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Estimating the dimensions of integrated calciner and carbonator for calcium looping process in a 7500 TPD capacity of cement plant

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Abstract

The calciner in cement factories plays a crucial role, particularly in the decomposition of calcium carbonate ($CaCO_3$) as primary raw materials into calcium oxide (CaO) and carbon dioxide (CO_2), a significant contributor to greenhouse gas (GHG) emissions. Hence, an integrated system has been proposed, combining conventional cement plants with calcium looping (CaL) cycles to reduce CO_2 emissions. CaL facilitates the capture of CO_2 by CaO, forming $CaCO_3$ as raw material for cement production. Given that CaL effectively reduces CO_2 emissions, the integration process with conventional cement plants requires careful consideration, particularly regarding raw materials, calciners, and carbonators. Integration parameters for CaL in raw materials include average diameter and logarithmic temperature difference. At the same time, calciners and carbonators encompass heat transfer coefficient (U), calciner dimensions, carbonation factor, and mass balance post-integration with CaL. These parameters will be calculated to facilitate the integration of the CaL cycle with conventional cement plants. In this study, based on raw materials with an average diameter of 3.28 μm and the mean heat transfer coefficient between hot gas and raw materials of 4 W/m^2 K, the calculated dimensions for the calciner are 9.6 m in diameter and 25 m in height. Since the plant studied has two preheater strings, two carbonator units are also required. The size of each carbonator is 4.75 m in diameter with a length of about 40 m, so it has a total volume approximately equal to the volume of the calciner to provide a longer residence time for particles.

Keywords: calcium looping (CaL) cycles; raw materials; cement calciner; cement carbonator; CO_2 capture; integrated system.

I. Introduction

Globally, cement is one of the largest energy consumers, making up approximately 7 % of the world's total energy consumption [1][2][3]. This makes sense because cement is an essential material [4] in infrastructure, such as construction or building and civil engineering projects [5]. The main energy demands in cement production and the limestone calcination process [6] that emits carbon dioxide gas

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indicate the cement factories as major CO_2 producers [7], contributing to greenhouse gas (GHG) effects [8][9][10]. Therefore, tackling this issue within cement production is necessary. One promising method is to capture CO_2 [11], and for cement plants, this promising method is calcium looping (CaL) [12]. In the calcium looping process, the CO_2 release by the calciner is due to the calcination process of calcium carbonate ($CaCO_3$) and is captured by calcium oxide (CaO) in the carbonator [13]. Current calcium looping application methods for CO_2 capture in cement plants include tail-end and integrated calcium looping [14]. Cement production generates CO_2 emissions of approximately one ton of CO_2 per ton of cement [15], with world production expected to reach four million tons per year [16]. While the indispensability of cement in human life is unquestionable, the challenge lies in mitigating CO_2 emissions from cement production and fuel combustion [17]. The challenge in the cement industry could be addressed by integrating the calciner and carbonator as a calcium looping method for cement plants. However, in determining the dimensions of equipment, research on the transportation of raw materials, including heat transfer with heating gas, needs to be carried out carefully. The transportation of raw materials in cement production has been examined by [18]. In that research, calciner dimensions were calculated and simulated, yet CO_2 emissions produced in cement plants were not integrated into the calculation to obtain adequate calciner dimensions.

The application of blended cement in cement factories, alongside a review of modern technologies for CO_2 capture, such as amine scrubbing, oxy-firing, and calcium looping, was also already carried out. However, this study focused solely on the theory of each modern technology without performing calculations for their application [19]. The strategies to reduce CO_2 emissions were also examined, including implementing energy-saving measures such as using energy-efficient equipment and transitioning the cement production process from wet to dry, as well as substituting conventional fuels with waste drive fuel (WDF). However, the study puts more emphasis on the examination of constraints in terms of technology and finance [20]. The study on three CO_2 capturing technologies, namely monoethanolamine (MEA), post-combustion calcium looping (CaL), and oxyfuel based on techno-economic assessment (TEA) and life cycle assessment (LEA), were examined. The results showed that CaL is one of the best technologies for reducing CO_2 emissions. Of course, this CaL will be more suitable for application in the cement industry, considering that this technology uses even absorbents derived from the raw materials of cement itself [21]. Hence, this study complements each by focusing on detailed calculations of total raw materials requirement in a cement plant if integrated with the CO_2 capture process in the calcium looping system. Therefore, this study aims to integrate calcium looping (CaL) into the cement plant. Before that, the heat transfer coefficient must be evaluated to examine the calciner's and carbonator's dimensions in the calcium looping (CaL). This study also estimated the carbonation factor of CaO with an average particle diameter of 180 μm and initial mass data [22], with the known value of carbonation factor of 77 % for a 3.3 μm diameter CaO [23], and with the assumption the CO_2 capture linear to particle surface area. As a final result, the necessity of calcium oxide (CaO) can be determined.

II. Materials and Methods

A. Materials and equations

The initial data of the existing cement plant, which was used for estimating the dimensions of the calciner and carbonator for integrated calcium looping (CaL), can be found in Table 1.

The sieve testing method is employed to determine raw materials' average particle size (diameter) and the corresponding surface area based on the grouped sizes. Utilizing the number of particles per group and the area of each particle, the total surface area of particles, referred to as A_{tp}, from one kg of the sample (in m^2) is calculated using equation (1), where n_{pi} is the number of particles in the group, and A_{pi} is the surface area of one particle in a group.

$$A_{tp} = \sum_{1=1}^{1=k}\left(n_{pi} * A_{pi}\right) \tag{1}$$

After that, the calculation method serves as a comparison to the sieve testing method to validate the accuracy of the calculated equivalent diameter of raw materials and the corresponding surface area. Using the

Table 1.
Initial data of cement production [24][25].

No	Parameter	Unit
1	Clinker production (ton/h)	314.45
2	Fuel rate in calciner (kg fuel/kg clinker)	0.1415
3	Calcination energy (kcal/kg $CaCO_3$)	425
4	Combustion efficiency (%)	85
5	Temperature input calciner (°C)	40
6	Temperature output calciner (°C)	900
7	Net heating value (kcal/kg)	5500
8	Average velocity of gas flow in calciner (m/s)	7
9	Percentage of $CaCO_3$ in raw mix (%)	80
10	Fuel consumption (kg/kg clinker)	0.156

assumption that the particles of raw materials are in the form of spherical geometry, the surface area, A, for a given diameter is computed using equation (2), where n_i the number of particles d_{mi} is the center of the diameter, and the value π is 3.14.

$$A_{tp} = \sum\left(n_{pi} * \pi\, d_{mi}{}^{2}\right) \tag{2}$$

Then, the heat transfer coefficient is employed to calculate the total energy transferred in cement manufacturing calciners and to determine the height of the oxy-calciner that will be used in the calcium looping process. Oxy-calciner is a limestone calcination equipment that burns the required fuel using pure oxygen. Specifically, the heat transfer coefficient utilized for calculating the height of the calciner in calcium looping is the average heat transfer coefficient between gas and particles in the the interconnected looping cycle (ILC) and separated looping cycle (SLC). This coefficient is calculated using the equation derived from the substitution method in equation (3), where q is the rate of heat transfer, U is the heat transfer coefficient and ΔT_{LMTD} is a logarithmic mean temperature difference.

$$q = UA\Delta T_{LMTD} \tag{3}$$

The height of the calciner, $h_{calciner}$, used in the calcium looping process, is determined by considering the time spent on gas in the calciner and the average velocity of gas flows inside the calciner, as indicated in Table 1. This calciner height is computed using equation (4), where t_g the time spent by gas, v_{gas} the average velocity of gas flow in the calciner, and 1.2 is the value of the safety factor.

$$h_{calciner} = t_g \times 1.2 \times v_{gas} \tag{4}$$

On the other side, carbonation is the process of binding CO_2 by CaO in the calcium looping process. This process takes place inside the carbonator. The carbonation factor is defined as the mass fraction of CaO that can bind CO_2 compared to the total mass of CaO that flows into the carbonator. The calculation involves defining the initial mass of CaO flowing through the carbonator and determining the quantity of CaO particles that remain unreacted with CO_2. The carbonation factor, FK_m, is calculated using the equation (5), where $m_{1,CaO}$ is the initial mass of CaO flowing in the carbonator and Δm is the mass of CaO that does not react with CO_2.

$$FK_m = \frac{m_{1,CaO} - \Delta m}{m_{1,CaO}} \tag{5}$$

The diameter of CaO that reacts with CO_2 and the diameter of CaO that does not react with CO_2 have differences or depths. This depth is calculated using the equation (6), where h is the depth of reacted and unreacted CaO with CO_2, d is the diameter of reacted CaO with CO_2, and d_2 is the diameter of unreacted CaO with CO_2.

$$h = d - d_2 \tag{6}$$

The calculation of surface area per kg of particles requires data on the surface area of each CaO particle and the number of CaO particles. The total surface area of CaO particles, $A_{particle}$, was calculated using equation (7), which multiplies the surface area of CaO particles, A_{CaO}, by the amount of CaO, n_{CaO}.

$$A_{particle} = A_{CaO} \times n_{CaO} \tag{7}$$

Then, the amount of carbon dioxide in the carbonator, $CO_{2,carbonator}$, was determined by considering the total mass of CO_2, $m, CO_{2,total}$, generated from the calciner and the CO_2 capturing efficiency, $\%\ CO_2$. The calculation of carbon dioxide in the carbonator can be executed by using equation (8).

$$CO_{2,carbonator} = m, CO_{2,total} \times \%CO_2 \tag{8}$$

The quantity of calcium carbonate in the carbonator, $CaCO_{3,carbonator}$ can be calculated by taking into account the total CO_2 present in the carbonator, the CO_2 capturing efficiency, the oxy-CaL efficiency, and the molar mass of calcium carbonate, $Mr, CaCO_3$, and carbon dioxide Mr, CO_2. The calculation for the amount of calcium carbonate in the carbonator is represented by equation (9).

$$CaCO_{3,carbonator} = Mr, CaCO_3 \times \frac{CO_{2,carbonator}}{Mr, CO_2} \tag{9}$$

The number of raw materials that enter that oxy-CaL is given by equation (10), where $\%CaCO_3$ is $CaCO_3$'s CO_2 capture efficiency (80 %).

$$OxyCaL_{rm} = \frac{CaCO_{3,carbonator}}{\%CaCO_3} \tag{10}$$

The fresh feed consists of fresh feed from cyclones in strings A and B. The calculation for the amount of fresh feed A is represented by equation (11).

$$Ff_{1A} = Cyclone\ 1A_{rm} - Unsep_{rm,2A} - OxyCaL_{rm,carbA} - CO_{2,carbA} \tag{11}$$

The amount of fresh feed B can be calculated by equation (12).

$$Ff_{1B} = Cyclone\ 1B_{rm} - Unsep_{rm,2B} - OxyCaL_{rm,carbB} - CO_{2,carbB} \tag{12}$$

where Ff_{1A} is fresh feed for string A, Ff_{1B} is fresh feed for string B, $Cyclone\ 1A_{rm}$ is raw mix entering 1A cyclone, $Cyclone\ 1B_{rm}$ is raw mix entering 1B cyclone, $Unsep_{rm,2A}$ is unseparated raw mix from 2A cyclone, $Unsep_{rm,2B}$ is unseparated raw mix from 2B cyclone, $OxyCaL_{rm,carbA}$ is raw mix that entering the oxy-CaL from string A, $OxyCaL_{rm,carbB}$ is raw mix that entering the oxy-CaL from string B, $CO_{2,carbA}$ and $CO_{2,carbB}$ is the amount of CO_2 in carbonator A and B, respectively.

B. Methods

The existing cement plant consists of double strings, such as preheater type (A and B strings). Therefore, the calciners also consist of ILC or In-Line Calciner in the A string and SLC or Separated Line Calciner in the B string. String A and B represent pipes or flows, respectively. To integrate the existing cement plant with calcium looping (CaL), first, the sieve testing method that employs multiple aperture sizes to classify the diameter of raw materials and ascertain the mass percentage corresponding to each diameter size [26] was applied to examine the initial data on the particle size distributions, the total particle surface area and mass of raw materials obtained from several cement factories in Indonesia. These raw materials consist of 80 % limestone ($CaCO_3$) and 20 % mixture of silica sand, clay, and iron sand. The procedure for sieve testing starts with taking some sampling raw material from five cement plants in Indonesia, such as Tonasa I, Tonasa II, Gresik, Padang IV, and Padang V. This raw material will be sieved using a standard set of sieves of different aperture size to make size analysis. The mass of raw material that retained by a particular aperture size can be determined. This mass is defined as the unfiltered mass of raw material that can be used to determine the mass filtered or the mass for the determined aperture size. The mass filtered is determined by subtracting the initial mass that is assumed to be 100 % from the unfiltered mass of raw material. The output of this sieve testing method is the diameter size of the raw material and the mass fraction of the raw material at that diameter. Those mass fractions from sieve testing of raw material taken from five cement plants in Indonesia will be averaged, and one mass fraction for each diameter size will be used as a representative. Then, the heat transfer method was used to determine the heat transfer coefficient (U) [26] and log mean temperature difference (LMTD) [27], which are essential for calculating the dimensions of the calciner and carbonator. The ideal gas law [28] was also used to define the calciner's dimensions, incorporating the previously determined gas quantity. These dimensions encompass both the diameter and height of the calciner and the carbonator's dimensions are based on a similar volume to that of the calculated calciner. After that, the fresh feed material from the preheater in strings A and B of the cement plant will be calculated based on the mass balance by using conservation equations.

The calcium looping method was utilized in the cement industry, integrating two reactors: a calciner and a carbonator. The calcium looping calciner collected $CaCO_3$ from cyclones 3A and 3B. The calcination reaction in the calciner would obtain energy from coal combustion while utilizing full oxygen (oxy-calcination) provided by the hot gas generator at temperatures ranging from 850 to 950 degrees Celsius. The calcium looping carbonator used CaO as a sorbent to absorb CO_2. The carbonator obtained CaO from the calcium looping cyclone that developed during the calcination process in the oxy-calciner. The CO_2 absorbed by CaO was converted into $CaCO_3$, which is then directed to cyclones 1A and 1B. The carbonation reaction generates $CaCO_3$, which can be utilized to create cement. The integrated schematic diagram of the calcium looping apparatus and the study of the strings of the ILC-SLC cement plant are presented in Figure 1.

III. Results and Discussions

The sieve testing method is employed to calculate the total surface area for particles within the diameter range of 0.5 μm to 700 μm, yielding a total surface area of 675.63 m^2. This result is then compared to the surface area of particles calculated using equations that have been presented previously as validation for the sieve testing method. According to the equation-based calculation, the surface area for particles with an average diameter of 3.28 μm is also determined to be 675.63 m^2, which matches the result obtained from the sieve testing method. Consequently, the accuracy of the result is confirmed. Besides that, the heat transfer coefficient for clinker production, 314.54 tons/h, with a total surface area of particles of 675.63 m^2, is calculated using equation (3). The operating temperature in the calciner includes the inlet and outlet temperatures of hot gas, and separated kiln feed from the cyclone is used to calculate the logarithmic temperature difference, which is used to determine the heat transfer coefficient.

The calculation results show that the heat transfer coefficient for the ILC is 3.24 W/m^2 K, while for SLC is 4.75 W/m^2 K. So, the average heat transfer coefficient for both calciners is 4 W/m^2 K. The average heat transfer coefficient is utilized to compute the total heat and gas residence time in the calciner. These parameters will be used to determine the height of the calciner that will be used in the calcium looping process. Accordingly, with a diameter of 9.6 m, the height of the calciner for an average heat transfer coefficient of 4 W/m^2 K is 25 m, as illustrated in Figure 2. This height

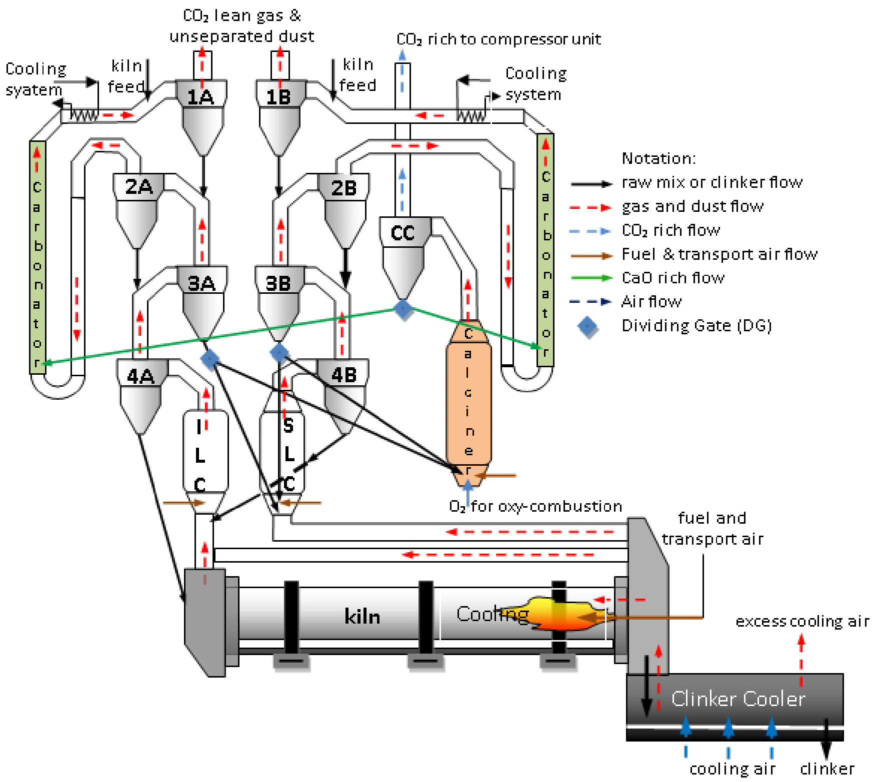

Figure 1. Schematic diagram of the configuration of calcium looping (CaL) with existing cement plant.

is considered the typical height of a calciner in a cement plant.

The calciner facilitates the decomposition of $CaCO_3$ into CaO and CO_2. Following the release of CO_2 in the calcination process, carbonation occurs in the carbonator, where CaO captures CO_2 from the cement plant, forming $CaCO_3$ as raw material for subsequent clinker production processes. However, it is essential to calculate the percentage of CO_2 captured in the carbonator to assess the effectiveness of the calcium looping cycle. This percentage is determined as the carbonation factor and evaluated using equation (5). The carbonation factor of CaO particles represents the percentage of CaO's capacity to capture CO_2 and produce $CaCO_3$. This study estimated the carbonation factor of CaO with an average particle diameter of 180 μm that was sourced from several locations in Indonesia, such as Bayah, Padang, Tuban, Cilacap, and Citereup with the initial mass data of CaO of 18 g with the CO_2 gas was supplied for a brief duration [22]. The result from those studies shows that the CaO captured the supplied CO_2 gas, causing an increase in the final mass of CaO. The increase in the final mass of CaO indicates that CO_2 gas has been captured, resulting in the formation of the final products $CaCO_3$ (limestone) and residual CaO from the reaction. The initial and final mass of CaO involved in the reaction were compared. Therefore, the mass of CaO that has reacted to form $CaCO_3$ can be determined. The carbonation factor is calculated as the ratio between the reacted CaO and the initial CaO. For the five CaO sources, the carbonation factor values were averaged to obtain a single value representing all five sources. This average value will serve as a reference for calculating carbonation factor values for other particle diameters. When expressed as a percentage of the volume of reacted particles, the carbonation factor also signifies

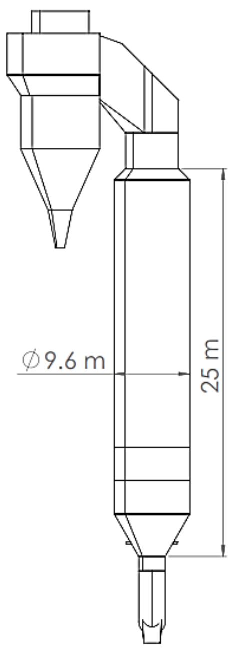

Figure 2. Dimension of calciner calcium looping.

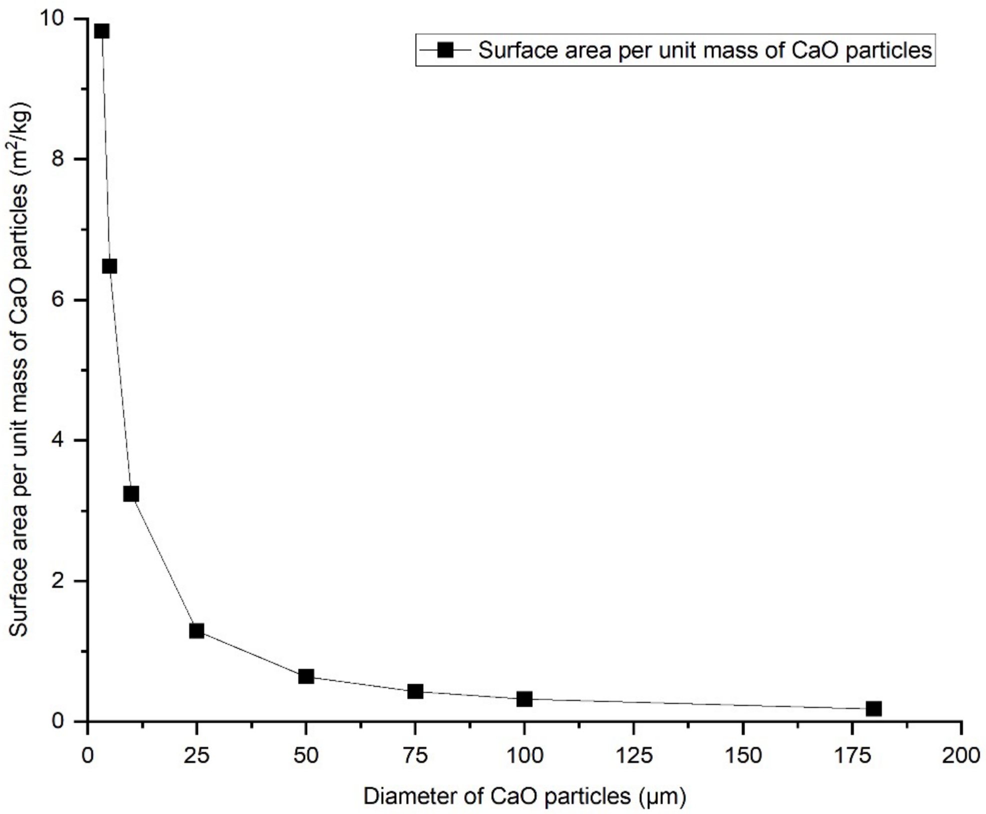

Figure 3. Diameter and surface area per unit mass of CaO particles.

the ratio between the volume of CaO that undergoes reaction and the initial volume of CaO.

The carbonation factor for 3.3 μm, 5 μm, 10 μm, 25 μm, 50 μm, 75 μm, 100 μm, and 180 μm in diameter is calculated by comparing the surface area per kg of particles between the 180 μm diameter CaO and those diameters, with a known value of carbonation factor of 77 % for a 3.3 μm diameter of CaO [23]. The result of this calculation for all CaO diameters is presented in Figure 3. The graph below shows that the surface area per unit mass of CaO particles, Ap, decreases with increasing particle size. This is because smaller particles have a bigger total surface area than larger particles, which means they have more surface area per unit mass. A larger particle surface area leads to increased conversion of CaO to $CaCO_3$.

After determining the surface area per unit mass of CaO particles, the carbonation factor can be calculated by comparing the surface area per unit mass of CaO particles and using the known carbonation factor values for particles with a diameter of 3.3 μm, which is 77 %, and for diameter of 180 μm is 42 %. The calculated carbonation factor for all observed particles can be seen in Figure 4. The graph shows that the diameter of the CaO particles increases, and the carbonation factor drops, reducing the particles' ability to collect CO_2. The shape of the particles is assumed to be spherical; a small particle diameter will have a high carbonation factor because the depth of the carbonation reaction is more effective than that of a larger particle diameter. The carbonation factor cannot be 100 % because, during the reaction, the CO_2 gas begins interacting with the CaO on the particle's outermost surface, forming the $CaCO_3$ compound. When all the pores in the CaO particles have become $CaCO_3$, the $CaCO_3$ compound layer will prohibit the CO_2 gas from reacting with the CaO within the particle.

In the CO_2 captured by CaO, if the entry of CO_2 reacting from the surface of CaO particles towards the interior occurs, then the unreacted CaO is assumed to be inside the particle. The shape of the particles is assumed to be spherical, and the depth of the reacted CaO portion, as well as the diameter of the unreacted CaO particles, can be evaluated. The difference in mass between the resulting $CaCO_3$ from the reaction and the mass of CaO that has absorbed CO_2 is used to calculate the remaining volume of unreacted CaO. Afterward, the diameter of CaO reacting with CO_2 and the diameter of CaO not reacting with CO_2 have differences in depth. Therefore, the depth is also calculated. These depth values were averaged to obtain a single value representing all five sources. The difference between the diameter of CaO that reacts with CO_2 and that does not react with CO_2 is called the depth of $CaCO_3$ from the CO_2 capture reaction. Based on the calculation, the average depth of the CO_2 capture reaction is 15.23 μm.

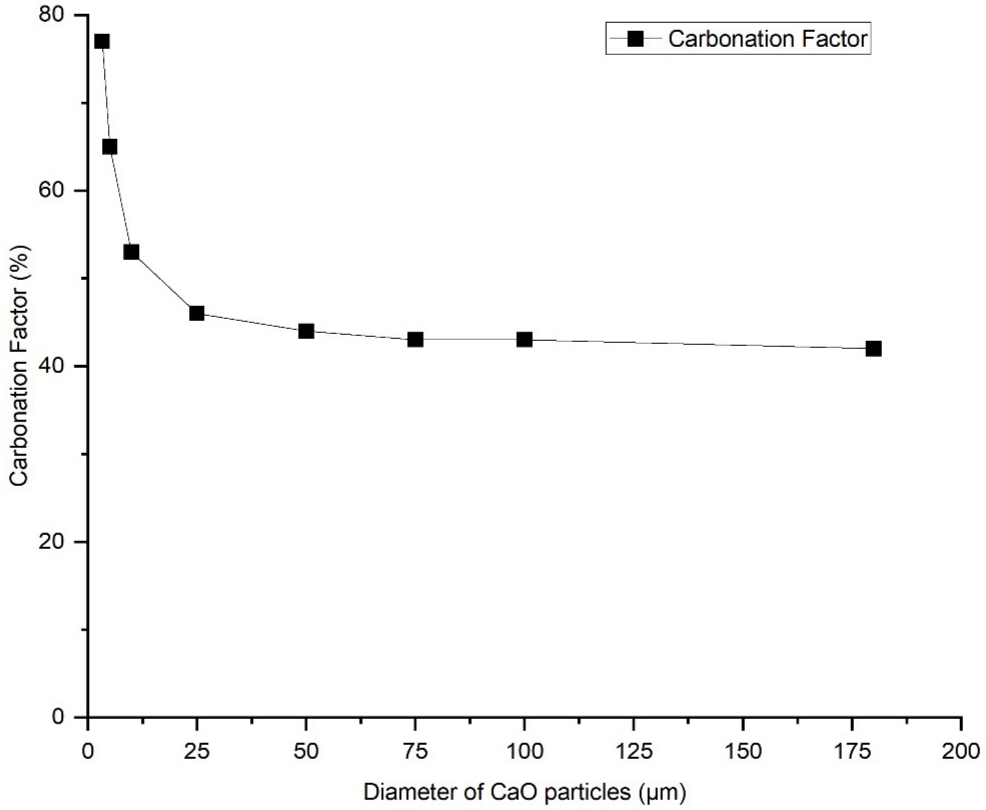

Figure 4. Diameter of CaO particles and carbonation factor.

The carbonator dimension design assumes that the carbonator's effective volume is close to that of the calciner for calcium looping. Meanwhile, the diameter of the carbonator adjusts to the dimensions of the upper cyclone gas to the cyclone below. The carbonator diameter is 4.75 m, which corresponds to the size of a gas line in the cement industry, as shown in Figure 5.

The carbon dioxide to be captured by a certain amount of CaO originates from the calcination process and fuel combustion of the cement manufacturing process. This CO_2 is calculated per unit mass of clinker produced by a cement plant (kg of clinker). Assuming the average CO_2 capture efficiency in the carbonator is known, and the percentage of CO_2 from the gas in the calciner to be captured in the carbonator is assumed to be 100 % or ideally captured all the CO_2 from the calciner, the CO_2 in the carbonator for clinker production of 314.54 tons/h is 0.58 kg/kg of clinker. With known CO_2 in the carbonator, the quantity of raw material required to capture CO_2 or the quantity of $CaCO_3$, is about 3.33 kg/kg of clinker. Since the quantity of raw material in the carbonator is known, the amount of raw mix entering the oxy-CaL is about 4.16 kg/kg of clinker.

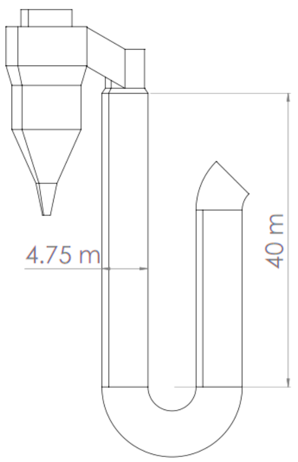

Figure 5. Dimension of carbonator.

With all the values obtained, the fresh feed for the integrated calcium looping system in the cement plant can be determined, that is, 1.37 kg/kg of clinker for string A and 1.45 kg/kg of clinker for string B.

IV. Conclusion

This research marks the initiation of calcium looping integration in cement factories, addressing the challenge of reducing CO_2 emissions in process industries. It presents the determination of calcium looping equipment dimensions, specifically the oxy-calciner and carbonator, designed to absorb carbon dioxide from cement plants. The study for raw materials with an average diameter of 3.28 μm with clinker production at a rate of 314.54 tons/h or 7550 tons/day yields several noteworthy conclusions, such as the average heat transfer coefficient between hot gas and raw materials is 4 W/m^2 K, the calculated dimensions for the calciner are 25 m in height and 9.6 m in diameter, the size of each carbonator is 4.75 m in diameter and 40 m in height. Therefore, it has a total volume approximately equal to the volume of the calciner to provide a longer residence time for particles. Also, the CO_2 to be captured in the carbonator is 0.5840 kg/kg of clinker, and the required $CaCO_3$ is about 3.335 kg/kg of clinker with a carbonation factor for CaO diameter of about 180 μm is 41.89 %, the amount of raw mix entering the oxy-CaL is about 4.1690 kg/kg of clinker, and the fresh feed for the integrated calcium looping system in the cement plant can be determined, that is, 1.3734 kg/kg of clinker for string A and 1.4496 kg/kg of clinker for string B.

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Declarations

Author contribution

T. Aulia: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation. **R.A. Prahmana**: Writing – review & editing, Formal analysis, Investigation, Data curation, Conceptualization, Supervision. **P.S. Darmanto**: Conceptualization, Resources, Supervision, Funding acquisition. **F.B. Juangsa**: Conceptualization, Resources, Supervision. **R.D.G.G. Permatasari**: Writing – review & editing, Software, Visualization. **K. Walad**: Writing – review & editing, Software.

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Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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