2 [26].

Regarding the engineering performance, the specific capture capacity of BOFS (EP_1) mainly depends on the operating conditions, but not directly correlated with

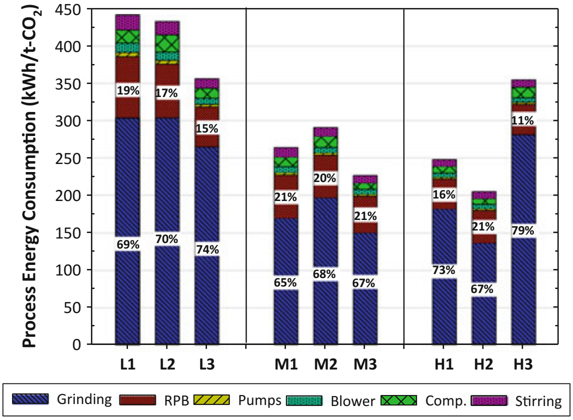

Fig. 19.8 Process energy consumption of HiGCarb for different scenarios, with their corresponding contribution in percentage. *Numbers* within each bar stack represent the contribution percentage

CO_2 removal ratio. Scenario H2 has the highest EP_1 value, corresponding to 316 kg CO_2 per ton BOFS. Among the nine scenarios, although CO_2 removal ratio in scenario H3 was the highest (i.e., 99.5%), the EP_1 of scenario H3 was the second lowest (i.e., 154 kg CO_2/t-BOFS). On the other hand, the CO_2 capture scale is mainly related to the amount of BOFS introduced into HiGCarb system per unit time period. In scenario H3, a relatively low L/S ratio (i.e., greater amount of BOFS) and higher slurry flow rate were used, the CO_2 capture scale of the entire HiGCarb system was promoted, i.e., 56.8 t-CO_2/year, becoming the top three high in the nine scenarios.

For the amount of wastewater neutralization (EP_2), scenario H3 exhibited the highest treatment capacity, followed by scenarios L2 and M2. The EP_2 was not related to CO_2 removal ratio but replied on the slurry flow rate and L/S ratio. Also, for the amount of carbonated product (EP_3), scenario 3 exhibited the highest capacity, followed by scenario H1 and then scenario L3. The carbonated product can be used as SCMs in blended cement mortar, where several mortar properties, such as early-stage compressive strength and soundness could be enhanced [27].

19.3.3.2 Environmental Performance: Impacts and Benefits

From the environmental aspect, the global warming potential (GWP), for instance, can be calculated by considering all the CO_2-equivalent emissions of each element or equipment from the life cycle point of view for the entire HiGCarb process. Figure 19.9 shows the GWP of each scenario as determined by LCA. The impact of the reactor manufacturing and its maintenance could be neglected since the magnitude of environmental impacts by RPB production is 10^4 to 10^5 times less than that of operating processes. The actual CO_2 capture amounts could be offset by the energy consumption due to the manufacturing and operation of equipment, causing additional CO_2 emissions. According to the direct measurement of CO_2 reduction in the flue gas, the capture capacity per ton of BOFS by carbonation reaction ranged between 140 and 320 kg CO_2. On the other hand, the carbonated BOFS product from the HiGCarb process can be used as SCMs in blended cement, thereby resulting in additional avoidance of CO_2 (i.e., indirect CO_2 reduction). Cement manufacturing is a CO_2-intensive process, where 0.73–0.99 tons of CO_2 would be generated for one ton of cement production [28]. To account for the environmental benefits from product utilization, the avoided burden approach [15, 16] has been applied in the LCA. The results indicated that a significant amount of 0.87–1.00 ton of CO_2 emission could be indirectly avoided by utilization of the carbonated BOFS as SCMs.

The HiGCarb process could be able to serve as a “real” CO_2 fixation technique from the viewpoint of LCA. As shown in Fig. 19.9, the highest GWP reduction was found in scenario H2, i.e., a reduction of roughly 1.28 ton CO_2-eq per t-BOFS between the BAU and HiGCarb. In scenario H2, additional CO_2 emissions from HiGCarb due to electricity uses (i.e., 33.8 kg CO_2-eq) were much lower than those of being directly captured by HiGCarb (i.e., 316.7 kg CO_2-eq).

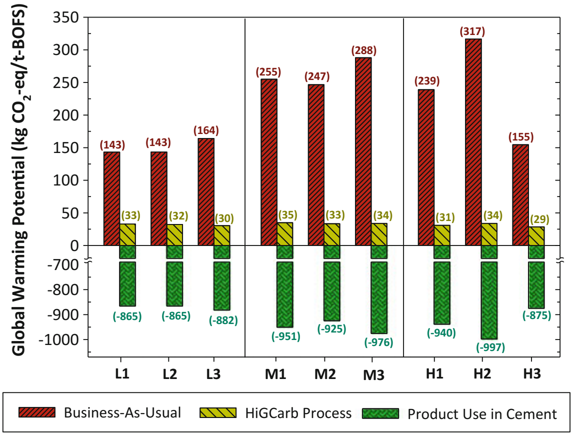

Fig. 19.9 Direct reduction by CO_2 capture and indirect CO_2 avoidance, in terms of global warming potential (GWP), in each scenario for one ton BOFS input to HiGCarb process

According to the LCA results, the HiGCarb process could reduce not only GHG emission but also environmental impacts on ecosystem quality, human health, and resource depletion. Figure 19.10 shows the results of the endpoint impact assessment for different scenarios by the ReCiPemethodology. Because of the various initial material flows such as wastewater discharge and CO_2 emission, the endpoint impacts of BAU (as indicated by the red bars) among the nine scenarios are quite different. For the HiGCarb (as indicated by the yellow bars), the particulate formation (PM) potential was found to be significantly higher than that of the BAU. It was attributed to the fact that the HiGCarb process would consume additional electricity, thereby resulting in a greater human health impact for all scenarios. However, the adverse impacts on human health due to the process of electricity usage could be compensated by the utilization of carbonated BOFS as SCM (as

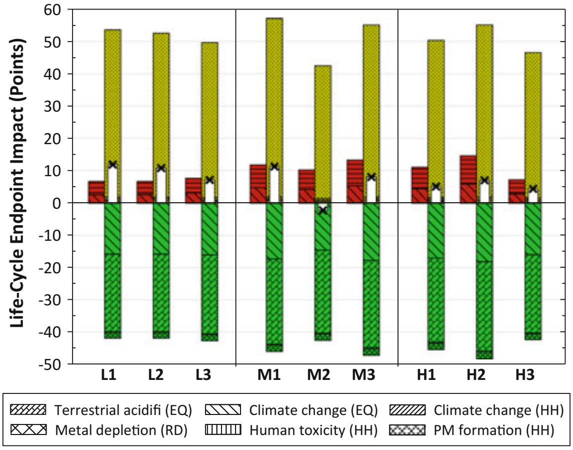

Fig. 19.10 Endpoint impact assessment on different scenarios. *Red bars* represent the BAU scenario; *yellow bars* represent the HiGCarb process; and *green bars* represent the product use as cement substitutes. *White bars with dots* represent the net impact between *yellow* and *green bars* in each scenario

presented in green bars). For this reason, the net endpoint impact (as presented in white bars) in the case of CO_2 removal ratio higher than 75% (scenario M2) could eventually be reduced by up to 12.4 points over that of BAU case. Nevertheless, the net endpoint impact was still relatively higher in scenarios of low CO_2 removal ratio (such as L1 and L2).

19.3.3.3 Economic Performance: Cost and Benefits Analysis

For the economic performance, Fig. 19.11 shows the effect of CO_2 removal ratio on operating costs and revenue gained in three different cases of electricity prices. Three different levels of average electricity price for industrial use in 2013 were used for economic performance evaluation: (1) Case A represents a low industrial electricity price of 0.091 USD/kWh; (2) Case B represents a medium industrial electricity price of 0.168 USD/kWh; and (3) Case C represents a high industrial electricity price of 0.319 USD/kWh. As shown in Fig. 19.11, the operating cost for processing one ton of BOFS was roughly 5.4–5.9 USD in Case A, while increasing to 19.0–20.8 USD per one ton of BOFS input in Case C. On the other hand, the profits from direct and indirect carbon credits were estimated to be 8.3–10.1 USD per ton of BOFS input to the HiGCarb process. Also, no additional CO_2 storage cost is needed for the HiGCarb because the CO_2-based mineral product can be directly used as SCMs for Portland cement in cement industry. As a result, the total profits returned including carbon credit and BOFS-related returns were approximately 25.8–29.0 USD per ton of BOFS input to HiGCarb. According to the above analysis, the highest revenue was gained with a CO_2 removal ratio greater than 93%. In Case A, the revenue gained was estimated to be 20.2–23.2 USD per ton of BOFS input.

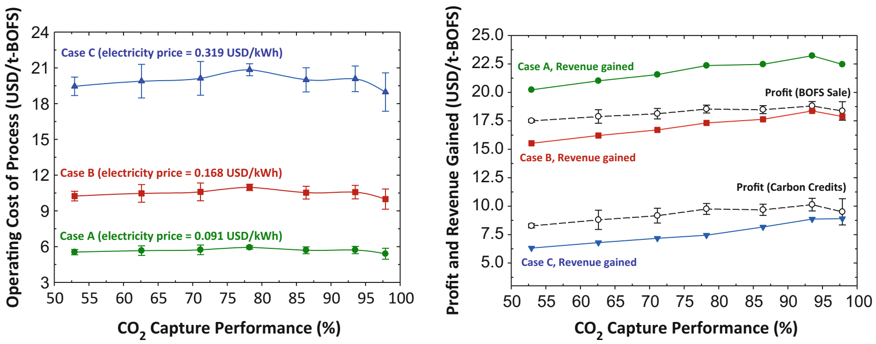

Fig. 19.11 Effect of CO_2 removal ratio on operating costs and revenue gained. *Case A* represents the one with low electricity price; *Case B* represents the one with medium electricity price; and *Case C* represents the one with high electricity price. *Note* Revenue gained is calculated by Eq. (19.18)

19.3.4 Optimization Using 3E Triangle Model

Figure 19.12 shows the results of performance evaluation for different scenarios of HiGCarb process via 3E triangle model. The results indicated that the effects of CO_2 removal ratio (η) on economic costs were not significant since the LCC scores typically ranged between 0.25 and 0.40. However, a poor engineering performance (i.e., scenarios L1, L2, and L3) is typically accompanied by severe environment impacts, with the LCEI scores ranging between 0.52 and 0.60. In other words, an increase in CO_2 removal ratio should effectively reduce the environmental impacts and make integration of the HiGCarb process into the steel industry more environmentally friendly. Among nine scenarios, scenario H2 exhibits a superior engineering performance (as indicated by line 1) with a relatively lower environmental impacts (as indicated by line 2) and a relatively lower economic costs (as indicated by line 3). Although the CO_2 removal ratio of scenario H3 was the highest (i.e., 99%) among all scenarios, the large quantity of BOFS input eventually resulted in medium environmental impacts and economic costs.

To evaluate the significance of HiGCarb process in an industry, data from China Steel Corp (CSC) is used and combined with the results of 3E triangle model. The annual production of BOFS in CSC is assumed to be 1.2 Mt [29], which should be treated and/or utilized. By applying the scenario H2, the annual direct CO_2 fixation by HiGCarb process is estimated to be 0.33 Mt, corresponding to a reduction potential of 1.5% in total CO_2 emission from the studied industry. Meanwhile,

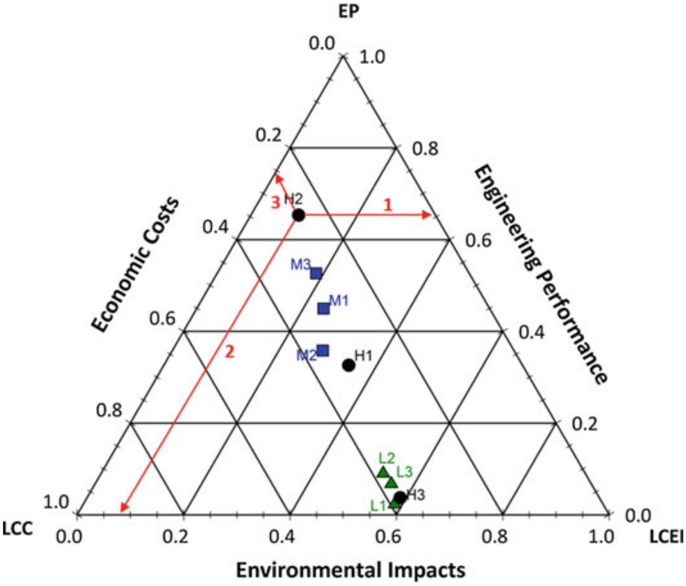

Fig. 19.12 Comprehensive performance evaluation of HiGCarb process for different scenarios via 3E triangle model

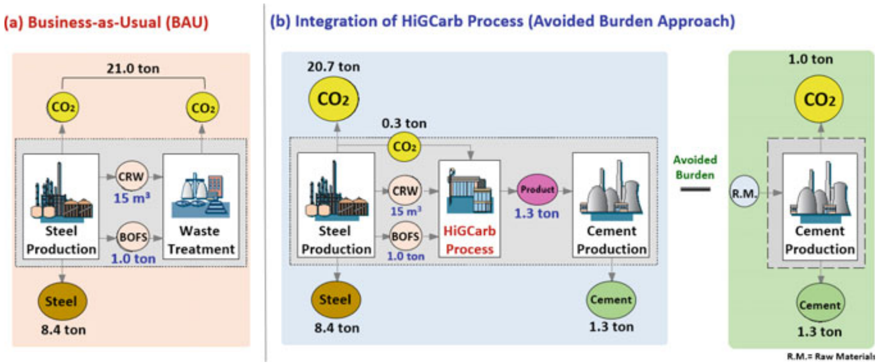

Fig. 19.13 Comparison of performance for business-as-usual and integrated HiGCarb process suggested by the results from 3E triangle model

based on the avoided burden approach, the annual indirect CO_2 reduction from the substitution of carbonated BOFS as cementitious materials is about 1.05 Mt, accounting for ~5% in total CO_2 emission from the steel industry.

As a result, a reduction potential of up to 6.5% in total CO_2 emission from the steel industry could be realized, as shown in Fig. 19.13. The HiGCarb process can establish a waste-to-resource supply chain between the steel and cement industries, thereby reducing the overall CO_2 emissions and resource consumption. At the same time, the alkaline wastes from steel industry could be stabilized and converted into valuable products such as green cement.

19.4 Technology Demonstration and Commercialization

Large-scale CO_2 separation from flue gases in power and/or industrial plants would make a huge volume of CO_2 available on-site. As a result, the subsequent fate of captured CO_2 would be either storage in natural geological structures or direct utilization and conversion. In practice, the suitable storage sites are few in number, and the procedures still involve high energy and economic costs with high risk and uncertainty in terms of long-term storage. Therefore, carbon utilization and conversion technologies are preferred, which should be directly integrated with the associated capture unit. Rather than the amount of CO_2 used, the most important consideration in CO_2 utilization is the development of innovative technologies for cleaner production, thereby directly leading to a reduction in the use of energy and materials.

Industrial wastes (by-products) are generally produced near places of CO_2 emissions. Accelerated carbonation using industrial alkaline wastes is attractive since an integrated approach to combining CO_2 capture and utilization with waste stabilization can be achieved. The potential environmental impacts caused by utilization of those untreated wastes, such as (1) highly alkaline and active properties and (2) heavy metal leaching, could be avoided. Therefore, using emitted CO_2 to carbonate industrial waste (i.e., accelerated carbonation) offers an improvement over existing methods because it does not require the CO_2 or the industrial waste to be transported. It also allows better monitoring of total pollution emissions.

19.4.1 Worldwide Demonstration Plans

Accelerated carbonation may involve several energy-intensive processes, such as material grinding, reactor heating, and gas pressurization. Energy and cost penalties largely depend on plant scale, operation conditions, and operation modulus (such as pretreatment and post-treatment processes). Several pilot studies and demonstration projects of accelerated carbonation using alkaline wastes can be found around the world, as summarized in Table 19.4. For example, since 2007, an accelerated carbonation plant at the Rocks (Wyoming, the USA) has been demonstrated in a 2120-MW coal-fired power plant using fly ash [30]. Another pilot study in the USA has been developed by Calera Corp. The Calera technology can fix approximately 30,000 t CO_2 per year from fossil fuel power plants and other industrial sources. The captured CO_2 is sequestered in geologically stable substances suitable for disposal, storage, and/or use as building materials. In 2009, Calera identified another ideal demonstration site at a brown-coal power plant in the Latrobe Valley, Victoria, Australia, for further demonstration [31].

In France, the Carmex project was initiated in 2007 and launched in 2009 to utilize various materials (such as harzburgite, wehrlite, iherzolite, olivine, and slags) through direct carbonation with and/or without organic ligand and mechanical exfoliation. The Carmex experiences indicate that the use of mineral carbonation is feasible for industries [34]. In this project, the accessible alkaline wastes are matched to large CO_2 emitters through a dedicated geographic information system (GIS). From the technical point of view, a high carbonation conversion of 70–90% can be achieved without additional heat activation of feedstock.

In Australia, the MCi project has been carried out to transform CO_2 into carbonates for use in building or non-fired products, such as bricks, pavers, and plasterboard replacements [35]. A total investment of US$9 million over four years was provided to establish the pilot plant at the University of Newcastle. The serpentine was used as the feedstock to mineralize CO_2 from the Kooragang Island plant.

In San Antonio (Texas, the USA), the Capitol SkyMine® plant was under construction by Skyonic on September 2013, and had been launched since October 2014. This plant can directly remove CO_2 at a scale of 83,000 t-CO_2 annually from industrial waste streams. The carbonate and/or bicarbonate material products can be

Table 19.4 Pilot-scale demonstration projects of accelerated carbonation around the world

Project name	Scale	Launch	Feedstock	Description of features	Performance	References
Rocks, Wyoming, the U.S.	Pilot scale	2007	Coal fly ash	• Conducted at Jim Bridger Power Plant • Direct mineralization of CO_2, SO_2, and Hg in flue gas	• CO_2 removal efficiency: 90% • SO_2 removal efficiency: > 85% • $HgCO_3$ formation: 0.4 mg/kg (25–58 °C)	[30, 32]
Calera (Moss Landing, CA, the U.S.)	Pilot scale	2007	Naturally occurring brines, waste materials	• Stabilize a number of pollutants (e.g., mercury, SO_2) in addition to CO_2 • Potentially low energy penalty compared to other carbon capture processes • Produces calcareous material and HCl	• CO_2 capture capacity: 30,000 t/y • CO_2 capture efficiency: > 50% • SO_2 removal efficiency: > 95% • Replace 40% of clinker in cement production	[31, 33]
Carmex (New Caledonia, France)	Pilot scale	2009	Mafic wastes (mine tailings)	• Ex situ mineral carbonation without heat activation • Matching accessible wastes to large CO_2 emitters through dedicated GIS • Environmental assessment through LCA	• Carbonation yield: 70–90% • 23% of initial Mg was carbonated in Ni-pyrometallurgical slag	[34]
MCi (New South Wales, Australia)	Pilot scale	2013	Serpentine	• Capture CO_2 emissions at its Kooragang Island plant • Solid product can be turned into various green products including building materials (bricks and pavers)	• Total investment of US$9 million over four years • Bricks were used as construction materials, acting as physical carbon sinks	[35]

(continued)

Table 19.4 (continued)

Project name	Scale	Launch	Feedstock	Description of features	Performance	References
HiGCarb process (CSC, Kaohsiung, Taiwan)	Small scale	2014	Basic oxygen furnace slag, cold-rolling mill wastewater	• High-gravity carbonation process • Alkaline wastewater was introduced • Carbonated BOFS was utilized as partial replacement of cement	• CO_2 capture capacity: ∼ 60 t/y • CO_2 removal efficiency: > 95% • Wastewater is neutralized to pH 7 • Free CaO and $Ca(OH)_2$ in BOFS are eliminated	[11, 27]
Capitol SkyMine (San Antonio, TX, the USA)	Business model	2014	Brine solution	• Groundbreaking on September 2013 • Facility opened on October 2014 • Produce green chemicals such as HCl, bleach, Cl, and H_2	• CO_2 capture capacity: 83,000 t/y • CO_2 removal anywhere from 15%–99% • Remove CO_2, SO_x, and NO_2 from flue gas • Removes heavy metals (e.g., Hg)	[36]
HiGCarb process (FPCC, Yunlin, Taiwan)	Small scale	2016	By-product lime	• High-gravity carbonation process • Carbonated product is used as SCM	• CO_2 capture capacity: ∼ 200 t/y • CO_2 removal efficiency: > 95% • Remove CO_2, SO_x, NO_x, and PM from flue gas	–

cogenerated for use in bioalgae applications to become a profitable process. Aside from mineralizing CO_2, the process can remove SO_x, NO_2, and heavy metals such as mercury from existing power plants and/or industrial plants that can be retrofitted with SkyMine®.

In Taiwan, the first small-scale high-gravity carbonation (HiGCarb) process was launched at China Steel Corporation (CSC) in 2013 to stabilize basic oxygen furnace slag (BOFS) and alkaline wastewater. The CO_2 removal efficiency of hot-stove gas was greater than 95%, with total elimination of CaO_f and $Ca(OH)_2$ content in the BOFS. The annual capture scale was ~60 tons CO_2 at a gas inflow rate of 0.9 m^3/min. Moreover, the carbonated BOFS was used as green cement substitutes in mortar. In 2016, the second HiGCarb process was established at Formosa Petrochemical Corporation (FPCC) using by-product lime to capture CO_2 in the flue gas. The capture scale of this plant was about 0.6 tons CO_2 per day, at a gas inflow rate of 1.8 m^3/min. Moreover, the carbonated by-product could be used as supplementary cementitious materials (SCM) in cement mortar or concrete.

19.4.2 Engineering Performance

19.4.2.1 CO_2 Capture Scale and Efficiency

Scale-up of the post-combustion CO_2 capture process is possible without significant developments or costs [37]. However, deployment of the post-combustion carbon capture process in industries is still challenging because the CO_2 emissions normally come from “multiple” sources. For example, in the steel industry, an integrated steelmaking process is composed of numerous facilities from the entire life cycle of iron ore to steel products including raw material preparation (such as coke production, ore agglomerating plant, and lime production), iron-making (such as blast furnace, hot metal desulphurization), steelmaking (such as basic oxygen furnace, ladle metallurgy), casting and finishing mills. The largest part of direct CO_2 emissions in steel mills is from power plants (about 48% in total CO_2 emissions), followed by blast furnaces at around 30% [38].

In large-scale tests, process integration should be considered to improve the performance of the entire industrial plant, thereby reducing the energy requirement for the capture process. Also, the selection of appropriate site for CO_2 capture should be referred to the management of the use of by-product gases, as well as on the definition of boundary limits.

19.4.2.2 Product Utilization

Accelerated carbonation technology of alkaline wastes could be moved toward commercialization, only if the produced carbonates can be used as a valuable product such as a substitute for components of cement [37]. In addition to the product utilization, the current challenges in the application of the process still include the following:

- Effect of impurities on removal performance
- Acceptance of the product by the cement industry
- Ability to capture large amounts of CO_2
- Energy requirements
- Finding an appropriate water source
- Production of alkalinity
- Having sufficient demand for the end product

For the product utilization, it was reported that both cement kiln dust (CKD) and fly ash (FA) have been successfully used to produce a green Portland ash [31, 39]. The suitability of the calcareous material as a partial replacement for cement clinker in cement has been documented in some non-structural applications in the USA, but the suitability of the calcareous material as a cement ingredient in concrete applications has not yet been demonstrated publicly [31].

19.4.2.3 Integrated Approach to CO_2 Fixation and Solid Waste Utilization

Figure 19.14 shows an integrated approach to deploying the high-gravity carbonation (HiGCarb) process, which could be considered for CO_2 fixation in flue gas and solid waste utilization within an industrial plant. It is noted that the CO_2 removal rate by the HiGCarb process could meet the timescale in industrial plants.

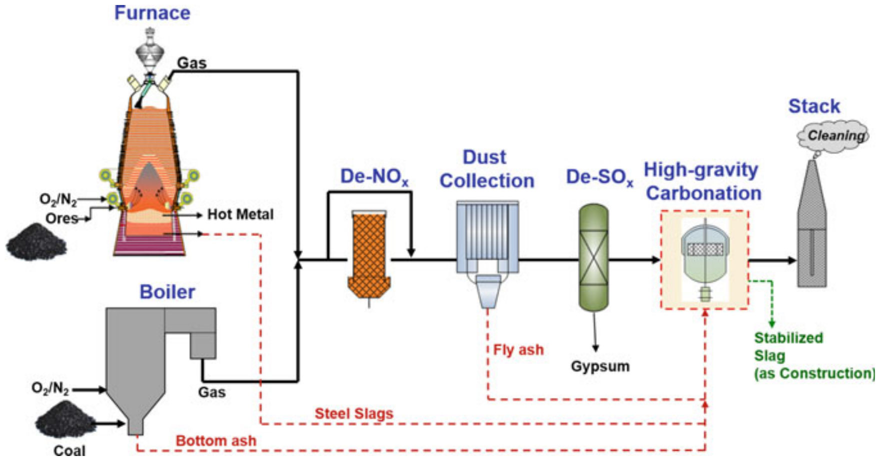

Fig. 19.14 Integrated approach to applying high-gravity carbonation (HiGCarb) process for CO_2 capture in flue gas and solid waste utilization within an industrial plant

19.4.3 Economic Perspectives

19.4.3.1 Energy Consumption

Accelerated carbonation might involve several energy-intensive processes, such as grinding of solid wastes, reactor heating, and gas pressurization. From the perspective of energy consumption, the extensive uses of electricity for any unit process in carbonation could easily diminish the credits from carbon fixation. A life cycle assessment for different types of direct carbonation processes indicates that energy consumption is responsible for the increase in additional CO_2 emission and offsets the overall CO_2 capture efficiency Xiao et al. [40]. In comparison, although the energy consumption of indirect carbonation is typically less than that of direct carbonation, the manufacturing of chemicals for the extraction step may generate additional CO_2 emission and lead to other environmental issues. This suggests that the recovery of the extractants (valuable chemicals) with low energy consumption should be included for implementing indirect carbonation [40, 41].

As suggested by the US Department of Energy (DOE) [25], a cost-effective CO_2 capture facility should meet the following criteria:

- Achieve a removal efficiency (η) of 90%
- Maintain <35% impact on the cost of electricity (COE)

Consequently, heat recovery is an important unit process for accelerated carbonation (exothermic reaction), which could not only improve carbonation performance but also reduce energy loss. For example, the temperature of flue gas streams is high enough for carbonation since it is usually above the dew point. Therefore, heat can be directly obtained from the gas streams or other heat-regenerating systems. To achieve this goal, Santos et al. [42] developed an integrated process where the high pressure is obtained by pumping liquid in an autoclave reactor through a long reaction chamber. Moreover, the heat generated by the carbonation reaction (i.e., exothermal) can be recovered via the integrated process.

Pan et al. [4, 43] introduced a high-gravity carbonation (HiGCarb) process for direct carbonation, where high micromixing between the slurry and gas phases could enhance the overall mass transfer, thereby improving the carbonation conversion and reducing the residence time. In the HiGCarb process, the slurry was first pumped into the center of the reactor, after which it flowed outward motivated by centrifugation. In the meantime, the CO_2 gas entered the reactor from the tangent direction and moved inward due to the pressure gradient.

Figure 19.15 shows the effect of CO_2 removal efficiency on energy consumption and CO_2 capture capacity of the HiGCarb process. Both steel slag grinding and HiGCarb (air compressors, stirring machines, blowers, pumps, and rotating packed bed reactor) processes were considered in energy consumption calculation. The scale of the HiGCarb process was operated at a capture capacity of ~170 kg CO_2 per day, producing ~690 kg of C-BOFS per day. The energy consumption for the

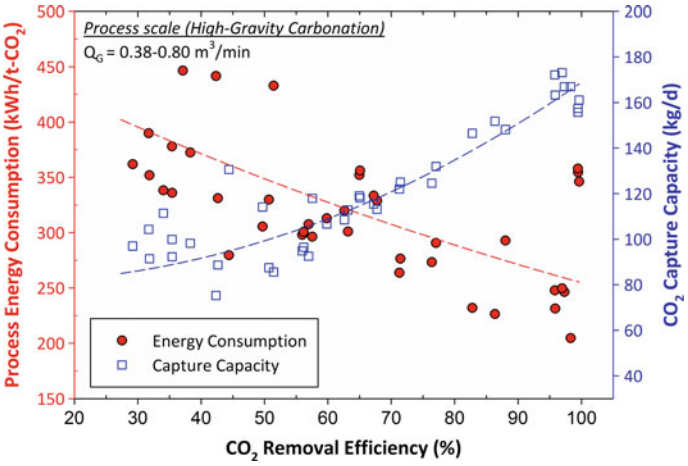

Fig. 19.15 Influence of CO_2 removal efficiency (η) on energy consumption and capture capacity in the case of high-gravity carbonation (HiGCarb) process. Adaption with the permission from Ref. [27]. Copyright 2015 American Chemical Society

high-gravity carbonation include grinding, pumps, blowers, stirring machines, air compressors, and rotation packed bed. The results indicate that the overall energy consumption of the HiGCarb process increases with the decrease in CO_2 capture efficiency. The total energy consumption of the HiGCarb process with CO_2 removal efficiency (η) > 90% was estimated to be 270 ± 60 kWh/t-CO_2 captured (with a 95% confidence interval). It is noted that this value was lower than the DOE requirement, i.e., 420 kWh/t-CO_2 [26]. In addition, the corresponding capture capacity was estimated at 150 kg of CO_2 per day. In this case, the COE of the HiGCarb process was estimated to be 22.4 ± 0.1%, which met the goal of maintaining <35% impact on COE set by the US DOE [25].

19.4.3.2 Operating Cost

Accelerated carbonation using industrial alkaline wastes should be an important part to the reduction in CO_2 from the industrial sector and the use of industrial wastes as cement replacement. Technology may not be the only barrier to the deployment of accelerated carbonation process in the industrial sector. Market competitiveness and the global nature of some of these industries are important issues that should be addressed. From an economic perspective, costs for grinding feedstock can be reduced using the slag in the as-received condition. If the grinding process is needed to improve efficiency of carbonation, it should be done only to the optimum particle size for carbonation to save energy costs. Similarly, the costs of CO_2 pressure can be reduced by using it at atmospheric pressure, and the costs of transportation can be reduced by equipping the slag-producing industry with the carbonation process.

Energy and cost penalties largely depend on plant scale, operation conditions, and operation modulus including pretreatment (e.g., grinding and thermal activation) and post-treatment processes (e.g., product separation and disposal) [44, 45]. Due to lack of commercialized plant studies, cost estimations of accelerated carbonation are based roughly on pilot- or laboratory-scale operations. As presented in Table 19.5, the energy consumption and cost evaluation of direct carbonation were relatively lower than those of indirect carbonation. In the case of direct carbonation, the energy requirement of the grinding process was the major cost in the overall process [4, 46]. Depending on the types of feedstock and operating modulus, the cost of *ex situ* direct carbonation typically ranged between US\$54/t-CO_2 and US \$133/t-CO_2. The handling of solid particles (powders) in the process has the potential to raise the operation and maintenance (O&M) costs, compared to CO_2 absorption technologies using ammonia and amine [47].

In contrast, for indirect carbonation using chemical extraction (such as CH_3COOH, HCl, HNO_3, and NaOH) without regeneration of chemicals, a fairly high cost of US\$600–4500 would be required for capturing one ton of CO_2 [52]. However, with the regeneration of the chemicals, the recovery process would generate more than 2.5 times the amount of CO_2 fixation in the carbonation process [53]. The operating costs depend largely on the purity of the precipitated calcium carbonate (PCC) product. An average cost of US\$80 is required per ton of the PCC production from two-stage indirect carbonation using cement wastes at 50 °C and 30 bar. The major energy consumption processes include pulverization, carbonation, CO_2 separation, CO_2 pressurization, and stirring process for both extraction and carbonation, which are considered at a total of 52.8 MW [54].

To make *ex situ* carbonation more economically feasible, a breakthrough on the use of carbonated solid wastes or products should be sought in the aspects of technology, regulation, institution, and finance. The global cement market is large: with ~3.5 billion metric tons used in 2011 at the processing cost of ~US\$100 per ton [37]. It is noted that the carbonated solid waste could potentially be used as partial cement replacement materials (i.e., SCM) [55, 56]. As a result, the benefits returned from carbonation product utilization should be considered in the fiscal analysis of the overall process. From the viewpoint of energy consumption, fine fly ash (FA) should be a good candidate for low-cost carbonation since no grinding process is needed in advance. Moreover, waste heat from manufacturing processes could be integrated instead of electrical heating to reduce the overall energy requirement and operating cost [57].

In comparison, without taking into account long-term monitoring costs, it is estimated that the total cost of *in situ* carbonation should be at US\$72–129 per t-CO_2, if transportation and storage cost was assumed to be ~US\$17 per t-CO_2 in basaltic rocks [58]. All of these costs are by far greater than the recent carbon price in European carbon market, i.e., ~US\$7 per t-CO_2 in 2014 [52]. However, it is noted that the CO_2 price may increase to US\$35–90 per t-CO_2 by 2040 [59]. In

Table 19.5 Energy consumption and cost evaluation of various processes for direct carbonation

Process descriptions	Evaluation consideration	Feedstock	Performance	Energy consumption (kWh/t-CO_2)	Cost[b] (US$/ t-$CO_2$)	References
Carbonation with pretreatment	• Mechanical (grinding to 38–75 μm) and thermal (630 °C for 2 h) pretreatment • Carbonation reaction	Olivine	• Mineral grade: 70%	633–653	54–55	[46]
		Wollastonite	• Mineral grade: 100% (lizardite/antigorite)	429	91	
Carbonation with thermal heat	• Heat activation pretreatment • Partial dehydroxylation of lizarite	Serpentine	• Heat integration • 63% of decrease in energy requirement	n.a.[a]	70	[48]
Carbonation in a fluidized bed	• Pilot-scale plant (at a 532 MW coal-fired power plant) • 70 °C for 120 min	Fly ash (No grinding was required)	• Efficiency: 90% • Capacity: 0.21 t-CO_2/t-ash	n.a.	7.4–27.3	[32, 49]
Carbonation in a spraying glass chamber	• Laboratory scale • Grinding: ~ 45 μm	Mixed steel slag and concrete wastes	• Efficiency: > 50% • Operating (pumps, reactor, sprayers) and labor costs • Cost of water consumption included	15.6 (laboratory scale)	8	[50]
Autoclave reactor (20 bar and 200 °C)	• Depreciation of investments • Viable costs (feedstock, electricity, cooling water) • Fixed costs (labor, supervision, maintenance, insurance, and laboratory)	Wollastonite	• Feedstock costs: US$70 • Grinding (from 100 mm to < 38 μm) • Compression costs: US$34	296 (power) -295 (Heat) (laboratory scale)	133	[51]
		Steel slag	• Fixed operating cost: US$36 • High depreciation cost: US$30	337 (power) -167 (Heat) (laboratory scale)	100	
High-gravity carbonation (HiGCarb)	• Grinding of slag • Stirring, Pumps, Carbonation Reactor (1 atm, 25 °C) • Scale factor: 0.7	Basic oxygen furnace slag	• Grinding costs: US$17.7 • Operating costs: US$24.3	485 (laboratory scale)	57	[4]

[a] *n.a.* Not Available. [b] Assumed EUR€1.0 = US$1.3

another scenario estimated by the International Panel on Climate Change (IPCC), the carbon price would give ~US\$55 per t-CO_2 as a lower bound estimate.

19.4.4 Environmental Impacts and Benefits

The effect of accelerated carbonation should be carefully weighed and compared according to changes in the environmental impacts. A life cycle assessment (LCA) of the accelerated carbonation process is of particular importance to maximize CO_2 capture capacity while minimizing additional CO_2 emissions due to the process energy consumption. Several operation units, such as material grinding, sieving, and heating, for accelerated carbonation are energy-intensive processes, thereby leading to additional CO_2 emissions. In particular, more than half of the process power consumption may come from the material grinding [8]. These unit processes may also increase other environmental impacts, such as eutrophication (midpoint), acidification (midpoint), and resource depletion (endpoint), due to increases in the concentrations of other pollutants.

19.4.4.1 Reduction in Greenhouse Gas Emission

Deployment of accelerated carbonation in industries and/or power plants will contribute to greenhouse gas (GHG) emission reduction and create additional environmental benefits. For instance, conventionally, precipitated calcium carbonate (PCC) is manufactured by carbonating calcined limestone; therefore, the produced CO_2 is greater than that bound during the carbonation process [60]. Traditional PCC manufacturing resulted in an additional 0.21 kg of CO_2 emissions per kg of PCC [61], mainly caused by oil combustion for lime calcinations. In the indirect carbonation process, the PCC can be produced from a carbon-free feedstock, being a more environmentally sustainable method for producing PCC since no calcination step is required. In this case using acetic acid with wollastonite, a net fixation of 0.34 kg CO_2 per kg of PCC can be achieved, indicating a substantial reduction in GHG emission. Since PCC can be used in the paper industry, a paper mill plant integrated with the indirect carbonation process can transform its CO_2 emissions into PCC toward carbon neutrality.

Similarly, the direct carbonation process can attain huge environmental benefits by taking the use of carbonated solid wastes as SCMs into account. The demand of cement could be reduced if the carbonated solid waste is used as substitutes to replace Portland cement in cement mortar or concrete. Cement production is energy- and material-intensive, which accounts for 4–5% annual CO_2 emission around the world [62]. China accounts for more than 60% of global cement production, where the carbon footprint of cement production in China in 2011 was 0.545 ton-CO_2/ton-cement [63]. The main contributors to CO_2 emission from cement production are

- Clinker process: The production of raw cement material lime (CaO) from limestone ($CaCO_3$), which produces ~0.5 ton CO_2 per ton cement [62].
- Intensive heat required during the cement production.

In addition, a huge amount of natural resources is used for cement production. For instance, Kumar et al. [64] estimated that 1.3–1.5 tons of limestone, 0.2–0.4 tons of clay, and 0.11–0.13 tons of coal are used per ton of cement clinker production.

19.4.4.2 Leaching Behavior of Heavy Metals from Alkaline Solid Wastes

The leaching potential of heavy metals from alkaline solid wastes is of great concern, in terms of human health and environmental impact. Extensive studies have been carried out to evaluate the effect of pH of the solution, carbonation reaction, and mineral structure on the leaching behavior of heavy metals from the solid wastes [55, 65, 66]. Also, different oxidant states of heavy metals in alkaline solid wastes would result in different leaching behaviors, especially for Cr species [67]. It has been demonstrated that accelerated carbonation could effectively reduce the leaching behaviors of most heavy metals from various types of solid wastes, such as steel slag [27], bottom ash [68], and fly ash [69].

As indicated in a few reports [65], the dissolution of calcium silicate minerals (e.g., C_2S) might break down the mineral structure during carbonation, as shown in Eq. (19.19), thereby potentially releasing heavy metals (e.g., vanadium), chlorine and fluoride ions into solution.

$$(\mathrm{CaO})_2\cdot\mathrm{SiO_2} + m\mathrm{H_2O} \rightarrow \mathrm{CaO}\cdot\mathrm{SiO_2}\cdot\mathrm{H_2O}_{m-1} + \mathrm{Ca(OH)_2} \tag{19.19}$$

Calcium-bearing components are the major alkalinity contributors in the alkaline solid wastes. For instance, the pH of the solution containing steel slag typically ranges between 11 and 13. Conversion of the CaO species in solid wastes into $CaCO_3$ with CO_2 gas could effectively decrease the pH of the solution to about 6–8. In parallel with the decrease in pH, the leaching potential of most heavy metals from wastes, such as Pb, Cr, Cu, Zn, Cd, and V ions, could be properly restricted due to the formation of insoluble carbonates [65, 66].

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