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Optimization of Cyclone-Type Rotary Kiln Reactor for Carbonation of BOF Slag

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Abstract: Mineral carbonation of the basic oxygen furnace (BOF) slag produced in the steel-making process not only provides an effective approach for carbon dioxide storage, but also stabilizes the slag such that it can be reused as a construction material. Generally speaking, carbonation performance improves as the time for which the carbon dioxide resides within the reactor increases. This research proposes a method to increase the residence time of carbon dioxide in the cyclone converter slag carbonization kiln by adjusting the inclination angle and length of the feed pipe. Therefore, it has the same effect of increasing the flow path length of the cyclone in the reactor. The optimal values of the inclination angle and length of the gas inlet tube are determined using the robust Taguchi design method. Computational fluid dynamics simulation results show that the optimized reactor design increases the average residence time of carbon dioxide gas by 60.4%, compared with the original rotating reactor design with a straight (non-cyclonic) flow path. Moreover, the experimental results show that the optimized design increases the carbon dioxide storage capacity from 12.15 g per kilogram of BOF slag in the original rotary kiln reactor to 16.00 g in the re-designed reactor.

Keywords: carbon dioxide storage; rotary reactor; residence time

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1. Introduction

With fossil fuels continuing to be the primary source of energy for meeting the world's power demands, the amount of carbon dioxide (CO_2) gas released into the atmosphere is increasing year by year. The greenhouse effect resulting from the buildup of CO_2 in the atmosphere has brought about many changes in the Earth's climate and has led to changing weather patterns, rising sea levels, frequent natural disasters, poor crop harvests, extended periods of drought and famine, and so on. Consequently, how to reduce the production of CO_2 and develop effective CO_2 storage methods has attracted much attention in recent years.

The steel industry is not only one of the largest contributors to annual CO_2 production, but also produces a large amount of basic oxygen furnace (BOF) slag as a waste product. BOF slag has a high free lime content, and hence its disposal poses a significant environmental challenge. As a result, many applications for reusing BOF slag have been proposed, including cement production, concrete aggregation, soil conditioning, marine reconstruction, and so on. However, the high content of free CaO in BOF slag not only has high alkalinity, but also has the problem of volume expansion, which severely limits its ability to be reused as a building material [1–3]. Therefore, effective methods to reduce the content of free CaO are needed to increase its reuse potential. As described by Santos et al. in [4], four basic methods are commonly used for the stabilization of BOF slag, namely (i) weathering in slag pits to convert the CaO into hydrated lime; (ii) steam hydration; (iii) the addition of SiO_2 and O_2 to the slag pot to dissolve the free lime and chemically bond it as silicate; and (iv) control of the slag cooling path to stabilize the tri-calcium silicate phase. However, all four methods require a long processing time and a large space, which limit their practicality for widespread application. Accordingly, Santos et al. [4] proposed a hot-stage carbonation approach for reducing the free CaO content of BOF slag during the pouring and solidification stages. It was shown that the proposed method yielded an effective reduction in the free CaO content and stabilized the volume of the slag as a result. Pan et al. [5] examined the optimum gas-phase mass transfer rate in a high-gravity carbonation process for BOF slag utilizing a rotating packed bed (RPB) furnace and cold-rolling mill wastewater (CRW). The results showed that the mass transfer resistance of carbonation lay mainly on the liquid side. Ghasemi et al. [6] performed a life cycle assessment of two carbonation processes (slurry phase and wet carbonation) aimed at permanent CO_2 storage using BOF slag as the alkalinity source. It was shown that, of the two methods, the slurry route resulted in a higher net avoided greenhouse warming potential, but led to a greater environmental impact due to its higher material and energy requirements. Polettini et al. [7] investigated the individual and joint effects of the main carbonation operating parameters (namely the total pressure, the CO_2 concentration in the gas phase, and the temperature) on the CO_2 sequestration yield of a direct aqueous carbonation process. The experimental results showed that the optimal sequestration yield was achieved using a pressure of 5 bar, a CO_2 concentration of 40%, and a temperature of 50 °C. Jiang and Ling [8] investigated the effects of two accelerated carbonation approaches, namely during-granulation and post-curing, on the properties of the artificial aggregates produced using 100% BOF slag. It was shown that the post-curing approach resulted in a significant improvement in the overall properties of the BOF slag; most notably including a strength enhancement of 220%. Chang et al. [9] investigated the effects of accelerated carbonation on the pH value and leaching behavior of BOF slag and found that the absorption of CO_2 reduced the alkalinity of the BOF slag from the initial value of 12.73 to the final value of 9.36. They also found that increasing the degree of carbonization can reduce the leaching of Ca but increasing the leaching of Si.

Current methods for CO_2 storage include mainly underground geological storage, ocean storage, mineral carbonation storage, and biological storage [10]. Mineral carbonation storage involves the use of natural minerals rich in calcium / magnesium and other alkaline substances such as olivine, wollastonite, and serpentine stones to absorb the CO_2 in a solid matrix. The use of BOF slag as a source material for CO_2 mineralization storage has attracted growing interest in recent decades as a means not only of reducing the level of CO_2 emissions to the atmosphere, but also stabilizing the BOF slag and improving its potential for reuse as a construction material [11–14]. Pan et al. [15] showed that the accelerated carbonation of BOF slag in a RPB furnace provides a viable approach for CO_2 capture, as rapid reaction kinetics are induced even under relatively mild reaction conditions. The same group [16] evaluated the engineering, environmental, and economic aspects of a high-gravity carbonation process. It was shown that the total CO_2 emissions of the steel and cement industries could be reduced by up to 6.5% by implementing a waste-to-resource supply chain between them. Ding et al. [17] investigated the thermochemical energy storage performance of methane reforming using high temperature slag. The results showed that the methane conversion and thermochemical energy storage efficiency both improved as the initial slag temperature increased.

Ko et al. [18] proposed an accelerated carbonation process for treating BOF slag, in which the traditional static reactor used for carbonation purposes was replaced with a rotary kiln reactor to increase the contact level between the solid phase and the gas phase. The experimental results showed that, given the use of appropriate carbonation parameters (e.g., the temperature, relative humidity, and CO_2 concentration), the bearing strength and particle cylindrical crushing strength of the BOF slag were both significantly improved following carbonation. However, studies have demonstrated that the CO_2 gas in the traditional rotary kiln reactor enters the reactor from the center of the left plate, flows along the rotation axis of the reactor tube, and then quickly leaves the reactor from the center of the right plate, so the flow path is very short. Thus, the CO_2 gas residence time in the reactor is relatively short; the carbonation performance is also reduced. Chang et al. [19] analyzed the flow field in a rotary kiln and proposed that arranging the inlet and outlet pipes obliquely to the reactor centerline can increase the residence time. However, the proposed sealed bearings are too large in diameter and difficult to maintain. Therefore, the applicability of the proposed system is impractical.

In order to solve the above problems, the present study proposes a novel rotary kiln reactor, which can increase the residence time by bending the outlet and inlet tubes of the reactor kiln into an inclination angle with the rotation axis of the reactor (see Figure 1); a slow-moving cyclone flow structure is produced in the rotary kiln reactor (see Figure 2). Notably, the inlet and outlet tubes have a diameter of just 60 mm, and hence the bearings are available as standard off-the-shelf components. As a result, their procurement cost is greatly reduced. Furthermore, the smaller scale of the bearings simplifies the disassembly process, and therefore reduces both the time and the expense of the maintenance process. As shown in Figure 1, the end-sections of the inlet and outlet tubes are bent in the downward and upward directions, respectively, so as to induce a longer cyclonic flow path within the reactor cylinder (see Figure 2). The optimal design of the inlet tube (i.e., the inclination angle and length) is determined using the robust Taguchi design method in conjunction with CFD simulations. The practical feasibility of the proposed cyclone-type reactor is demonstrated both numerically and experimentally. It is demonstrated that the proposed reactor increases the CO_2 absorption per kilogram of BOF slag by 31.7%, compared to the original (non-cyclone) rotary kiln reactor proposed by Ko et al. [18].

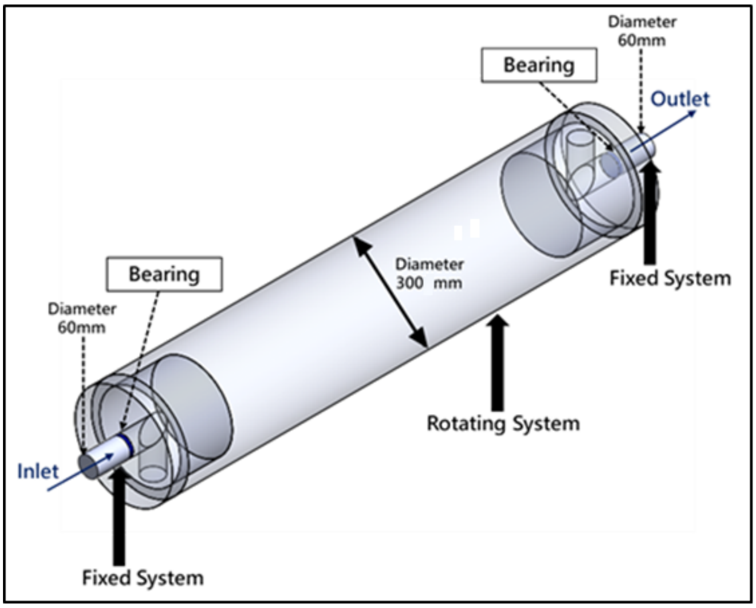

Figure 1. CAD model of the proposed cyclone-type rotary kiln reactor.

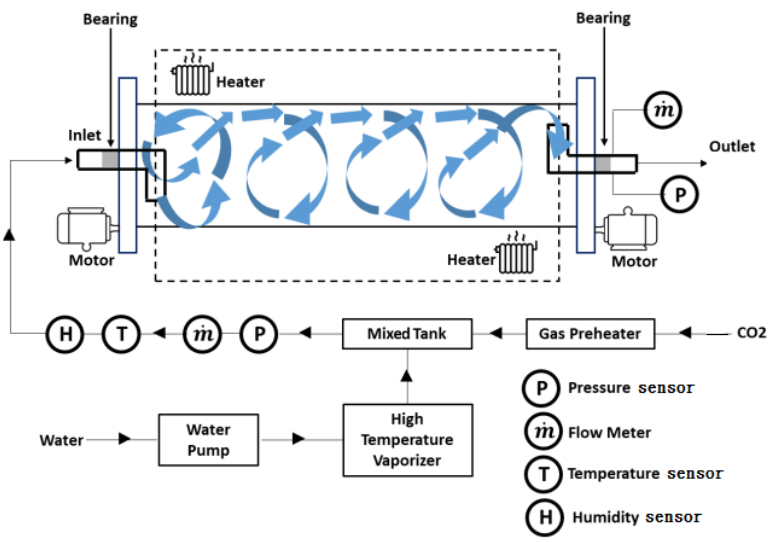

Figure 2. Flow field trajectory in the proposed cyclone-type rotary kiln reactor.

2. Reactor Model

In constructing the CAD model of the proposed cyclone-type rotary kiln reactor, the length was specified as 1500 mm and the diameter was specified as 300 mm. Moreover, the flow rate of CO_2 gas was set to 1 L/s. The other design and operation parameters are listed in Table 1.

Table 1. Design parameters for the proposed cyclone-type rotary kiln reactor.

Diameter of reactor	∅ 300 mm
Length of reactor	1500 mm
Diameter of inlet/outlet tubes	∅ 60 mm
Rotation speed of reactor	2 rpm
Flow rate of CO_2	1 L/s

As discussed above, the end sections of the inlet and outlet tubes within the reactor were bent in order to enhance the cyclonic airflow within the reactor cylinder. In particular, the length of the cyclonic flow path was increased by optimizing two parameters of the inlet tube, namely the inclination angle (θ) and length (L). The inclination angle (θ) is the angle between the axis of the inlet tube and the vertical line (θ) and the length (L) is measured from the center point of the bending cross-section of the tube to the center point of the exit cross-section, as shown in Figure 3. Moreover, as shown in Table 2, both parameters were assigned four levels and the optimal combination of level settings was determined via CFD simulations, as described in the following.

Table 2. The control parameters and levels set in this study.

Code	Control Parameter (Unit)	Level 1	Level 2	Level 3	Level 4
A	Inclination angle θ (°)	0	30	45	60
B	Length L (mm)	30	60	90	120

In this study, the CFD software was used to predict the CO_2 gas flow in the reactor. The governing equations were formulated as follows:

Mass conservation:

$$\frac{\partial \bar{u}}{\partial x}+\frac{\partial \bar{v}}{\partial y}+\frac{\partial \bar{w}}{\partial z}=0 \tag{1}$$

Momentum conservation:

In accordance with the FVM method, the momentum change rate of the control volume is equal to the sum of the forces acting on the control volume. For a 3D model, these forces contain many tangential force stress and normal force stress components in the *x*, *y*, and *z* directions, respectively. The Navier–Stokes equations mathematically express conservation of momentum for Newtonian fluids as follow:

$$\frac{\partial \bar{u}}{\partial t}+\bar{u}\frac{\partial \bar{u}}{\partial x}+\bar{v}\frac{\partial \bar{u}}{\partial y}+\bar{w}\frac{\partial \bar{u}}{\partial z}=-\frac{1}{\rho}\frac{\partial \bar{p}}{\partial x}+\frac{\partial}{\partial x}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial x}-\overline{u'u'}\right)+\frac{\partial}{\partial y}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial y}-\overline{u'v'}\right)+\frac{\partial}{\partial z}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial z}-\overline{u'w'}\right) \quad (2)$$

$$\frac{\partial \bar{v}}{\partial t}+\bar{u}\frac{\partial \bar{v}}{\partial x}+\bar{v}\frac{\partial \bar{v}}{\partial y}+\bar{w}\frac{\partial \bar{v}}{\partial z}=-\frac{1}{\rho}\frac{\partial \bar{p}}{\partial x}+\frac{\partial}{\partial x}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial x}-\overline{v'u'}\right)+\frac{\partial}{\partial y}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial y}-\overline{v'v'}\right)+\frac{\partial}{\partial z}\left(\frac{u}{\rho}\frac{\partial \bar{u}}{\partial z}-\overline{v'w'}\right) \quad (3)$$

$$\frac{\partial \bar{w}}{\partial t}+\bar{u}\frac{\partial \bar{w}}{\partial x}+\bar{v}\frac{\partial \bar{w}}{\partial y}+\bar{w}\frac{\partial \bar{w}}{\partial z}=-\frac{1}{\rho}\frac{\partial \bar{p}}{\partial x}+\frac{\partial}{\partial x}\left(\frac{u}{\rho}\frac{\partial \bar{w}}{\partial x}-\overline{w'u'}\right)+\frac{\partial}{\partial y}\left(\frac{u}{\rho}\frac{\partial \bar{w}}{\partial y}-\overline{w'v'}\right)+\frac{\partial}{\partial z}\left(\frac{u}{\rho}\frac{\partial \bar{w}}{\partial z}-\overline{w'w'}\right) \quad (4)$$

In this study, the Lagrangian multiplier method was used to determine the residence time of CO_2 gas in the rotary kiln. Assume that the CO_2 gas entering the rotary kiln contains many small dust particles, which are carried by the CO_2 stream through the reactor tube. In the CFD simulation, the trajectory of each dust particle is tracked, and the flow time required from the entrance to the exit is calculated. Then the average flow time of all dust particles is estimated as the residence time of CO_2 gas.

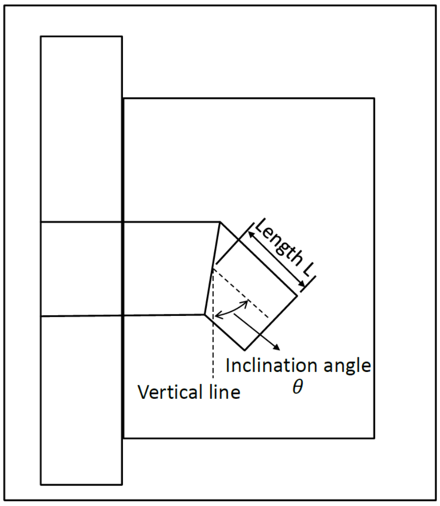

Figure 3. Schematic diagram and definition of inclination angle (θ) and length (L).

As described above, the optimal design of the inlet tube was determined using the robust Taguchi design method [20]. The optimization process involved two control factors (θ and L) and four level settings (Table 2). Thus, in accordance with the Taguchi design method, the experiments were configured in an L16(4^2) orthogonal array (OA), as shown in Table 3. Since the optimization process involved only two factors and four levels, the experiments in the OA were performed in full (i.e., 16 experiments). Consequently, it was unnecessary to calculate the S/N ratio to evaluate and compare the quality of the experimental outcomes. In other words, the optimal settings of θ and L were determined directly from an inspection of the experimental results.

Table 3. $L_{16}(4^2)$ Taguchi orthogonal array.

No:	A Level	B Level	Inclination Angle θ (°)	Length L (mm)
1	1	1	0	30
2	1	2	0	60
3	1	3	0	90
4	1	4	0	120
5	2	1	30	30
6	2	2	30	60
7	2	3	30	90
8	2	4	30	120
9	3	1	45	30
10	3	2	45	60
11	3	3	45	90
12	3	4	45	120
13	4	1	60	30
14	4	2	60	60
15	4	3	60	90
16	4	4	60	120

3. Materials and Test Methods

3.1. BOF Slag

The BOF slag used in the experiments was acquired from a steel manufacturing company in Kaohsiung, Taiwan. The CO_2 gas is manufactured by Sanjie Company with a purity of 99.9%; the source of water vapor is obtained by high-temperature vaporization of DI water, and the resistance value of DI water is above 18 Ω. In order to control the carbonation conditions and ensure a fair comparison between the carbonation performance of the proposed cyclone-type reactor and the carbonation performance of original reactor, a gas mixing chamber was used to adjust the composition of the inlet gas mixture (comprising CO_2 and H_2O) installed in the rotary kiln upstream. (Full details of the preparation and analysis process of the BOF slag are available in [18], so for the sake of brevity, it is omitted here.)

3.2. Carbonation Process

In the process of mineral carbonation, the amount of CO_2 adsorbed is related to the degree of carbonation of the free CaO in the BOF slag. In this experiment, the inlet gas mixture comprised 40% CO_2 and 60% H_2O. The purpose of H_2O vapor was to enhance the combination of the free-CaO and CO_2 to form $CaCO_3$ [3]. The chemical reactions in the resulting CaO-H_2O-CO_2 system were as follows:

$$\mathrm{CaO} + \mathrm{CO_2} \rightarrow \mathrm{CaCO_3} \tag{5}$$

$$\mathrm{CaO} + \mathrm{H_2O} \rightarrow \mathrm{Ca(OH)_2} \tag{6}$$

$$\mathrm{Ca(OH)_2} + \mathrm{CO_2} \rightarrow \mathrm{CaCO_3} + \mathrm{H_2O} \tag{7}$$

$$\mathrm{CO_2} + \mathrm{H_2O} \rightarrow \mathrm{H_2CO_3} \tag{8}$$

$$CaO + H_2CO_3 \rightarrow CaCO_3 + H_2O \quad (9)$$

$$Ca(OH)_2 + H_2CO_3 \rightarrow CaCO_3 + 2H_2O \quad (10)$$

Equation (5) describes the direct reaction between CaO and CO_2 to form $CaCO_3$. Equations (6)–(9) show the joining of CaO and CO_2 to form $CaCO_3$ through the reaction of H_2O. The molar ratio CaO and CO_2 to $CaCO_3$ is 1:1:1. Thus, by measuring the weight (or mole number) of the $CaCO_3$ produced in the reaction process, the weight (or mole number) of CO_2 absorbed can be inferred by deduction. In this study, thermogravimetric analysis (TGA) was used to determine the weight of $CaCO_3$ per kilogram of BOF slag after the carbonation process. The CO_2 absorption per kilogram of BOF slag was then calculated accordingly.

3.3. Thermogravimetric Analysis

The amount of carbonates in the BOF slag can be regarded as the degree of carbonation, which refers to the weight percentage of carbonates in the BOF slag after the accelerated carbonation experiment. Using thermogravimetric analysis (TGA), $CaCO_3$ in the BOF slag can be decomposed to produce CO_2 gas and dissipate due to high temperature. The original mass of $CaCO_3$ can be calculated from the weight loss. The test procedure is to put 30 g of the carbonated BOF slag sample on the platinum sample pan, and then put it into the chamber of the thermogravimetric analyzer, and pass nitrogen into the chamber of the thermogravimetric analyzer at a flow rate of 4 L/min. Then heat the sample to 1000 °C at a heating rate of 20 °C/min and observe the weight change.

4. Results and Discussions

4.1. CFD Simulation Results

Table 4 shows the CFD simulation results of the average residence time of the CO_2 gas in the reaction chamber under the 16 cases in Table 3. It can be observed that the average residence time is between 55.81 s and 101.08 s. In other words, the two design parameters have a significant effect on the carbonation performance of the proposed reactor.

The results presented in Table 4 show that the longest and shortest average residence times are obtained in the 4th and 15th experiments, respectively. The corresponding geometries of the inlet tube are shown in Figure 4a,b, respectively. Figure 5 shows the flow field trajectory within the reactor for the level settings considered in the 4th experiment (i.e., θ = 0° and L =120 mm). It is seen that the inlet gas impacts directly on the reactor wall and is imparted a rotation effect by the rotating wall surface, which immediately produces a cyclonic gas flow. Figure 6 shows the flow field trajectory within the reactor for the level settings considered in the 15th experiment (i.e., θ = 60° and L = 90 mm). In this case, the gas moves forward through a certain distance before impacting the reactor wall. Consequently, the onset of the cyclonic rotation effect is delayed, and hence the residence time of the CO_2 gas is reduced in the rotary kiln.

Table 4. CFD simulation results for the average residence time of the CO_2 gas within the reaction chamber.

Experimental Run	Inclination Angle θ (°)	Length L (mm)	Residence Time (s)
1	0	30	84.140
2	0	60	89.574
3	0	90	97.077
4	0	120	101.08
5	30	30	72.089
6	30	60	78.625
7	30	90	78.146
8	30	120	82.743
9	45	30	70.731
10	45	60	79.041
11	45	90	74.642
12	45	120	80.181
13	60	30	70.333
14	60	60	69.444
15	60	90	55.818
16	60	120	57.007

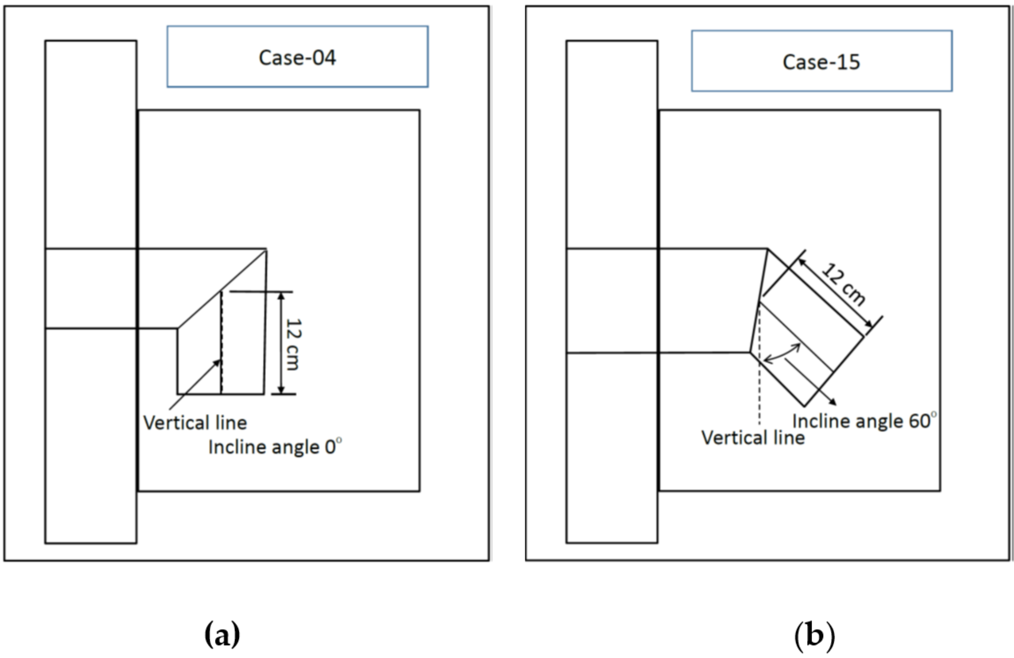

Figure 4. Geometry of inlet tube in: (**a**) 4th experiment; and (**b**) 15th experiment.

Table 5 compares the average residence times of the CO_2 gas in the original rotary kiln (without bending the inlet and outlet tubes) and the cyclone-type rotary kiln reactor proposed in the present study (Run #4 in Table 4). As shown, the residence time of the CO_2 gas in the proposed cyclone-type reactor (101.08 s) is 38.05 s longer than that in the original (non-cyclone flow) rotary kiln reactor (i.e., 63.03 s), which corresponds to an improvement of around 60.4% in the available carbonation reaction time.

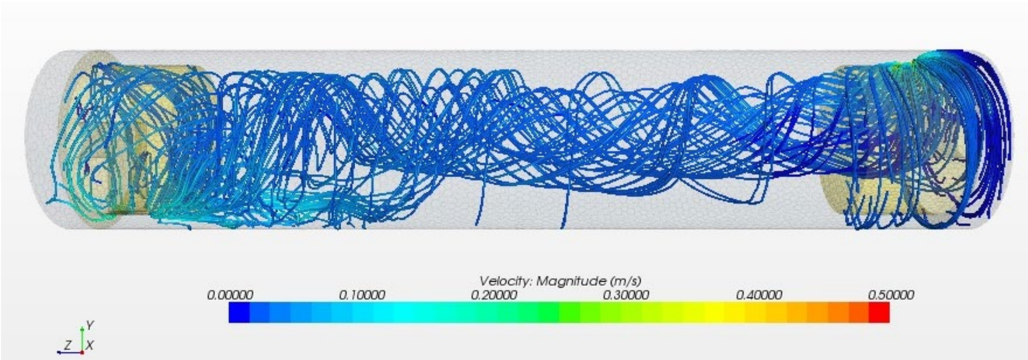

Figure 5. Flow field trajectory in reactor for 4th experiment in Taguchi OA.

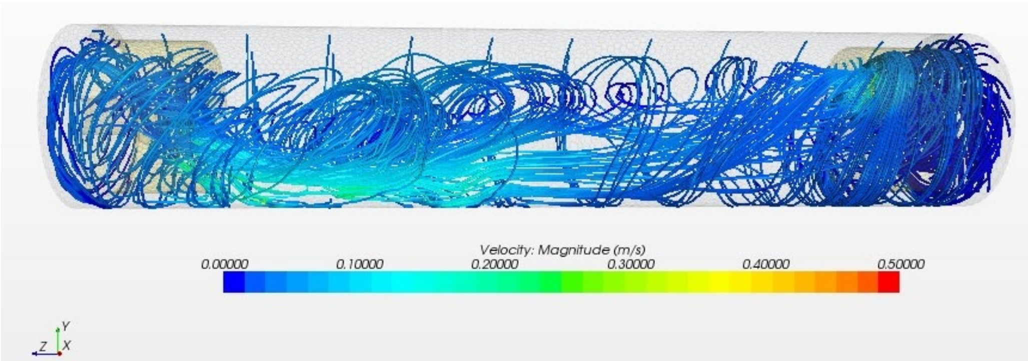

Figure 6. Flow field trajectory in reactor for 15th experiment in Taguchi OA.

Table 5. Comparison of average residence times in original rotary kiln and present study.

	Average Residence Times of CO_2 Gas
Original rotary kiln	63.03 s
The proposed cyclone-type rotary kiln	101.08 s

4.2. Carbonation Experiments

The validity of the CFD simulation results was confirmed experimentally using a prototype cyclone-type rotary kiln (see the schematic illustration shown in Figure 1 and the photograph presented in Figure 7). The experimental setup consisted of three major components, namely the reactor, a high temperature vaporizer, and a reactive gas control unit. The reactor was based on the original (non-cyclone) rotary kiln design proposed in [18] and had the dimensions shown in Table 1. In accordance with the Taguchi results, the inlet tube was designed with the optimal geometry parameters of θ = 0° and L = 120 mm.

Figure 7. Photograph of prototype cyclone-type rotary kiln.

Table 6 compares the carbonation performance of the prototype cyclone-type rotary kiln with that of the original reactor. It is seen that the proposed reactor increases the CO_2 absorption per kilogram of BOF slag from 12.15 g to 16.00 g. In other words, compared to the original rotary kiln reactor, the proposed reactor improves the CO_2 absorption capacity by 31.7%.

Table 6. CO_2 absorption per kilogram of BOF slag in original rotary kiln and proposed cyclone-type rotary kiln.

	CO_2 Absorption per Kilogram of BOF Slag (g/kg)
Original rotary kiln	12.15
The proposed cyclone-type rotary kiln	16.00

5. Conclusions

The present study proposes a novel rotary kiln reactor. The diameter of the sealed bearing of the reactor is just 60 mm and the end-sections of the inlet and outlet tubes are bent in such a way as to lengthen the flow path of the cyclonic airflow within the reactor; thereby increasing the residence time of the CO_2. The optimum inclination angle (θ) and length (L) of the inlet tube have been determined using the Taguchi robust design method. The feasibility of the proposed rotary kiln reactor has been demonstrated both numerically and experimentally. The key findings of the present study can be summarized:

1. The Taguchi results have shown that the maximum CO_2 residence time is achieved when using an inclination angle (θ) of 0° (i.e., parallel to the vertical line) and a bent tube length (L) of 120 mm.
2. The simulation results have shown that the CO_2 gas residence time in the optimized cyclone-type rotary kiln (i.e., 101.08 s) is 38.05 s longer than that in the original (non-cyclonic) rotary kiln reactor (i.e., 63.03 s). In other words, the proposed design increases the carbonation reaction time by 60.4%.
3. The experimental results have shown that the proposed cyclone-type rotary kiln achieves carbon dioxide absorption per kilogram of BOF slag of 16.00 g; representing an improvement of 31.7% over that of the original rotary kiln reactor, i.e., 12.15 g.
4. In 2013, the annual production of BOF slag in Taiwan was about 1.5 million tons [18]. Based on the annual production of BOF slag, it is estimated that this study has a CO_2 absorption potential of 24,000 tons/year in Taiwan.

Author Contributions: M.-S.K. contributed to perform the carbonation experiments; T.-B.C. contributed to conceive and design the experiments, and write the paper; C.-Y.L. contributed to perform the CFD simulations and write the paper; J.-W.H. contributed to perform the CFD simulations; C.-F.L. contributed to perform the CFD simulations. All authors have read and agreed to the published version of the manuscript.

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