Theoretical Study on the Production of Environment-Friendly Recycled Cement Using Inorganic Construction Wastes as Secondary Materials in South Korea

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Received: 12 September 2018; Accepted: 21 November 2018; Published: 27 November 2018

Abstract: The cement industry endeavors to reduce CO2 emissions from cement manufacturing by utilizing industrial by-products as alternative fuels and developing secondary concrete products from construction wastes. With these efforts, the cement industry is attempting to become more eco-friendly and reduce environmental load. This study analyzed the possibility of using inorganic construction wastes to produce environmentally friendly recycled cement using the process of proportioning. To this end, the types and production trends of recyclable construction wastes and previous studies on the development of recycled cement using such construction wastes were analyzed. Based on this analysis, recyclable inorganic construction wastes were selected, and real waste was collected. The chemical composition of each inorganic construction waste was analyzed using X-ray fluorescence, and the composition of ordinary commercial cement was used as the baseline. After the collected inorganic construction wastes were mixed, they were fired using the Bogue formula. The mineral components of clinker, which was generated from the firing process, were predicted and analyzed. Waste gypsum board and ceiling materials were shown to contain large amounts of CaO, which could substitute limestone—a key component of cement. These results suggested that if the limestone content was greater than 85 wt %, mixing inorganic construction wastes in appropriate proportions could be used to develop various types of Portland cement.

Keywords: recycled cement; inorganic waste; construction waste; secondary materials

1. Introduction

The cement industry continues to have a large impact on industrial and economic development. Accordingly, significant efforts are being made to transform the cement industry into a sustainable industry from an environmental perspective [1–3]. However, despite these attempts, the construction industry still faces social problems, which are related to increasing quantities of wastes and treatment issues, and environmental challenges that arise as a result of resource depletion and global pollution caused by the greenhouse gases generated from the production of materials [4–6]. Cement production is a highly energy intensive production process. The energy consumption by the cement industry is estimated at about 2% of the global primary energy consumption, or almost 5% of the total global industrial energy consumption. China produces the most cement globally by a large margin, at an
estimated 2.4 billion metric tons in 2017, followed by India at 270 million metric tons in the same year, as shown in Table 1. It was also reported that China, India, the United States, and South Korea produce the largest quantities of cement globally [7–9].

### Table 1. Major countries in worldwide cement production (million metric tons).

| Major Countries | 2012  | 2013  | 2014  | 2015  | 2016  | 2017  |
|-----------------|-------|-------|-------|-------|-------|-------|
| China           | 2210  | 2420  | 2480  | 2350  | 2410  | 2400  |
| India           | 270   | 280   | 260   | 270   | 290   | 280   |
| United States   | 74.9  | 77.4  | 83.2  | 83.4  | 85.9  | 86.3  |
| Vietnam         | 60    | 58    | 60.5  | 61    | 70    | 78    |
| Turkey          | 63.9  | 71.3  | 75    | 77    | 77    | 77    |
| Indonesia       | 32    | 56    | 65    | 65    | 63    | 66    |
| Saudi Arabia    | 50    | 57    | 55    | 55    | 61    | 63    |
| South Korea     | 48    | 47.3  | 63.2  | 63    | 55    | 59    |
| Egypt           | 46.1  | 50    | 50    | 55    | 55    | 58    |
| Russia          | 61.5  | 66.4  | 68.4  | 69    | 56    | 58    |
| Iran            | 70    | 72    | 65    | 65    | 53    | 56    |
| Brazil          | 68.8  | 70    | 72    | 72    | 60    | 54    |
| Japan           | 51.3  | 57.4  | 53.8  | 55    | 56    | 53    |

In the field of the cement industry, eco-friendly materials are being actively developed in order to reduce the quantity of resources required as inputs and wastes generated as by-products [10–14]. In South Korea, a “Basic Plan for Recycling of Construction Wastes” was established to enhance the reuse of construction wastes. However, studies on recycling of construction wastes reported in the literature to date have focused predominantly on waste concrete [15,16]. Since waste concrete is highly available to improve the rate of recycling, recycled aggregate and cementitious powder have been highlighted. By contrast, only a few studies have dealt with secondary products arising from inorganic construction wastes, such as waste tiles, waste cement blocks, and waste gypsum board [17–20].

This study aimed to propose a theoretical proportioning for the development of eco-friendly recycled cement using inorganic construction wastes as secondary materials. In order to achieve this goal, this study analyzed different inorganic construction wastes that could substitute the existing raw materials utilized in the production of cement. Moreover, real inorganic construction wastes were collected, and their chemical composition was analyzed. A proportioning of eco-friendly recycled cement containing inorganic construction wastes was theoretically derived. Finally, the Bogue formula was used to predict chemical factors and to analyze the mineral components produced by clinker calcination.

### 2. Types and Trends of Construction Wastes

Construction wastes account for approximately 25–30% of all waste generated in the EU. This type of waste contains materials with high resource value such as metals, wood, glass, concrete, etc. Therefore, there is a high potential for recycling and material recovery of construction wastes which so far is under-exploited. The level of recycling varies significantly—from 10% to 90%—within the EU as shown in Figure 1 [21].

The potential to increase construction sector resource efficiency by increasing the construction waste recycling rate is significant. Construction wastes arise from activities such as the construction of buildings and civil infrastructure, total or partial demolition of buildings and civil infrastructure, and road planning and maintenance. However, the construction wastes produced from the construction sites provide only uneven qualities depending on the time and place. Due to social recognition on the quality issue as well as the quality degradation in application to concrete products, studies have been experimental rather than leading to field application. Furthermore, most studies have been conducted on waste concrete, for example, recycled aggregate and waste concrete fine powder, while further studies are required on construction materials that recycle various construction wastes [22–28].
Figure 1. The level of recycling and material recovery of construction wastes in the EU.

Figure 2 shows the yearly waste generation, which was based on statistical data from “A Current Status of National Waste Generation and Treatment” provided by the South Korean Ministry of Environment [29]. The examination of the total amount of waste generated in South Korea revealed that the percentages of municipal waste and general industrial waste increased gradually. Construction waste accounted for the largest portion of total waste, and its percentage reached 48.9% in 2015. In addition, as the term of reconstruction for row houses and apartments has been shortened from 40 years to 30 years, the number of reconstruction and remodeling works on buildings that were constructed in the 1990s is expected to increase. Accordingly, the amount and percentage of construction waste will most likely increase [30,31].

Figure 2. Current status of waste generation.

During the life cycle of a building, i.e., processes involving the design, construction, maintenance, and destruction of a real building, a large amount of construction waste is generated. These waste materials can be treated as follows. Initially, waste collection and transport licensees receive construction wastes from sites and classify the wastes in their own collection yard. Recyclable wastes
are sent to intermediate collection centers, local logistic centers, recycling centers, and other specialized facilities [32,33]. Subsequently, these wastes are recycled as secondary concrete products. Other wastes are sent to final treatment facilities, such as incineration plants or backfilling of construction waste.

Table 2 provides an overview of the classification of construction wastes to be treated, and reveals that waste concrete and asphalt occupy the largest shares. Various recyclable inorganic construction wastes are mostly classified as non-combustible wastes and construction waste materials.

Table 2. Classification and occurrences of different construction wastes.

| Waste Classification          | 2010  | 2011  | 2012  | 2013  | 2014  | 2015  |
|------------------------------|-------|-------|-------|-------|-------|-------|
| Construction waste material  |       |       |       |       |       |       |
| Concrete                     | 114,302 | 121,181 | 117,754 | 111,653 | 114,908 | 124,451 |
| Asphalt concrete             | 32,535  | 35,245  | 35,738  | 35,398  | 33,725  | 35,509  |
| Other (1)                    | 2132    | 2339    | 2957    | 3280    | 2393    | 3230    |
| Subtotal                     | 148,969 | 158,765 | 156,448 | 150,331 | 151,026 | 163,190 |
| Combustible waste            |       |       |       |       |       |       |
| Wood                         | 636    | 592    | 683    | 704    | 866    | 923    |
| Synthetic resin              | 839    | 1096   | 1261   | 1695   | 1586   | 1654   |
| Other (2)                    | 98     | 20     | 21     | 19     | 67     | 11     |
| Subtotal                     | 1573   | 1708   | 1964   | 2418   | 2519   | 2588   |
| Non-combustible waste        |       |       |       |       |       |       |
| Construction sludge          | 645    | 1403   | 644    | 1052   | 707    | 995    |
| Other (3)                    | 9      | 4      | 7      | 6      | 170    | 41     |
| Subtotal                     | 654    | 1407   | 651    | 1058   | 877    | 1036   |
| Construction soil debris     | 5347   | 4838   | 5904   | 5067   | 5863   | 7659   |
| Mixed construction waste (4) | 21,577 | 19,699 | 22,471 | 24,664 | 25,097 | 23,787 |
| Total (tons/day)             | 178,120 | 186,417 | 186,629 | 183,538 | 185,382 | 198,260 |

(1) Other construction waste material: waste brick, waste block, and waste roofing tile; (2) Other combustible waste: waste fiber and waste wallpaper; (3) Other non-combustible waste: waste metal, waste glass, waste tile, and waste ceramics; (4) Other mixed construction waste: mixed construction waste, waste board, and waste panel.

The non-combustible wastes and construction waste materials, which are discharged, are mixed and stored in arm roll boxes. When they are transported to collection yards, they are typically buried in landfills without accurate classification. Accordingly, the management and effective classification of these recyclable inorganic construction wastes would enable the establishment of eco-friendly construction and production systems, which would minimize construction wastes and maximize recycling rates [34,35].

3. Analysis of Inorganic Construction Wastes

3.1. Chemical Composition

Cement is an inorganic powder that is produced by combining pulverized limestone and clay. When combined with water, a chemical reaction occurs and the cement hardens. In this respect, cement is a critical construction material. As shown in Table 3, the representative chemical components of Portland cement are calcium oxide (CaO), silicon dioxide (SiO$_2$), alumina (Al$_2$O$_3$), and ferric oxide (Fe$_2$O$_3$). Depending on the mixing rates of chemical components, the mineral composition of cement after calcination can vary. Since the mixing rates change the properties of cement, it is very important to quantitatively assess the chemical compositions of materials before calcination and to match the final composition to that of conventional cement [36–38].

Table 3. The average chemical composition of Portland cement [39].

|     | CaO (wt %) | SiO$_2$ (wt %) | Al$_2$O$_3$ (wt %) | Fe$_2$O$_3$ (wt %) |
|-----|------------|----------------|-------------------|-------------------|
| Type I | 63.8       | 22.1           | 5.0               | 3.0               |
| Type II | 63.6      | 23.3           | 3.9               | 3.9               |
| Type III | 64.9      | 20.8           | 4.5               | 2.8               |
| Type IV | 63.0       | 25.9           | 3.0               | 2.8               |
| Type V | 65.0       | 22.4           | 3.4               | 4.4               |
Various recyclable inorganic construction wastes are mostly classified as non-combustible wastes and construction waste materials, as shown in Table 2. Non-combustible wastes and construction waste materials, such as tiles, glass, bricks, gypsum boards, and concrete powder, contain a large amount of CaO and SiO$_2$. We performed a literature review in order to collect chemical composition data of six inorganic construction wastes, as shown in Table 4. This review revealed that waste gypsum board contains a small amount of SiO$_2$ and a large amount of CaO, which represents the largest portion in cement. Accordingly, the waste gypsum board appeared to be the most useful substitute for (natural) limestone [40-44]. As for SiO$_2$, which represents the second largest component of cement, waste tiles, waste glass, and waste clay bricks were selected as potential substitutes. Lightweight foamed waste concrete and waste concrete powder were also shown to contain adequate ratios of both SiO$_2$ and CaO. However, these materials include many impurities that prevent their use as limestone and clay substitutes; these materials cannot be used for cement manufacturing without further processing. Generally, the process of construction waste grinding is as shown in Figure 3. Construction waste was selected to have no contaminants. Through a two-day drying operation, the moisture inside the construction waste was evaporated. Construction waste is crushed through jaw crusher and three-phase induction motor. Finally, the construction waste is finely grinded to 90 $\mu$m specimen size using a vibratory micro mill.

Table 4. Investigation of the chemical compositions of inorganic construction wastes though the existing literature review [40-44].

| No. | Construction Waste                  | SiO$_2$ (wt %) | Al$_2$O$_3$ (wt %) | Fe$_2$O$_3$ (wt %) | CaO (wt %) | MgO (wt %) | $\text{Na}_2\text{O}$ (wt %) | $\text{K}_2\text{O}$ (wt %) | SO$_3$ (wt %) | TiO$_2$ (wt %) | L.O.I* (wt %) | Total (wt %) |
|-----|------------------------------------|----------------|-------------------|-------------------|------------|------------|----------------|----------------|--------------|---------------|--------------|---------------|
| 1   | Waste tiles                         | 61.40          | 17.43             | 1.73              | 8.80       | 0.80       | 1.27           | -              | 0.36         | 6.61          | 99.41         |               |
| 2   | Waste glass                         | 71.00          | 1.47              | 0.07              | 8.91       | 0.04       | 13.10          | 0.24           | -            | -             | 99.66         |               |
| 3   | Waste bricks                        | 64.34          | 24.10             | 4.81              | 0.57       | 1.13       | 1.78           | 2.89           | -            | 1.09          | 99.71         |               |
| 4   | Waste autoclaved lightweight concrete | 48.30         | 3.69              | 1.88              | 28.10      | 1.59       | 0.26           | 0.62           | 1.59         | 13.30         | 99.40         |               |
| 5   | Waste gypsum boards                 | 1.60           | 0.69              | 0.22              | 54.32      | 0.14       | 0.46           | 0.23           | 41.47        | 0.49          | 99.58         |               |
| 6   | Waste concrete powder               | 45.50          | 11.90             | 1.90              | 29.80      | 1.90       | 3.00           | 1.40           | 2.30         | -             | 97.70         |               |

* L.O.I: Loss on ignition.

Figure 3. Process of construction waste grinding: (a) jaw crusher (crushing); (b) three-phase induction motor (grinding); (c) vibratory micro mill (fine grinding); (d) material dryer.
3.2. Theoretical Consideration of Chemical Components

As mentioned earlier, the four main chemical components of cement are chemically combined at approximately 1450 °C in a kiln, which results in new solid minerals. These resulting new minerals are $C_3S$ (3CaO·SiO$_2$), $C_2S$ (2CaO·SiO$_2$), $C_A$ (3CaO·Al$_2$O$_3$), and $C_{4AF}$ (4CaO·Al$_2$O$_3$·Fe$_2$O$_3$), and are termed as alite, belite, aluminate, and ferrite, respectively. As an investigation based on the process of proportioning, the present study considered the following chemical factors [41–43].

3.2.1. Bogue Formula

The mixing and burning of raw materials required to produce cement in a kiln at 1450 °C results in the generation of new solid minerals. The Bogue formula is conventionally used to predict the compounds of these clinker minerals. The Bogue formula was proposed in the 1920s by Robert Herman Bogue, and has been applied widely as the prediction formula for mineral composition of cement. The Korean standard on cement products (KS L 5201) specifies the application of the Bogue formula.

According to this standard, the chemical composition of cement is calculated depending on the content ratio of alumina and ferric oxide (Al$_2$O$_3$ (wt %)/Fe$_2$O$_3$ (wt %)) and are shown as follows [44,45].

\[
\begin{align*}
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 > 0.64 & \quad C_S = [4.071 \times \text{CaO (wt %)}] - [7.600 \times \text{SiO}_2 \text{ (wt %)}] - [6.718 \times \text{Al}_2\text{O}_3 \text{ (wt %)}] - [1.430 \times \text{Fe}_2\text{O}_3 \text{ (wt %)}] - [2.852 \times \text{SO}_3 \text{ (wt %)}] \\
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 > 0.64 & \quad C_S = [2.867 \times \text{SiO}_2 \text{ (wt %)}] - [0.7544 \times C_S \text{ (wt %)}] \\
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 > 0.64 & \quad C_A = [2.650 \times \text{Al}_2\text{O}_3 \text{ (wt %)}] - [1.692 \times \text{Fe}_2\text{O}_3 \text{ (wt %)}] \\
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 < 0.64 & \quad C_S = [4.071 \times \text{CaO (wt %)}] - [7.600 \times \text{SiO}_2 \text{ (wt %)}] - [4.479 \times \text{Al}_2\text{O}_3 \text{ (wt %)}] - [2.859 \times \text{Fe}_2\text{O}_3 \text{ (wt %)}] - [2.852 \times \text{SO}_3 \text{ (wt %)}] \\
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 < 0.64 & \quad C_S = [2.867 \times \text{SiO}_2 \text{ (wt %)}] - [0.7544 \times C_S \text{ (wt %)}] \\
\text{Al}_2\text{O}_3/\text{Fe}_2\text{O}_3 < 0.64 & \quad C_{4AF} = [2.100 \times \text{Al}_2\text{O}_3 \text{ (wt %)}] - [1.702 \times \text{Fe}_2\text{O}_3 \text{ (wt %)}] \\
\end{align*}
\]

The Bogue formula typically calculates the percentage of each cement mineral as follows: $C_3S$ (55 wt %), $C_2S$ (10 wt %), $C_A$ (10 wt %), and $C_{4AF}$ (10 wt %). The ASTM standard specifies the mean values of each mineral. However, the KS standard does not specify such values, and each manufacturer applies somewhat different criteria. In this study, we obtained and adopted reference values as shown in Table 5 from a real cement manufacturer [46].

| Portland Type | $C_3S$ (wt %) | $C_2S$ (wt %) | $C_A$ (wt %) | $C_{4AF}$ (wt %) |
|--------------|--------------|--------------|-------------|-----------------|
| Type I       | 52           | 24           | 9           | 9               |
| Type II      | 47           | 32           | 4           | 11              |
| Type III     | 62           | 14           | 9           | 8               |

3.2.2. Other Prediction Formulas and Chemical Factors

In addition to the percentage values of each cement mineral, properties such as lime saturation factor (LSF), silica modulus (SM), and iron modulus (IM) need to be considered for clinker manufacturing and management in cement and recycled cement production. LSF, SM, and IM formulas are shown as follows.

\[
\text{lime saturation factor (LSF)} = \frac{1.00 \text{CaO}}{2.8\text{SiO}_2 + 1.18\text{Al}_2\text{O}_3 + 0.65\text{Fe}_2\text{O}_3} \tag{7}
\]

LSF expresses the maximum quantity of CaO that can be combined with acidic components, such as SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$, during the normal burning and cooling of a clinker. The degree of burning of the clinker is indicated by the quantity of unreacted CaO, namely free lime [47].
If there is a small amount of free lime, it can be estimated that calcination was adequate. When LSF is low, even if the calcination in the kiln was adequate, the decrease in $C_3S$ can also reduce the initial strength. By contrast, when LSF is high, even if the calcination temperature or period is increased, calcination can be difficult and free lime may remain. However, the increase in $C_3S$ improves the initial strength, and LSF needs to be sufficiently high to fabricate cement with a high content of $C_3S$. The appropriate range of LSF is 0.92–0.96, and good-quality clinker contains 1.0 to 1.5 wt % of free lime [48].

\[
\text{silica modulus (SM)} = \frac{\text{SiO}_2}{\text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3}
\]  (8)

SM is an important value that affects clinker quality as well as the behavior of the mixture in the rotary kiln where clinker is calcinated. If SM increases, the mixture of materials becomes difficult to calcinate, which results in insufficient generation of lumps and causes powder to fly in the kiln. Moreover, as the calcination process is difficult, a higher temperature is required and more fuel is consumed, which makes it difficult to fabricate cement with stable quality. When cement erodes the kiln refractories, it contains a large amount of $C_2S$, which delays the strength development of cement. The optimal range of SM is from 2.3 to 2.8 [16,49].

\[
\text{iron modulus (IM)} = \frac{\text{Al}_2\text{O}_3}{\text{Fe}_2\text{O}_3}
\]  (9)

IM indicates the quantitative relationship between $\text{Al}_2\text{O}_3$ and $\text{Fe}_2\text{O}_3$. Any material with a low IM facilitates the generation of clinker, even at a low calcination temperature. In addition, if the value of IM is low, the $C_3A$ content decreases and $C_4AF$ content increases, thereby reducing the initial strength of cement. However, the hydration heat is lowered, and chemical resistance increases. By contrast, if a material compound has a high IM, it is difficult to calcinate, which increases fuel consumption, as in the case of material with high SM. In addition, since the clinker generated is hard, significant energy is needed to pulverize it, thereby raising the production cost [50,51].

The optimal range of IM is 1.6–2.0. Apart from LSF, SM, and IM, magnesium oxide (MgO), which is contained in materials, facilitates the melting process (from solid to liquid) or lowers the melting temperature, thereby promoting calcination. However, if the MgO content is too high, moisture expansion can occur when concrete hardens. For this reason, the KS standard specifies the upper limit of MgO as 5 wt %.

### 3.3. Collection and Chemical Analysis of Inorganic Construction Wastes

In this study, we analyzed the chemical compositions of inorganic construction wastes that had been deemed, based on the results of the literature review, as viable substitutes for the raw materials used in cement production. Based on the analysis, we visited real construction sites, intermediate- and final-stage treatment licensees, and recycling plants to collect six types of wastes, including tile, ceiling material, and cement block as shown in Figure 4. As 100% inorganic construction wastes did not appear to be fully available for the manufacturing of recycled cement, limestone and electric furnace slag were obtained from cement manufacturers and industrial by-product treatment facilities [52].

The chemical composition of each collected inorganic construction waste was analyzed using X-ray fluorescence analysis, as shown in Table 6.
Figure 4. Specimens of inorganic construction wastes: (a) waste concrete powder and 90 µm specimen; (b) waste tile and 90 µm specimen; (c) waste cement block and 90 µm specimen; (d) waste ceiling material and 90 µm specimen; (e) waste gypsum board and 90 µm specimen; (f) waste cement brick and 90 µm specimen; (g) waste brick and 90 µm specimen; (h) waste limestone and 90 µm specimen.

Table 6. The results of chemical composition analysis of inorganic construction wastes performed using X-ray fluorescence.

| No. | Construction Waste        | SiO$_2$ (wt %) | Al$_2$O$_3$ (wt %) | Fe$_2$O$_3$ (wt %) | CaO (wt %) | MgO (wt %) | Na$_2$O (wt %) | K$_2$O (wt %) | SO$_3$ (wt %) | TiO$_2$ (wt %) | Other (wt %) | L.O.I* (wt %) | Total (wt %) |
|-----|--------------------------|----------------|---------------------|---------------------|------------|------------|----------------|--------------|---------------|----------------|--------------|-------------|-------------|
| 1   | Waste ceiling material   | 5.84           | 1.10                | 1.06                | 39.21      | 0.54       | 0.00           | 0.32         | 24.62         | 0.41           | 0.52        | 26.39       | 100.00      |
| 2   | Waste gypsum board       | 0.91           | 0.32                | 0.30                | 36.42      | 0.22       | 0.30           | 0.12         | 39.29         | 0.03           | 0.13        | 21.96       | 100.00      |
| 3   | Waste concrete powder    | 50.69          | 10.01               | 3.93                | 19.56      | 1.26       | 0.67           | 1.26         | 3.69          | 0.99           | 0.46        | 0.63        | 8.12        | 100.00      |
| 4   | Waste cement block       | 55.46          | 10.28               | 3.42                | 22.35      | 1.16       | 0.92           | 1.02         | 2.23          | 0.44           | 0.72        | 1.32        | 8.22        | 100.00      |
| 5   | Waste tile               | 63.05          | 21.00               | 5.80                | 21.30      | 0.71       | 1.33           | 0.44         | 2.13          | 0.02           | 0.74        | 0.50        | 1.24        | 100.00      |
| 6   | Waste brick              | 16.19          | 11.22               | 36.32               | 23.10      | 2.71       | 0.00           | 0.07         | 0.21          | 0.84           | 0.46        | 0.00        | 10.01       | 100.00      |
| 7   | Electric furnace slag    | 12.21          | 2.40                | 0.77                | 44.75      | 2.15       | 0.01           | 0.64         | 0.00          | 0.00           | 0.00        | 36.55       | 99.48       |

* L.O.I: Loss on ignition.

The inorganic construction wastes that were analyzed in this study exhibited no significant differences in their chemical compositions from those collected in the previous studies [53, 54]. Each inorganic construction waste primarily contained the main chemical components of cement, including CaO, SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$. Waste ceiling material and waste gypsum board appeared...
to be suitable substitutes for CaO in the manufacturing of cement, which was in agreement with the results reported in previous studies. Waste tile and waste clay brick were found to be viable substitutes for silicate materials. However, SiO$_2$ and Al$_2$O$_3$ accounted for over 80 wt % of waste brick, while the content of CaO was very small. This composition could result in cement with hydraulic properties. Moreover, the waste brick included a higher proportion of alkalins, such as Na$_2$O and K$_2$O, than other materials; thus, this type of waste could only be used as a cement material in a limited ratio. Although waste cement and concrete powder contain only a small amount of CaO, they contain a large amount of SiO$_2$. The large amount of SiO$_2$ indicates a high content of sand; consequently, only a small amount of waste cement and concrete could be used to adjust the material composition.

3.4. Analyses of Inorganic Construction Wastes and Theoretical Combinations

As shown in Table 7, this study combined various inorganic construction wastes with a focus on achieving optimum contents of CaO and C$_2$S, which are closely related to the cement strength and are major influential factors in the generation of calcium silicate compounds, such as C$_3$S and C$_2$S. The goal of this study was to develop eco-friendly, recycled cement by combination of inorganic construction wastes in appropriate ratios. Consequently, we examined theoretical combinations of inorganic construction wastes in which the ratios of limestone were set to 75, 80, and 85 wt % in order to establish how much limestone containing a significant proportion of CaCO$_3$ could be theoretically reduced so as to decrease the CO$_2$ emissions generated by decarboxylation. In comparison with limestone (natural resource), waste ceiling material and waste gypsum board were shown to contain similar levels of CaO.

Table 7. The combination of inorganic construction waste.

| No. | Construction Waste     | Combination I | Combination II | Combination III |
|-----|------------------------|---------------|----------------|-----------------|
| 1   | Waste tile (wt %)      | 4.1           | 4.4            | 4.8             |
| 2   | Waste cement block (wt %) | 0.7          | 0.5            | 0.3             |
| 3   | Waste ceiling material (wt %) | 17.5       | 12.4           | 7.2             |
| 4   | Limestone (wt %)       | 75.0          | 80.0           | 85.0            |
| 5   | Electric furnace slag (wt %) | 2.7          | 2.7            | 2.7             |
|     | Total (wt %)           | 100.0         | 100.0          | 100.0           |

However, these materials contained considerably more sulfur trioxide (SO$_3$) than other inorganic construction wastes. This issue had to be considered as a variable when the mineral composition was predicted using the Bogue formula. Accordingly, the substitution of limestone with waste ceiling material and waste gypsum board was below our expectations. Waste cement block and waste concrete powder contained a small amount of CaO, as demonstrated in previous studies, but contained a significant amount of SiO$_2$. For this reason, these materials could not be utilized in the theoretical combinations. The waste tile and waste clay brick were both expected to possess a large amount of silicate materials. However, in the present study, the waste tile was applied in the theoretical combination since it contained more CaO and less SiO$_2$. When we performed the simulations using the four major minerals (C$_3$S, C$_2$S, C$_3$A, and C$_4$AF), no issues occurred despite the addition of an industrial by-product, such as electric furnace slag. However, the values of LSF, SM, and IM, which are conventionally used in cement manufacturing and management, were predicted to exceed their optimal ranges. Accordingly, the use of electric furnace slag, which contains a large amount of ferric dioxide (Fe$_2$O$_3$), as a composition modifier had to be considered in order to lower LSF, SM, and IM. When about 2.7 wt % of electric furnace slag was added to the mixture, the values of the target parameters stabilized. Consequently, slag was included at a fixed value in each combination.
4. Predictive Analysis of Clinker Calcination

4.1. Analysis of Clinker Minerals Using the Bogue Formula

The present study combined different inorganic construction wastes by applying the mineral composition values specified for ordinary Portland cement, which are widely accepted by South Korean cement manufacturers. The mineral composition of clinker, which would be produced by a real calcination process using the mixing ratios, was predicted and analyzed using the Bogue formula, as presented in Figure 5 which illustrates the results of the simulations. As for C$_3$S, which is the most influential factor when it comes to initial strength, Combination III (51.27 wt %) produced the most optimal prediction.

Figure 5. The predicted clinker mineral component compositions.

The remaining two combinations (Combination I: 48.38 wt %, Combination II: 50.23 wt %) did not satisfy the reference value for C$_3$S (52 wt %). C$_2$S makes a more significant contribution to long-term strength than initial strength, and has both low reactivity and low calories. All three simulated combinations failed to reach the reference value of C$_2$S (Combination I: 20.15 wt %, Combination II: 20.82 wt %, Combination III: 22.34 wt %). In addition, the reference value of C$_3$A, which is highly reactive and has very high calories, was not satisfied in any of the three combinations (Combination I: 7.15 wt %, Combination II: 7.61 wt %, Combination III: 8.12 wt %). By contrast, C$_4$AF was predicted to exceed its reference value. As for LSF and IM, Combination III (LSF: 0.94, IM: 1.74) produced the best prediction when compared to the reference values. As for SM, Combination I (SM: 2.7) exhibited the best outcome. Overall, Combination III afforded the best results in terms of approximating the ordinary Portland cement, which was taken as the standard, because it predicted the highest levels of C$_3$S and C$_2$S, which affect the initial and long-term strength, respectively. Combination II was ranked as second, with Combination I coming last.

4.2. Analysis and Discussion of the Mineral Composition Predictions

The mineral composition of clinker, which is produced by calcination, was predicted and analyzed by applying the Bogue formula. The results revealed that, unless the limestone content of cement was greater than 85 wt %, the reference values of mineral composition required by the cement manufacturers could not be achieved in the theoretical simulations. In other words, it was necessary to increase the percentage of natural limestone or inorganic construction waste containing some amount of CaO in order to obtain satisfactory mixing ratios of C$_3$S and C$_2$S. This study selected waste ceiling material and waste gypsum board as substitutes for limestone. However, as mentioned earlier, although these...
waste materials contained almost the same level of CaO as limestone, they also included excessive SO$_3$. For this reason, the Bogue formula could not predict a sufficient value of C$_3$S. Consequently, the SO$_3$ issue needs to be examined in more depth in future studies on recycled cement. If SO$_3$ is removed from these materials by further processing and the content of CaO is increased, the applicability of the waste ceiling material and waste gypsum board to the development of recycled cement would increase significantly.

4.3. Effects Expected by Developing Recycled Cement

As reported in previous studies, inorganic construction wastes contain the main chemical components of cement, and they can be good substitutes for the raw materials used in cement production. However, each inorganic construction waste is produced at a different time and place using different methods. Accordingly, these waste materials do not exhibit a uniform chemical composition, and often contain many impurities. Although recycled cement is already being produced, such impurities will become an obstacle to achieving uniform product quality and may make quality management challenging.

To address this issue, the management system for all the related stakeholders involved in the procedure, ranging from the construction sites, which discharge wastes, to collection yards and the transport stage, needs to be improved. Such improvements could include a new manual and incentive system that would promote the effective classification of construction wastes and subsequent disposal based on their properties. If construction wastes are classified more effectively and their collection is more rapid, it will be possible to secure uniform cement substitutes more predictably. Moreover, when relevant techniques are developed to utilize construction waste materials, it will be possible to produce and develop a variety of secondary construction products in addition to recycled cement in the construction material industry.

5. Conclusions

The present study examined the theoretical combinations of various inorganic construction wastes as a means of developing eco-friendly recycled cement. The analysis revealed that the inorganic construction wastes, which were identified by the literature review as suitable substitutes for the raw materials used in cement production and collected in this study, had similar chemical compositions to those reported in previous studies. For limestone as the main raw material of cement as well as other minerals as subsidiary raw materials, the selected inorganic construction wastes were substituted by 25%, 20%, and 15%, which were defined as Combination I, Combination II, and Combination III, respectively.

The inorganic construction wastes were collected based on the contents specified for ordinary Portland cement. The chemical compositions of the waste materials were put into the Bogue formula to predict the generation of minerals after clinker calcination. The results of these predictions showed that if the limestone content was greater than 85 wt %, a new type of cement with the same quality level as that of Portland cement could be developed. The analysis also revealed that waste ceiling material and waste gypsum board contain significant amounts of SO$_3$; if this amount could be reduced through further processing, these waste materials could substitute limestone in the manufacturing of recycled cement. Because most of the examined inorganic construction wastes contained large quantities of SiO$_2$, many new types of cement could be developed in addition to the ordinary Portland cement.

Consequently, the present study confirmed that inorganic construction wastes contain several major chemical components of cement. We utilized the Bogue formula to examine potential combinations of these wastes, and used the chemical composition of ordinary Portland cement as a reference. This examination showed that the development of recycled cement is theoretically possible. However, as this study was theoretical, further experimental studies including physical and chemical considerations need to be performed to advance the development of recycled cement and secondary construction products from inorganic construction wastes.
Author Contributions: All authors contributed substantially to all aspects of this article.

Acknowledgments: This research was supported by a grant (Code 17CTAP-C129766-01) from the Construction Technology Research Program (CTIP) funded by the Ministry of Land, Infrastructure and Transport. And this work was supported by the research fund of Hanyang University (HY-2017).

Conflicts of Interest: This manuscript has not been published or presented elsewhere in part or in entirety and is not under consideration by another journal. We have read and understood your journal’s policies, and we believe that neither the manuscript nor the study violates any of these. There are no conflicts of interest to declare. The authors declare no conflict of interest.

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