



Performance evaluation for carbonation of steel-making slags in a slurry reactor

E.-E. Chang^a, Chung-Hua Chen^b, Yi-Hung Chen^c, Shu-Yuan Pan^b, Pen-Chi Chiang^{b,*}

^a Department of Biochemistry, Taipei Medical University, Taipei, Taiwan

^b Graduate Institute of Environmental Engineering, National Taiwan University, No. 71 Chou-shan Rd., Taipei 106, Taiwan

^c Department of Chemical Engineering and Biotechnology, National Taipei University of Technology, Taiwan

ARTICLE INFO

Article history:

Received 17 February 2010

Received in revised form 10 August 2010

Accepted 9 November 2010

Available online 18 November 2010

Keywords:

Mineral sequestration of CO₂

Aqueous carbonation

Basic oxygen furnace slag

Calcite

Shrinking-core model

Kinetics

ABSTRACT

CO₂ sequestration by the aqueous carbonation of steel-making slag under various operational conditions was investigated in this study. The effects of the operational conditions, including type of steel-making slag, reaction time, reaction temperature, and CO₂ flow rate, on the performance of the carbonation process were evaluated. The results indicated that the BOF slag had the highest carbonation conversion, approximately 72%, at a reaction time of 1 h, an operating pressure of 101 kPa and a temperature of 60 °C due to its higher BET surface area of BOF slag compared to UF, FA, and BHC slags. The major factors affecting the carbonation conversion are reaction time and temperature. The reaction kinetics of the carbonation conversion can be expressed by the shrinking-core model. The measurements of the carbonated material by the SEM and XRD instruments provide evidence indicating the suitability of using the shrinking-core model in this investigation. Comparison of the results with other studies suggests that aqueous carbonation by slurry reactor is viable due to its higher mass transfer rate.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Carbon sequestration is a promising option for reducing carbon dioxide emissions and helping to improve global warming issue. Both CO₂ capture from emission sources and subsequent transport of the captured CO₂ to isolated reservoirs are essential to carbon sequestration. Carbon-capture technology is a critical process in carbon sequestration, which is affected by environmental factors, carbon-capturing capacity, and cost. In Taiwan, the physical storage of CO₂ is impractical owing to the shortage of available space and suitable locations.

Mineral sequestration is a method that accelerates the natural weathering of silicate minerals to react with CO₂ and form carbonate minerals and silica [1], which are stable end products for further usage or disposal. The mineralogical carbon sequestration contributes to remarkable CO₂ sequestration in the proximity of the emission source, without the need of storing the gas into a geological reservoir. This technology is called *ex situ* mineral sequestration of CO₂ [2]. Besides controlling the reaction conditions, the choices of suitable mineral feedstocks as well as the proper design of a reactor are crucial in achieving high efficiencies in CO₂ sequestration. A possible feedstock for mineral CO₂ sequestration is industrial solid waste including combustion residues, slags, and fly ashes. These materials are generally alkaline and

rich in calcium with the advantages of low costs and widespread availability in industrial areas compared to ores [3]. Using steel-making slag as a feedstock is very attractive, as it has a high calcium oxide content and high alkalinity, which should enhance CO₂ mineralization during the carbonation process. The carbonation reactions were carried out primarily through the reaction of CO₂ with raw CaO-based materials, and CaCO₃ was observed as the predominant carbonation product [4]. Simultaneously, it can reduce the amount of waste and neutralize hazardous material.

In selecting an efficient reactor, the slurry reactor is recommended because it can provide a higher contact frequency between CO₂ and the feedstock and can be operated in a continuous mode. Thus, the objectives of this research work were to investigate the carbonation of the steel-making slags in a slurry reactor. The effects of the operational conditions, including type of steel-making slag, reaction time, reaction temperature, and CO₂ flow rate, on the performance of the carbonation process were evaluated. The reaction kinetics of the carbonation conversion were tested by the shrinking-core model. The results of this study were also compared with those of previous studies in the literature.

2. Materials and methods

2.1. Aqueous carbonation experiments

The aqueous carbonation experiments were conducted using various steel-making slag feedstocks, including ultrafine (UF),

* Corresponding author. Tel.: +886 2 23622510; fax: +886 2 23661642.

E-mail address: pcchiang@ntu.edu.tw (P.-C. Chiang).

fly-ash (FA), blended hydraulic-cement (BHC), and basic oxygen-furnace (BOF) slags from China Hi-ment Corporation (Kaohsiung, Taiwan). All slags were ground and sieved. Slags with particle size of less than 44 μm were selected and then placed in a reactor that contained distilled water at a designated temperature. CO_2 was injected into the reactor continuously at 101.3 kPa and a constant flow rate. The schematic diagram for carbonation of steel-making slag in a slurry reactor is shown in Fig. 1. The slurry reactor used for carbonation consisted of a glass column 150 cm tall and 3 cm in inner diameter.

The operation factors including the reaction time (t), feedstock, reaction temperature (T), and flow rate (Q) were performed at different levels of operating conditions. The above experiments were conducted to minimize energy and chemical consumption. The composition of the fresh slag was analyzed by inductively coupled plasma atomic-emission spectroscopy (ICP-AES) after total digestion. The conversion of carbonation products was determined quantitatively by thermogravimetric analysis (TGA) and qualitatively by X-ray diffractometer (XRD), and scanning-electron microscope (SEM).

2.2. TGA

A thermogravimetric analyzer (TGA-51, Shimadzu) is an analytical device for determining the change of sample weight with respect to the temperature change. Three weight fractions according to (1) moisture, (2) organic, elemental carbon and MgCO_3 and (3) CaCO_3 contents were determined under the following temperatures: (1) 25–105 $^\circ\text{C}$, (2) 105–500 $^\circ\text{C}$, and (3) 500–780 $^\circ\text{C}$. The weight loss of calcium carbonate between 520 and 780 $^\circ\text{C}$ is due to the release of CO_2 [4–6]. The weight variation of the slags from the TGA measurements is presented in Fig. 2 which indicates that the CO_2 release at high temperature (above 800 $^\circ\text{C}$) can be neglected. In addition, the initial carbonation level of the fresh slags was found to be negligible based on TGA curves. Thus, the CO_2 in the carbonation process was expressed by $\text{CO}_2(\text{wt}\%) = \Delta m_{520-780^\circ\text{C}}/m_{105^\circ\text{C}}$. The carbonation conversion (δ_{Ca}) can be determined from the total calcium

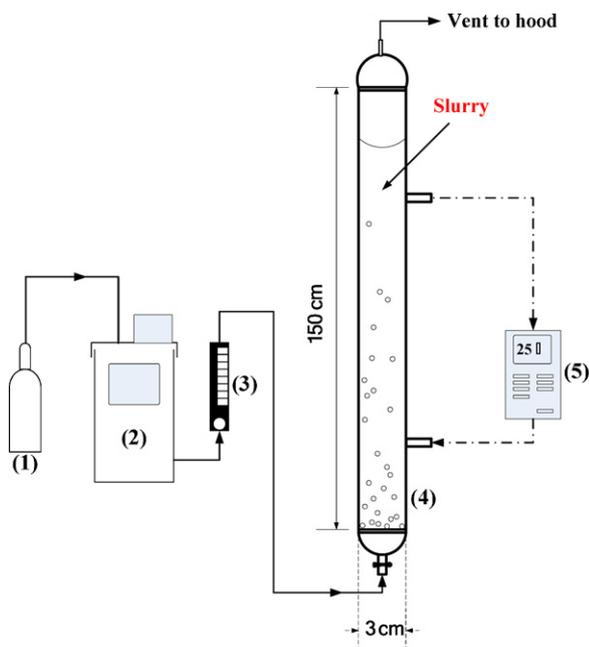


Fig. 1. The schematic diagram of the experimental set-up for carbonation of steel-making slag in a slurry reactor. 1: carbon-dioxide gas cylinder; 2: circulating bath; 3: rotameter; 4: slurry reactor; 5: thermo couple.

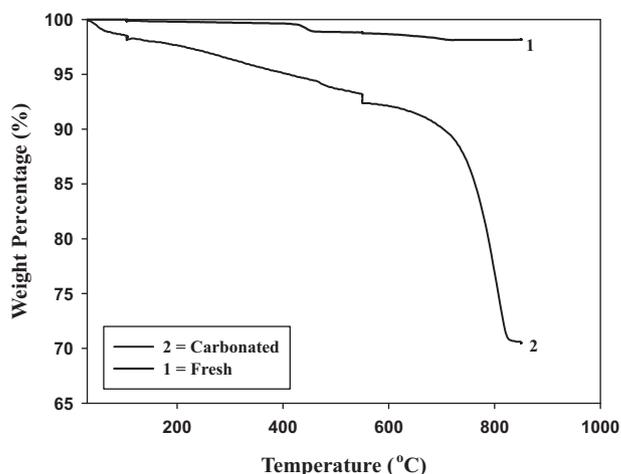


Fig. 2. TGA curves of the fresh and carbonate basic oxygen furnace slags (carbonation conditions: pressure = 14.7 psig CO_2 ; temperature = 70 $^\circ\text{C}$; particle size <44 μm ; L/S ratio = 10 mL g^{-1} ; gas flow rate = 0.1 L min^{-1}).

content of the carbonation product, and initial carbonate content is negligible [6]:

$$\delta_{\text{Ca}} (\%) = \frac{(\text{CO}_2(\text{wt}\%)/(100 - \text{CO}_2(\text{wt}\%))) \times (MW_{\text{Ca}}/MW_{\text{CO}_2})}{\text{Ca}_{\text{total}}} \quad (1)$$

where δ_{Ca} is carbonation conversion, MW_{Ca} and MW_{CO_2} are the molar weight of Ca and CO_2 in kg mol^{-1} , respectively, and Ca_{total} is the total Ca content of the fresh sample in kg kg^{-1} .

2.3. SEM and XRD

The SEM (JSM-6500F, JEOL) used in this study is capable of producing high-resolution images of a sample and identifying CaCO_3 precipitated in the carbonation products. X-ray powder diffraction (XRD) (X'Pert Pro, PANalytical) is an analytical technique, used herein to identify and characterize CaCO_3 crystals in the carbonation products. Monochromatic X-rays were used to determine the interplanar spacing of the sample atoms with Cu as the anode material ($K\alpha$ -1 wavelength = 1.540598 \AA and $K\alpha$ -2 wavelength = 1.544426 \AA) and 2θ scanning, ranging between 20 $^\circ$ and 70 $^\circ$. When the Bragg conditions for constructive interference are obtained, a “reflection” is produced, and the relative peak height is generally proportional to the number of grains in a preferred orientation.

2.4. Aqueous carbonation

Theoretically, the extent of carbonation increases with reaction time. The aqueous carbonation experiments were conducted with reaction times of up to 240 min. The experimental procedures included the following three steps: aqueous carbon dioxide dissolution, calcium leaching, and calcium carbonate precipitation. Previous studies by Huijgen et al. [7] and Chu [8] indicated that the influence of the liquid-to-solid (L/S) ratio on carbonation was insignificant. Therefore, the effect of the L/S ratio was fixed at 10 mL g^{-1} in this study. In summary, the basic conditions of the carbonation experiment were performed as follows: $T = 70^\circ\text{C}$, $P_{\text{CO}_2} = 101.3 \text{ kPa}$, particle size <44 μm , $Q = 0.1 \text{ L min}^{-1}$, and reaction time = 60 min unless specified.

Table 1
Physico-chemical properties of ultra fine, fly ash, blended hydraulic cement and basic oxygen furnace slags (China Hi-cement Corporation).

	Parameters	UF slag	FA slag	BHC slag	BOF slag
Physical properties	True density (g cm^{-3})	2.89	2.78	2.94	3.51
	Mean diameter (μm)	11.67	17.35	20.63	14.98
	BET surface area ($\text{m}^2 \text{kg}^{-1}$)	148	237	115	279
Chemical properties	SiO_2 (%)	33.74 ± 3.20	39.78 ± 4.33	26.58 ± 1.20	11.15 ± 1.83
	Al_2O_3 (%)	14.38 ± 0.85	16.78 ± 0.93	8.88 ± 0.42	1.55 ± 0.46
	Fe_2O_3 (%)	0.45 ± 0.11	2.98 ± 0.89	2.12 ± 0.53	24.03 ± 1.35
	CaO (%)	41.89 ± 1.50	31.86 ± 6.47	54.19 ± 2.73	51.11 ± 4.82
	MgO (%)	5.56 ± 1.15	5.23 ± 0.76	3.97 ± 1.50	4.17 ± 2.10
	Sulfide sulfur (%)	0.46 ± 0.23	–	0.27 ± 0.27	–
	Sulfur trioxide (%)	0.66 ± 0.16	0.76 ± 0.30	1.55 ± 0.14	0.39 ± 0.39
	Total (%)	97.14 ± 0.37	97.39 ± 0.25	97.56 ± 0.82	92.4 ± 1.56

3. Results and discussion

3.1. Composition analysis of steel-making slags

The chemical compositions of the four feedstocks used in this study, i.e. UF, FA, BHC, and BOF slags, are presented in Table 1. The major components of these four steel-making slags include SiO_2 , Al_2O_3 , Fe_2O_3 , CaO, MgO, S^{2-} and SO_3 . The CO_2 -capturing capacity of the slag material is mainly attributed to two components: CaO and MgO. The higher CaO content would present higher reactivity of the slags for carbonation reaction. The CaO content in the UF, FA, BHC, and BOF slags are 41.89 ± 1.50 , 31.86 ± 6.47 , 54.19 ± 2.73 , and $51.11 \pm 4.82\%$, respectively. Assuming all calcium oxides are converted to calcium carbonate, the capacity of the UF, FA, BHC, and BOF slags would be 0.33, 0.25, 0.43 and 0.40 $\text{kg CO}_2 \text{kg}^{-1}$, respectively.

Fig. 3 shows the SEM images of the fresh (Fig. 3a and b) and carbonated (Fig. 3c and d) BOF slags, which were carbonated at 101.3 kPa and 70 °C. Comparison of the SEM images of the feedstock

before and after carbonation showed that the particle size shrank and cubic particles adhered to the feedstock after the carbonation. According to the images and qualitative analysis, the cubic particles can be composed of calcium carbonate with sizes ranging from 1 to 2 μm in diameter, which was similar to the results reported by Huijgen et al. [3]. The dark cubic particles in the SEM image were also identified as calcium carbonate by XRD analysis. Similar observations were also found in the SEM images of the fresh and carbonated BHC slags.

Fig. 4 shows the XRD images of non-carbonated and carbonated material blended with BHC (Fig. 4d) and BOF (Fig. 4b and e) slags, respectively. The XRD results indicate that the main crystal phase of the fresh BOF slag is calcium oxide (Fig. 4b) and the main phase of the carbonated BHC (Fig. 4d) and BOF (Fig. 4e) slags is calcium carbonate. The peaks in the XRD results of carbonated material appeared at $2\theta = 23.02^\circ$, 29.41° , 35.97° , 43.15° , 47.49° , 48.50° , 57.40° , 60.68° , and 64.68° , as shown in Fig. 4d and e, which can be determined as a pure calcite (CaCO_3). This suggests that the BHC and BOF slags should be carbonated with CO_2 to form CaCO_3 .

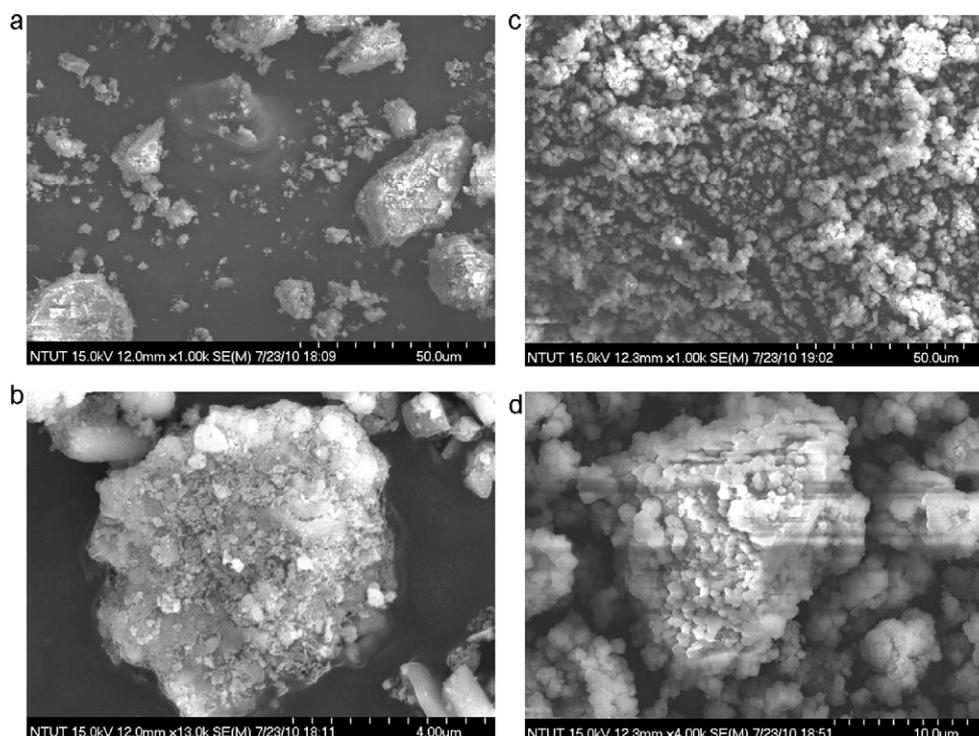


Fig. 3. Scanning-electron micrographs (SEM) of the steel-making slag: (a) fresh BOF slag (50 μm); (b) fresh BOF slag (4 μm); (c) carbonated BOF slag (50 μm); (d) carbonated BOF slag (5 μm).

3.2. Effect of reaction time and feedstocks

The effect of the reaction time on the conversion ratio for various slags is shown in Fig. 5. The results indicate that the carbonation rate decreased as the reaction time elapsed. The reaction leveled off after 60 min, indicating that the carbonation reaction had a limited conversion because of the barrier formed during the carbonation reaction, which was consistent with the findings of Huijgen et al.

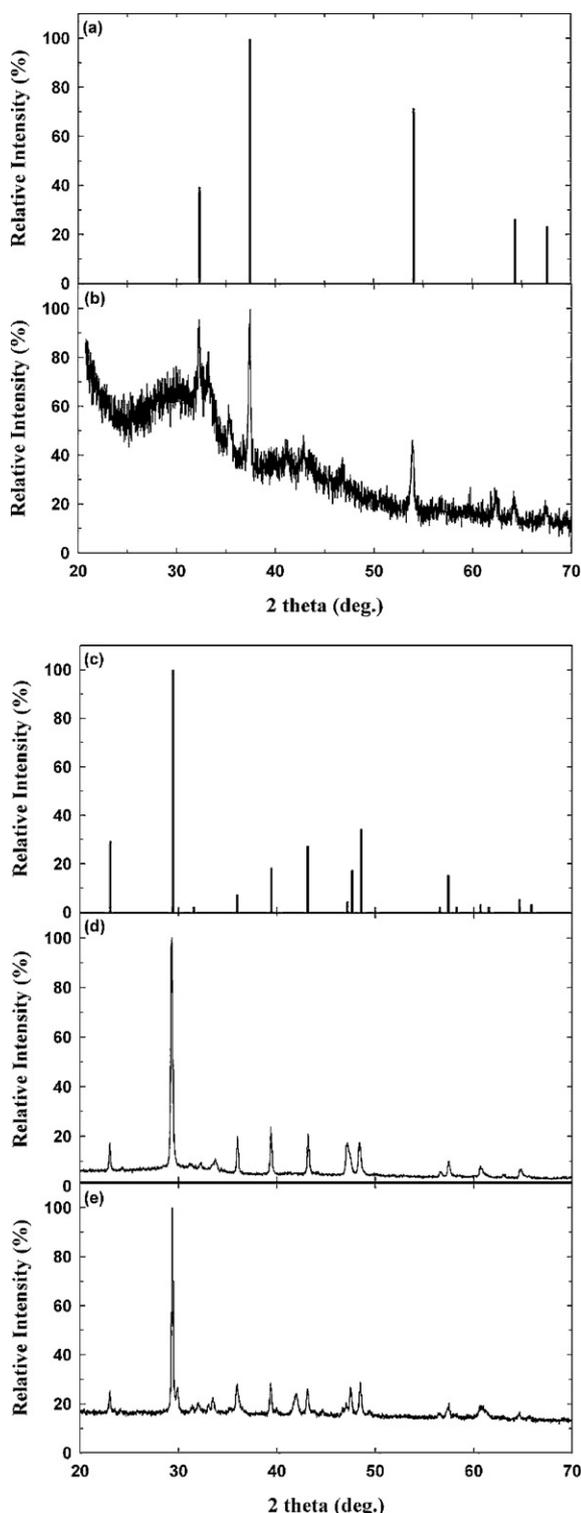


Fig. 4. XRD spectra with peak identifications: (a) pure calcium oxide (CaO); (b) fresh BOF slag; (c) pure calcite (CaCO₃); (d) carbonated BHC slag; (e) carbonated BOF slag.

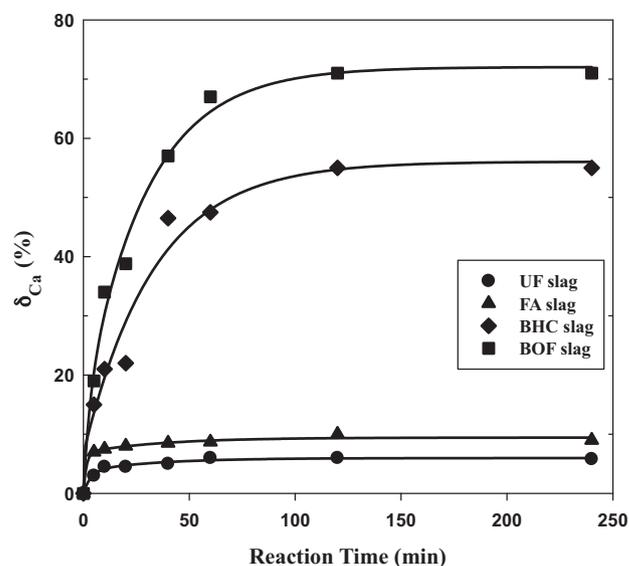


Fig. 5. Influence of reaction time on the carbonation conversion of the steel-making slag (carbonation conditions: pressure = 14.7 psig CO₂; temperature = 70 °C; particle size <44 μm ; L/S ratio = 10 mL g⁻¹; flow rate = 0.1 L min⁻¹).

[3]. It was noted that the maximum efficiencies in carbonation conversion (δ_{Ca}) at 60 min for BHC and BOF slags were 48 and 68%, respectively, which indicates that BOF has a relatively higher conversion.

A possible explanation of the above phenomenon is the different pH values observed for various feedstocks during the carbonation process. Furthermore, in aqueous carbonation, carbon-dioxide solubility is pH-dependent due to the dissolution of carbonic acid into bicarbonate ions and subsequently carbonate ions. The main carbonate species found after CO₂ dissolution in water is bicarbonate when the pH value is near 8.5 or carbonate when the pH value is higher than 11. A high pH value provides more carbonate ions in the water and enhances the carbonation process. After aqueous carbonation, the pH of the system gradually dropped to about 3. Since the optimal pH for the aqueous carbonation was around 10, the reaction time was selected at 60 min in this investigation.

In addition, the above phenomenon could also be explained by the physico-chemical properties of slags. According to Table 1, the content of calcium oxide of BHC is 54.19 ± 2.73 (wt.%), which is higher than that of BOF, i.e. 51.11 ± 4.82 (wt.%). However, the carbonation conversion (δ_{Ca}) of BOF (68%) is higher than that of BHC (48%) due to the relatively higher BET surface area of BOF shown in Table 1. The actual capacities of CO₂ captured per gram of slag were measured at about 0.02, 0.03, 0.21, and 0.27 g CO₂ captured/g for the UF, FA, BHC, and BOF slags, respectively.

3.3. Effect of flow rate

The conversion decreases moderately with increasing flow rate in the slurry reactor, as illustrated in Fig. 6. This is because the channeling effect in the slurry reactor becomes significant at a high flow rate, resulting in a poor gas–liquid mass transfer rate. For the BOF slag, the conversion (δ_{Ca}) would decrease sharply at flow rates lower than 1 L min⁻¹ and then gradually approach a stable value of about 52%. It is noted that the conversion consistently decreases with increasing flow rate in the case of the BHC slag. Therefore, a small flow rate of 1 L min⁻¹ is required for a high conversion in a slurry reactor, suggesting that the flow rate should be limited to a certain value that is able to make the slurry reactor in a fluidization mode.

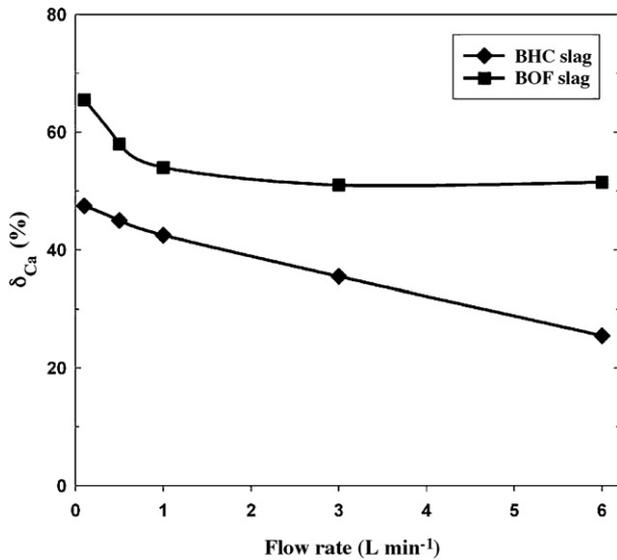


Fig. 6. Influence of flow rate on the carbonation conversion of the BHC and BOF slags (carbonation conditions: pressure = 14.7 psig CO₂; temperature = 70 °C; particle size <44 μm; L/S ratio = 10 mL g⁻¹).

3.4. Effect of reaction temperature

The aqueous carbonation experiments using the BOF were conducted at reaction temperatures ranging from 30 to 80 °C with a reaction time of 60 min, as shown in Fig. 7. The carbonation conversion is dependent on the reaction temperature and exhibits the maximum value at 60 °C. The reaction temperature would affect several system parameters including the reaction kinetics, equilibrium, CO₂ dissolution, and calcium leaching simultaneously. The carbonation reaction rate significantly increased with increasing reaction temperature, since the reaction rate constant exponentially increases with increasing temperature as expressed by the Arrhenius equation. In contrast, the carbonation reaction is exothermic; therefore, an increase in temperature may lead to a decrease in the equilibrium constant based on Le Chatelier's principle. As a result, the dissolution of CO₂ and the leaching rate of

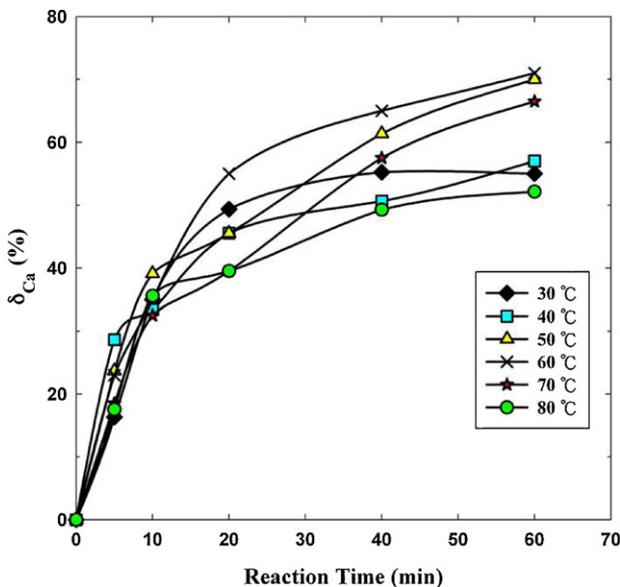


Fig. 7. Influence of reaction temperature on the carbonation conversion of the BOF slag (carbonation conditions: pressure = 14.7 psig CO₂; particle size <44 μm; L/S ratio = 10 mL g⁻¹, gas flow rate = 0.1 L min⁻¹).

calcium ions at higher temperature would decrease and increase, respectively.

According to the results shown in Fig. 7, the carbonation reaction can be categorized into two main regimes. At reaction temperatures below 60 °C, the carbonation conversion increased with increasing reaction temperature due to the higher leaching rate of calcium ions [7,9]. The CaCO₃ crystallization reaction was thus accelerated at higher temperature in this regime (30–60 °C). However, when the reaction temperatures were above 60 °C, boiling in the slurry reactor was accompanied by the low dissolution of CO₂, resulting in a decrease in the carbonation conversion. In this regime (60–80 °C), the CO₂ solubility is likely to be the key factor affecting the carbonation conversion.

3.5. Kinetic modeling of carbonation reaction

A shrinking-core model can be performed to analyze the experimental data during trials of carbonation conversion of calcium oxide [10]. The reaction mechanism consists of (a) diffusion of a gaseous reactant through the boundary film surrounding the solid particles; (b) penetration and diffusion of the reactant through the layer of solid product until it reaches the surface of the unreacted core; and (c) reaction over the surface of the core.

The measurements of the carbonated material by the SEM in conjunction with XRD provide evidence indicating the suitability of using the shrinking-core model in this investigation. The surface composition and molecular structure were found to be changed in the course of carbonation. The small CaCO₃ particles were formed on the surface of slags, which accounted for the formation of a protective layer around the reacting particles. A main assumption corresponding to the shrinking-core model is that the chemical reactions are infinitely faster than the CO₂ diffusion through the reacted layer [11].

Therefore, a carbonation process using CaO with flue gases at high CO₂ concentrations by considering the limitation of the product-layer diffusion could be expressed as follows [12,13]:

$$t = \frac{\rho_B r^2}{6bD_e C_A} [1 - 3(1 - X_B)^{2/3} + 2(1 - X_B)] \quad (2)$$

where ρ_B is the molar density of the slag of 3.19×10^{-2} mol cm⁻³, r is the particle size of 5×10^{-4} cm, b is the stoichiometric coefficient of unit for the carbonization of CaO, D_e is the effective diffusion coefficient (cm² s⁻¹), and C_A is the molar concentration of CO_{2(aq)} (mol cm⁻³). C_A can be calculated by Henry's law and the Van't Hoff equation as 2.98×10^{-5} , 2.31×10^{-5} , 1.82×10^{-5} , 1.46×10^{-5} , and 1.18×10^{-5} mol cm⁻³ at 30, 40, 50, 60, and 70 °C, respectively.

The values of D_e are estimated based on the experimental data as shown in Fig. 8. The D_e values, which apparently increase with increasing temperature, are in the range of 2.85×10^{-4} to 8.68×10^{-4} cm² s⁻¹ with high R^2 values ranged from 0.92 to 0.99. The obtained D_e values are consistent with the findings of studies by Nikulshina et al. [14] and Xie et al. [15], which indicates that the diffusion-control mechanism should be the rate-determination step of the carbonation reaction, which is also consistent with the findings of Nikulshina et al. [14] and Xie et al. [15].

3.6. Comparison of carbonization conversion in literatures and this study

The consumption of energy for mineral sequestration has decreased with recent technology developments. Many researchers have attempted to capture CO₂ with lower power and chemical usage. In this study, the highest conversion (δ_{Ca}) for the BOH oxygen-furnace slag was 72%, when the aqueous carbonation was conducted at 101.3 kPa and 60 °C in a slurry reactor. The factors

Table 2

Comparison of experimental results in the literatures and those of this study.

Feedstock	Method	Reactor	Temperature [°C]	Pressure [psig]	Particle size [μm]	Time [min]	Conversion [%]	Reference
Mg(OH) ₂	Direct carbonation	BR	500	4930	<20	120	100	Lackner et al. [17]
Serpentine	Direct carbonation	BR	300	4930	<50	120	30	Lackner et al. [17]
Olivine	Aqueous carbonation	BR	185	1682	<37	1440	91	O'Connor et al. [18]
Serpentine	pH-swing	BR	70	14.7	<75	10	42	Park and Fan [19]
Converter slag	pH-swing	CSTR	40	1.9	<63	60	72.8	Kodama et al. [16]
Blast-furnace slag	pH-swing	CSTR	30	14.7	<10	15	72.5	Eloneva et al. [20]
Ultrafine slag	pH-swing	BR	40	50	<44	15	46.2	Chu [8]
Blended hydraulic-cement slag	Aqueous carbonation	BR	160	700	<44	60	58.2	Chu [8]
Basic oxygen-furnace slag	Aqueous carbonation	FBR	60	14.7	<44	60	72.2	This study

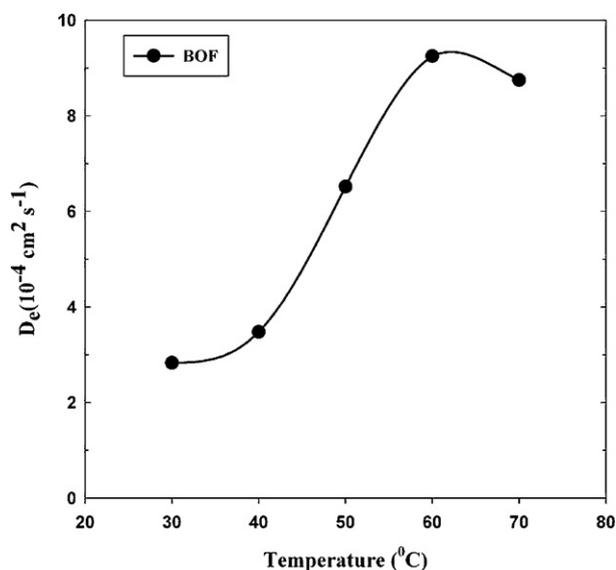


Fig. 8. Variation of D_e with reaction temperature for the carbonation conversion of the BOF slag (carbonation conditions: pressure = 14.7 psig CO_2 ; particle size <44 μm ; L/S ratio = 10 mL g^{-1} , gas flow rate = 0.1 L min^{-1}).

affecting the system operation were reaction temperature, initial pH with various feedstocks, and CO_2 flow rate. The optimal operating conditions, calculated by the response surface methodology (RSM), were found to be $T=70^\circ\text{C}$, $P_{\text{CO}_2} = 101.3 \text{ kPa}$ and $Q=0.1 \text{ L min}^{-1}$ under a reaction time of 1 h. The predicted optimal conversion (δ_{Ca}) is 71%, which is very close to the observed value.

Table 2 shows the comparison of the experimental results obtained in this study with others reported in the literature. It appears that the results of the pH-swing method from Kodama et al. [16] were similar to those observed in this study. However, previous studies using the pH-swing method consumed a great amount of base and acid solvents, which would cause adverse effects on the environment. On the other hand, the studies reported by Lackner et al. [17] and O'Connor et al. [18] exhibited a very high carbonation conversion (91–100%) at the expense of higher temperature (185–500 °C) and pressure (1682–4930 psig). This study demonstrates that a higher carbonation conversion (72%) can be achieved at relatively lower temperature (60 °C) and pressure (14.7 psig).

4. Conclusions

UF, FA, BHC and BOF slags were selected as feedstocks for the aqueous carbonation process in this study. The initial carbonation level of the fresh slags was found to be negligible based on TGA curves. Although the BHC slag has the highest theoretical sequestration capacity estimated by the calcium oxide content, the experimental results indicated that the BOF slag exhibited better performance in terms of carbonation due to its relatively higher

BET surface area and higher pH value in the course of aqueous carbonation. The feedstock in this reaction was alkaline calcium-rich particles, which reacted with CO_2 dissolved in the aqueous slurry to form calcium carbonate. Most of the carbonation reaction occurred in the first 1 h of reaction time. The conversion efficiency was significantly lower after 4 h of reaction time. The optimal temperature for carbonation was at 60 °C.

The surface composition and molecular structure were found to be varied in the course of carbonation based on the measurements of SEM and XRD instruments. Therefore, the kinetics of this reaction can be described by the shrinking-core model, which indicates that the ash diffusion-control mechanism should be the rate-determination step of the carbonation reaction. In the product analysis, the CaCO_3 product formed was a crystallized calcite type, which was also confirmed by the SEM and XRD analyses. It is thus concluded that aqueous carbonation of the steelmaking slags by slurry reactor is viable due to its high mass-transfer rate under rapid-mixing operating conditions.

It also suggests that grinding BOF slag into the powder utilizing energy thereby leading to CO_2 emission should be critically assessed by using the LCA (Life Cycle Assessment). In other words, if the technology of mineral sequestration in a slurry reactor could be developed and deployed, our future research work should be focused on the carbon and energy foot print studies.

Acknowledgment

This study was support by National Science Council of Taiwan under Grant No. NSC 98-3114-E-007-013.

References

- [1] K.S. Lackner, A guide to CO_2 sequestration, *Science* 300 (2003) 1677–1678.
- [2] G. Montes-Hernandez, R. Pérez-López, F. Renar, J.M. Nieto, L. Charlet, Mineral sequestration of CO_2 by aqueous carbonation of coal combustion fly-ash, *J. Hazard. Mater.* 161 (2009) 1347–1354.
- [3] W.J.J. Huijgen, G.J. Witkamp, R.N.J. Comans, Mineral CO_2 sequestration by steel slag carbonation, *Environ. Sci. Technol.* 39 (2005) 9676–9682.
- [4] D.N. Huntzinger, J.S. Gierke, S.K. Kawatra, T.C. Eisele, L.L. Sutter, Carbon dioxide sequestration in cement kiln dust through mineral carbonation, *Environ. Sci. Technol.* 43 (2009) 1986–1992.
- [5] G. Villain, M. Thiery, G. Platret, Measurement methods of carbonation profiles in concrete: thermogravimetry, chemical analysis and gammadensimetry, *Cement Concrete Res.* 37 (2007) 1182–1192.
- [6] E. Bouquet, G. Leyssens, C. Schönnenbeck, P. Gilot, The decrease of carbonation efficiency of CaO along calcination–carbonation cycles: experiments and modelling, *Chem. Eng. Sci.* 64 (2009) 2136–2146.
- [7] W.J.J. Huijgen, G.J. Witkamp, R.N.J. Comans, Mechanisms of aqueous wollastonite carbonation as a possible CO_2 sequestration process, *Chem. Eng. Sci.* 61 (2006) 4242–4251.
- [8] H.W. Chu, CO_2 sequestration by carbonation of alkaline solid waste, Master Thesis, Graduate Institute of Environmental Engineering, National Taiwan University, 2007.
- [9] S.N. Lekakh, C.H. Rawlins, D.G.C. Robertson, V.L. Richards, K.D. Peaslee, Kinetic of aqueous leaching and carbonization of steelmaking slag, *Metall. Mater. Trans. B* 39 (2008) 125–134.
- [10] S.W. Park, D.H. Sung, B.S. Choi, K.J. Oh, K.H. Moon, Sorption of carbon dioxide onto sodium carbonate, *Sep. Sci. Technol.* 41 (2006) 2665–2684.

- [11] M. Castellote, C. Andrade, Modelling the carbonation of cementitious matrixes by means of the unreacted-core model, UR-CORE, *Cement Concrete Res.* 38 (2008) 1374–1384.
- [12] H. Gupta, L.S. Fan, Carbonation–calcination cycle using high reactivity calcium oxide for carbon dioxide separation from flue gas, *Ind. Eng. Chem. Res.* 41 (2002) 4035–4042.
- [13] D. Lee, An apparent kinetic model for the carbonation of calcium oxide by carbon dioxide, *Chem. Eng. J.* 100 (2004) 71–77.
- [14] V. Nikulshina, M.E. Galvez, A. Steinfeld, Kinetic analysis of the carbonation reactions for the capture of CO₂ from air via the Ca(OH)₂–CaCO₃–CaO solar thermochemical cycle, *Chem. Eng. J.* 129 (2007) 75–83.
- [15] X.Y. Xie, B.J. Zhong, W.B. Fu, Y. Shi, Measurement of equivalent diffusivity during the calcination of limestone, *Combust. Flame* 129 (2002) 351–355.
- [16] S. Kodama, T. Nishimoto, N. Yamamoto, K. Yogo, K. Yamada, Development of a new pH-swing CO₂ mineralization process with a recyclable reaction solution, *Energy* 33 (2008) 776–784.
- [17] K.S. Lackner, D.P. Butt, C.H. Wendt, Progress on binding CO₂ in mineral substrates, *Energy Convers. Manage.* 38 (1997) S259–S264.
- [18] W.K. O'Connor, D.C. Dahlin, D.N. Nilsen, S.J. Gerdemann, G.E. Rush, R.P. Walters, P.C. Turner, Research status on the sequestration of carbon dioxide by direct aqueous mineral carbonation, in: 18th Annual International Pittsburgh Coal Conference, Newcastle, Australia, 2001.
- [19] A.H.A. Park, L.S. Fan, CO₂ mineral sequestration: physical activated dissolution of serpentine and pH swing process, *Chem. Eng. Sci.* 59 (2004) 5241–5247.
- [20] S. Eloneva, S. Teir, J. Salminen, C.J. Fogelholm, R. Zevenhoven, Fixation of CO₂ by carbonating calcium derived from blast furnace slag, *Energy* 33 (2008) 1461–1467.