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Study on CO_2 Emission Reduction Effects of Using Waste Cementitious Powder as an Alternative Raw Material

Park, Dong-Cheon[1] Kwon, Eun-Hee[1] Hwang, Jong-uk[1] Ahn, Jae-Cheol[2*]

Department of Architectural Engineering, Korea Maritime University, Youngdo-Gu, Busan, 606-791, Korea [1]
Department of Architecture, Dong-A University, Saha-Gu, Busan, 604-714, Korea [2]

Abstract

With environmental regulations continuously being strengthened internationally the need to control environmental pollution and environmental load is emerging in Korea. The purpose of this study is to seek methods or using waste cementitious powder as an alternative raw material for limestone through the optimization of raw material and to quantitatively analyze the resulting reduction of CO_2 emission in order to contribute to solving the issue of waste, which is the biggest issue in relation to construction and global warming. The results of the study, show that waste cementitious powder can be used as an alternative raw material for limestone at OPC level, but it was also found that mixing fine aggregate cementitious powder into waste cementitious powder significantly affected the substitution rate for limestone with waste cementitious powder and the reduction of greenhouse gas. In particular, when fine aggregate cementitious powder was used at a rate of 0~20%, the substitution rate for limestone and the reduction in the rate of greenhouse gas emission was significantly reduced. It is thought that a technique to efficiently separate and discharge the fine aggregate cementitious powder mixed in waste cementitious powder needs to be developed in the future.

Keywords : cementitous powder of waste concrete, fine aggregate additional ratio, CO_2 emission

1. Introduction

1.1 Background of a study

Internationally, environmental regulations are being strengthened with the goal of sustainable development. In major developed countries, specific studies have been conducted on minimization of pollution discharge, recycling and resource recovery of waste and improvement of eco-friendly processes. In Korea, issues related to environmental pollution and environmental load are emerging. Among them, the biggest issue related to construction is waste and global warming. Construction waste accounts for 51.7% of all industrial waste, and concrete waste and asphalt waste account for 87.1%. This makes them a key target for material recycling, and various approaches have been pursued[1]. Greenhouse gas emission is the most significant factor affecting global warming. Greenhouse gas emission by the cement industry accounts for 18.9% of total emissions. As the second largest industry after the steel industry in terms of emissions, it has been receiving attention. Emissions have tended to continuously increase with the development of the cement industry. For this reason, various studies have been conducted to find approaches to using recycled concrete. Concrete recycling has now reached the practical stage, in which it is being used for low-added value materials such as recycled aggregates and sub base course materials. This study

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* Corresponding author: Ahn, Jae-Cheol
[Tel: 82-10-9313-9232, E-mail: jcan222@nate.com]

is very useful from the perspective of the reuse of aggregate resource which is lacking, because the chunks of concrete generated by reconstruction are crushed and are used as aggregates and rubble. Moreover, when reduction of cost for waste treatment, conservation of natural resource and the impact on the natural environment caused by indiscriminate quarrying are considered, concrete recycling could be a measure that is beneficial for the protection of earth's environment.

Thus, the purpose of this study is to use the paste ingredients contained in waste concrete cementitious powder (hereinafter, waste cementitious powder) focusing on clinker production as an alternative raw material for limestone, and analyze the reduction effect achieved through conservation of raw materials for limestone and estimates of emission of greenhouse gas

1.2 Theoretical overviews

1.2.1 Research trend of CO_2 reduction technology in the cement industry

Measures for CO_2 reduction in the cement industry can largely be divided into four types, which are utilization of alternative fuels, introduction of high efficiency equipment for improved energy efficiency, increased use of blended cements and increased use of additives and admixtures. Increased usage of additives and admixtures is considered the most effective technology for CO_2 reduction. The reason for this is that measures of using alternative fuels and CO_2 reduction through high efficiency can reduce the CO_2 generated by combustion and power except that related to decarbonation by up to 10%.

1) Domestic research trend of CO_2 reduction technology

To reduce the energy used for manufacturing cement, Japan is promoting the supply of high efficiency equipment, including the installation of reserve grinders, introduction of high performance classifiers and expansion of usage of energy alternative sources. The British government is presenting a vision to reduce CO_2 emission by 60% over 60 years in the same manner as Japan through step-by-step short term (1990 - 2010), mid-term (2010 - 2030) and long-term (2030 - 2050) plans. In addition, the American Association of Cement has reported that 45 companies and 101 factories have established a voluntary long-term goal of reducing waste by 10% in 2020 compared to 1990 levels through the development of a CMS (Cement Manufacturing Sustainability) program, and this was achieved in 2009[2].

2) International research trend of CO_2 reduction technology

In the report ‘research on measures to cope with climate change conventions,’ it was reported that the average total consumption calories required for domestic cement manufacturing was 780.1kcal/kg · cement. It was found that among the main energy sources of cement manufacturing processes such as heat and electricity, supply heat was 690kcal/kg · cement and power was 90.1kcal/kg · cement. This was lower than the average value of 841kcal/kg · cement in Japan by approximately 7%. The proportion of installation of the latest NSP Kilns in Korea was 85%, which is higher than Japan's adoption rate of 83%. This implies that domestic cement manufacturing facilities are almost the same as those in developed countries in terms of energy efficiency and performance. As such, it is expected that measures to introduce a high efficiency facility will achieve a relatively lower reduction effect. On the other hand, the method to reduce CO_2 emission in the calcination process, which represents the biggest share of CO_2 emission in the cement manufacturing process, is the use of mixed cement, which can reduce the production of cement clinker. In Korea, only approximately 20% of cement is produced as mixed cement, and thus the expansion of the use of mixed cement is required. In addition, research has been conducted on recovery of the hydration of recycled cement based on calcination at low temperature. It has been reported that high compressive strength of 40MPa or higher is represented under the condition of calcination at low temperature of approximately 700℃[3]. It is thought that this may affect the reduction of CO_2 emission related to combustion and power, which accounts for about 40% of the entire calcination process.

1.2.2 CO_2 emission mechanism

The stage of the cement manufacturing process responsible for the largest share of CO_2 emission is the calcination process (83.1%), with other stages being responsible for emissions in the following amounts: mining (Aggregate: 1.2%, limestone 0.3%), grinding (1.4%), mixing (5.4%), and transport (4.3%)[4]. In this paper, the CO_2 emissions from the calcination process, which represent the majority of total CO_2 emission, were investigated.

CO_2 emission sources in the calcination process, which represents the largest share of greenhouse gas emission in the cement manufacturing process, can be largely divided into decarbonation of limestone, combustion of fossil fuels to maintain the kiln temperature required for calcination constant and power usage in the facility. Each CO_2 source is independent. According to previous studies, as shown in Figure. 1, the average CO_2 generated to manufacture 1 ton of cement was 822 kg-CO_2 based on raw fuel and electricity usage added through the manufacturing by each cement manufacturer A-F. In particular, decarbonation of limestone generated 555 kg-CO_2, 67.5% of CO_2 emission, the highest proportion. CO_2 emission caused by combustion was 219 kg-CO_2, which was 26.7% of all CO_2 emission. CO_2 emission caused by power usage was 48kg-CO_2, which accounted for 5.8% of all CO_2 emission[5]. However, according to some studies, CO_2 emission caused by decarbonation accounted for 52.15% of all emission and CO_2 emission caused by combustion and power usage accounted for 47.85%, a fairly significant difference that is accounted for by differences in the heat efficiency of fuel and the water content of raw materials[6]. In this study, the average of each ratio was used as follows: CO_2 emission caused by decarbonation, combustion and power was 59.83%, 32.9% and 7.2%, respectively. This is shown in Figure 2. Here, the decarbonation process of limestone, which accounted for the highest proportion of CO_2 emission during the cement manufacturing process, was shown in equation (1). As limestone ($CaCO_3$) is decomposed into lime (CaO) at high temperature, CO_2 is discharged. Thus, the CaO required for production with the main compounds of cement is estimated according to equation (2). If hydroxyl calcium ($Ca(OH)_2$), which is a hydrated product in waste cementitious powder, is substituted through a re-decomposition process, a significant amount of CO_2 emission can be reduced.

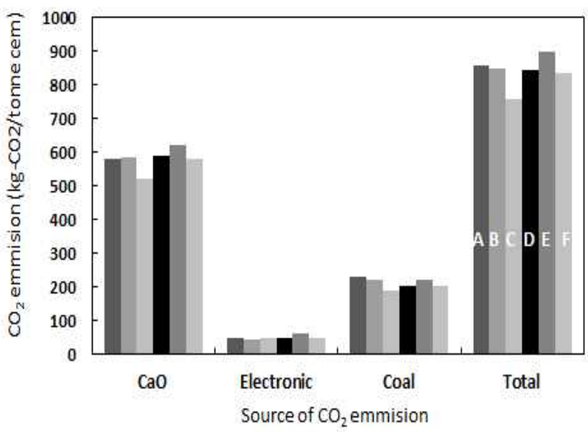

Figure. 1 CO_2 emission arising from the energy source and material

Figure. 2 CO_2 emission ratio from the energy source and material

$CaCO_3$(100g/mole) + Heat →
CaO(56g/mole) + CO_2 (44g/mole)------- (1)

$Ca(OH)_2$ (76g/mole)+Heat →
CaO(56g/mole) +H_2O(20g/mole)--------- (2)

1.3 Fine aggregate added in waste cementitious powder

In the author's previous research, the chemical composition of waste cementitious powder actually collected from 11 sites nationwide was analyzed, and the addition ratio of fine aggregate was approximately 65%. Contents of each ingredient were different from those in OPC. CaO and Fe_2O_3 were approximately 43% and 0.6% less, respectively. SiO_2 and Al_2O_3 were approximately 30% and 5% more, respectively. It was estimated that this was due to the addition of fine aggregate powder, which was difficult separate from cementitious powder because of its similar grain size and grain form. According to previous papers, major components were α-quartz, feldspar and calcite. α-quartz and feldspar added in cementitious powder and calcite added in coarse and fine aggregates in waste concrete were formed through grinding and carbonation reaction of $Ca(OH)_2$ formed by hydration reaction of cement, respectively. Thus, in this study, the effects of the addition ratio of each fine aggregate cementitious powder on substitution rate for limestone and CO_2 reduction were quantitatively analyzed[7].

Table 1. Chemical compositions of Actual Waste cementitous powder and Fine aggregate Addition ratio of 65%

Classification	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3
Actual Waste Cementitous powder	15.1	55.8	10.2	2.9	1.7
Fine aggregate Addition ratio of 65%	17.5	55.2	9.2	2.2	17.5

2. Research methods

2.1 . Raw materials

Since the formulation and age of actual waste cementitious powder in each site are unknown, a waste cementitious powder model was made and a comparative analysis was performed in this study. The raw material was fine aggregate aged 6 months in 75 ㎛ or lower, which was difficult to separate and discharge because it had a grain size and grain form similar to waste cementitious powder. Limestone and converter slag were used as ingredient adjusters. Here, converter slag was used to supplement the Fe_2O_3. In particular, in the case of fine aggregate powder, it was performed as the addition ratio was increased to 0~100%. Multi-purpose optimal combination was performed by using the above 4 kinds as raw materials for clinker. At the time, Bogue values, which were commonly used by cement manufacturers as the standards for quality assessment, were 90~100, SM 2.41~2.8, IM 1.48~1.73, which were LSD values of clinker, an intermediate product in OPC manufacturing. These values were used as standards.

The chemical compositions of each raw material used in the waste cementitious powder model and ingredient adjuster are shown in Tables 1 and 2.

Table 1. Chemical compositions of waste cementitous powder model

Classification	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3
Paste	47.7	18.2	3.9	2.6	0.0
Fine aggregate	1.3	75.1	12.1	2.0	0.0

Table 2. Chemical compositions of each raw material and byproduct raw materials

Classification	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3
Limestone	45.2	11.2	2.1	0.81	0.2
Converter Slag	31.8	14.9	2.7	38.4	0.1

2.2 Methods to estimate CO_2 emission

The cement manufacturing process largely consists of a mining process, a raw material grinding process, a cement calcination process, production and a shipping process. Since CO_2 emission during the production of clinker, which is the intermediate product of cement in the calcination process, accounts for 84% or more of emissions in the entire process, CO_2 emission is estimated based on the calcination process. At this time, all CaO contents are assumed to be discharged through $Ca(OH)_2$ and $CaCO_3$ depending on the optimal combination of cement raw materials based on the addition ratio of fine aggregate cementitious powder. It is assumed that CO_2 emission caused by combustion and power is the same under all conditions.

According to the findings of Kim, Sang Hyo et al., the CO_2 emission ratio generated upon manufacturing clinker can be broken down as follows: decarbonation (67.5%), combustion (26.7%) and power (5.8%). The CO_2 emission factor of binder used in concrete that is caused by decarbonation includes emission from decarbonation 0.82(t CO_2-e/tonne) and from slag 0.143(t CO_2-e/tonne). CO_2 emission is estimated based on the fine aggregate addition ratio mixed in waste cementitious powder using the above factors.

The equation used to estimate CO_2 emission is shown in the following (3), (4), (5) and (6).

Decarbonation

$$\frac{A_l \times Ac \times 0.82 \times 44}{100 \times 56} \quad (3)$$

Where, Al : usage amount of limestone depending on addition ratio of each fine aggregate

AC : amount of CaO production

Combustion

$$\frac{D_c \times 0.329}{0.598} \quad (4)$$

Where, Dc : CO_2 emission by Decarbonation Reaction

Electric Power

$$\frac{D_c \times 0.072}{0.598} \quad (5)$$

Slag

$$As \times 0.143 \quad (6)$$

Where, As : use amount of slag

3. Research results and analysis

3.1 Optimal combination of clinker raw materials

In terms of the results of combination of raw materials in each addition ratio of fine aggregate cementitious powder through optimization, which are shown in Figure 3, it was found that the substitution rate of waste cementitious powder for limestone was decreased as the addition ratio of fine aggregate cementitious powder was increased. As shown in Figure 4, each chemical ingredient required based on OPC standards such as CaO and SiO_2 was constant, because SiO_2 was increased as fine aggregate cementitious powder mixed in waste cementitious powder increased, but CaO was decreased. At the time, according to chemical characteristics, waste cementitious powder was highly likely to be utilized as an alternative to clay, which was SiO_2 raw material, rather than limestone. Therefore, if fine aggregate cementitious powder was actually about 65%, the substitution rate of waste cementitious powder for limestone was about 9%, which was a significantly low level. In order to increase the substitution rate by utilizing cement hydrates with high-added value among waste cementitious powder, it is thought that efficient separation of fine aggregate cementitious powder is required.

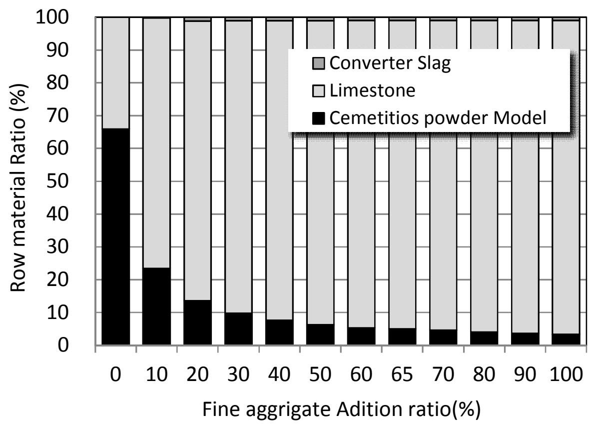

Figure 3. Raw material ratio from fine aggregate addition ratio

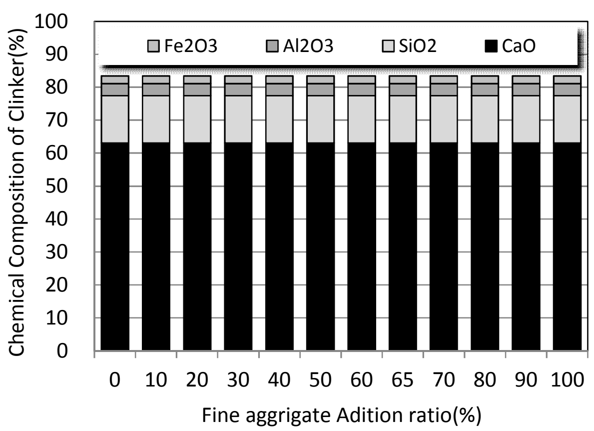

Figure 4. Chemical composition of clinker from fne aggregate addition ratio

3.2 Estimation of CO_2 emission

The results of estimation of CO_2 emission based on the addition ratio of fine aggregate cementitious powder were obtained as shown in Figure. 5. Under the assumption that CO_2 emission related to combustion and power was the same in all conditions, it was estimated as 15.4 and 3.3 (t CO_2 -e/tonne), respectively. However, in the case of decarbonation reaction, substitution rate of waste cementitious powder ($Ca(OH)_2 \rightarrow CaO+H_2O$) for limestone ($CaCO_3 \rightarrow CaO+ CO_2 \uparrow$) was increased with the increasing addition ratio of fine aggregate cementitious powder. At this time, a greenhouse gas emission of up to 56.56% was shown when addition ratio of fine aggregate was adjusted. In addition, as shown in Figure 6, it was found that the rate of CO_2 emission reduction was decreased with the increase in the addition ratio of fine aggregate cementitious powder. In particular, when the addition ratio of fine aggregate fell between 0%~20%, the reduction rate sharply declined, such as from 100% to 37.5%, 37.5% to 7.2%. However, when it was over 30%, the reduction of CO_2 emission was very minor. This reminds us of the need to develop technology to efficiently separate and discharge the fine aggregate cementitious powder mixed into waste cementitious powder.

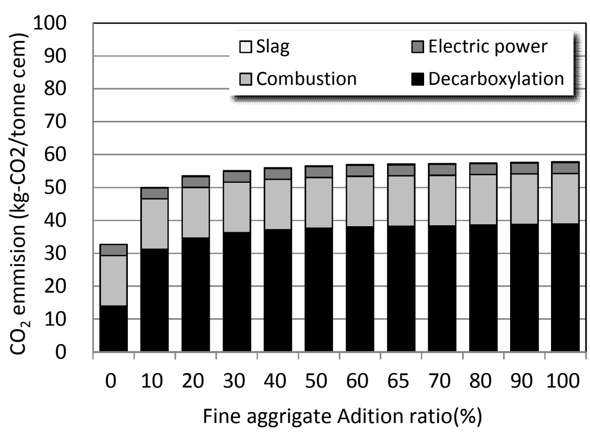

Figure 5. CO_2 emission from fine aggregate addition ratio

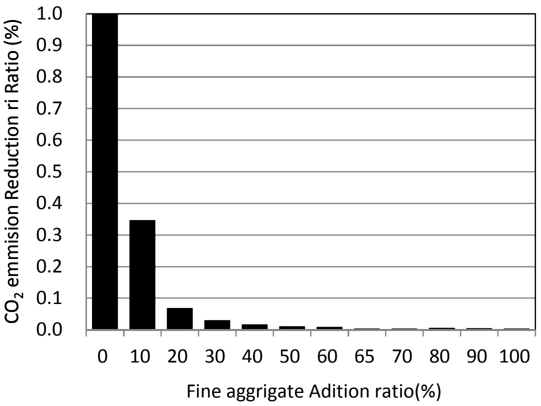

Figure 6. CO_2 emission reduction ratio from fine aggregate addition ratio

4. Conclusion

This study analyzed whether waste cementitious powder among the construction waste can be utilized as an alternative raw material, and handled the reduction of CO_2 emission. In particular, the purpose of this study was to reduce CO_2 emission in the clinker calcination process, which was responsible for the largest share of CO_2 emission in the entire cement manufacturing process.

Through the research, the following conclusions were obtained.

1) Through multi-purpose optimization, it was found that waste cementitious powder could be used as an alternative raw material for limestone to produce cement at the OPC level.
2) It was found that CO_2 emission was reduced in decarbonation reaction to produce CaO when waste cementitious powder was used as an alternative raw material for limestone.
3) It was found that the range in which waste cementitious powder was utilized as an alternative raw material for limestone was reduced as the addition ratio of fine aggregate in waste cementitious powder was increased, and the reduction of CO_2 emission was decreased. In addition, the actual addition ratio of fine aggregate in waste cementitious powder was 65% and the reduction rate of emission was only 0.27%. Thus, it is thought that technology to efficiently separate and discharge fine aggregate cementitious powder in waste cementitious powder should be developed in order to produce more greenhouse gas reduction effects.
4) It was found that the rate of greenhouse gas reduction was dramatically reduced when the addition ratio of fine aggregate in waste cementitious powder was increased. A significant reduction was shown when addition ratio of fine aggregate was 0%~20%.

Through this study, it was found that the utilization of waste cementitious powder as an alternative raw material to limestone had more effects on reduction of greenhouse gas by suppressing decarbonation reaction. It should be examined to determine whether this represented compressive strength and fluidity at the appropriate level for producing the actual recycled cement through property tests in the future. It is thought that the usage rate of waste cementitious powder as an alternative raw material for limestone will be increased, and that technology to efficiently separate and discharge fine aggregate cementitious powder should be developed to increase the reduction rate of greenhouse gas. In addition, study of the entire cement manufacturing process such as mining, grinding, mixing, transportation for CO_2 reduction is needed.

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