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# Development of a new ecological concrete with CO<sub>2</sub> emissions below zero

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#### HIGHLIGHTS

- $\gamma$ -C<sub>2</sub>S reacted with CO<sub>2</sub>, generated CaCO<sub>3</sub> and SiO<sub>2</sub> gel, formed a dense matrix.
- The amount of CO<sub>2</sub> can be measured with good accuracy by an inorganic carbon analyzer.
- The concrete added  $\gamma$ -C<sub>2</sub>S captured CO<sub>2</sub> included in the exhaust gas.
- The amount of CO<sub>2</sub> emission of the new ecological concrete achieved below zero.

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#### ABSTRACT

A new ecological concrete has been developed which utilizes CO<sub>2</sub> emitted from a power plant and which can achieve CO<sub>2</sub> emission levels below zero. This ecological concrete is named CO<sub>2</sub>-SUICOM (CO<sub>2</sub> Storage under Infrastructure by Concrete Materials). This concrete is based on two typical features. One is using a special additive (the dicalcium silicate  $\gamma$  phase:  $\gamma$ -2CaO·SiO<sub>2</sub>) and coal-ash instead of cement. These materials have very low level of CO<sub>2</sub> emissions, and γ-2CaO·SiO<sub>2</sub> hardens concrete by reaction with CO2. The other feature of this concrete is that CO2 in the exhaust gas of a power plant can be captured. After this concrete is manufactured, it is set in a chamber. Exhaust gas is drawn into the chamber, and CO<sub>2</sub> included in the exhaust gas is captured in the concrete. A method of evaluating the amount of CO2 in concrete was developed, which involved analyzing concentrations of CO2 using an inorganic carbon analyzing device. In this method it is important to use samples of a particular size. The method was used to confirm that amounts of CO<sub>2</sub> emitted during preparation of the concrete were below zero.

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#### 1. Introduction

The reduction of greenhouse gases has become a priority in various industries. The electricity industry accounts for approximately 30% of the CO<sub>2</sub> emissions generated in Japan. Therefore, enhancement of thermal efficiency in power stations and expansion of renewable energy sources such as hydro power, solar power and wind power are being promoted. Efforts are also being made to reduce CO<sub>2</sub> emissions, for example in the concrete industry in Japan [1]. A large amount of CO<sub>2</sub> is emitted during the process of manufacturing cement. For this reason, using by-products such as fly ash or granulated blast-furnace slag with a small amount of CO2 emissions instead of cement is the main method used to reduce

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CO<sub>2</sub> emissions in the concrete industry. For instance, according to the Japan Society of Civil Engineers (JSCE), CO<sub>2</sub> emissions can be reduced about 15% by using fly-ash cement type B, and reduced about 40% by using blast-furnace slag type B.

In this situation, the authors had found in the past that a very dense matrix could be obtained by mixing into concrete a special admixture containing  $\gamma$ -2CaO·SiO<sub>2</sub> (hereafter referred to as  $\gamma$ -C<sub>2</sub>S) as a major component, followed by forcibly carbonating the mixture. In addition, the authors developed a new ecological concrete by applying this "carbonation" technique to "CO<sub>2</sub> absorption - fixation of concrete [2,3]". A phenomenon called "neutralization" occurs during reaction with CO<sub>2</sub> in the air in ordinary concrete. However, the neutralization depth after 20–30 years may be only about 10 mm from the surface. In contrast, when a large quantity of CO<sub>2</sub> is absorbed in the new ecological concrete by controlling the curing temperature and humidity, CO2 emissions of the

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Fig. 1. Precast panel of CO<sub>2</sub>-SUICOM.

concrete can be significantly reduced, even if CO<sub>2</sub> emissions during manufacture of the concrete are not considered. This ecological concrete was named "CO<sub>2</sub>-SUICOM"; CO<sub>2</sub> Storage under Infrastructure by Concrete Materials [4]. Fig. 1 shows one example. Fig. 2 describes the SUICOM concept. The amount of CO<sub>2</sub> produced in ordinary concrete is about 400 kg/m³, which mainly comes from cement production. On the other hand, the amount of CO2 in SUI-COM is below 0 kg/m<sup>3</sup>. This SUICOM concrete is based on two typical features. One is that it uses a special admixture the  $\gamma$ -C<sub>2</sub>S instead of cement. This material has a very low level of CO<sub>2</sub> emissions, with the quantity of CO<sub>2</sub> emissions for this material being about one-fifth of that of ordinary Portland cement. Also, this material hardens concrete by reaction with  $CO_2$ . Since  $\gamma$ - $C_2S$  does not react with water, it does not contribute any strength development to ordinary concrete. However, when it reacts with CO<sub>2</sub>, it contributes strength development greater than that in ordinary Portland cement [5–7]. Also, the SUICOM concrete uses coal-ash instead of cement. As a result, this concrete can not only reduce the amount of CO<sub>2</sub> emitted from thermal power stations, but can also use by-products such as coal-ash effectively.

The other feature of the SUICOM concrete is its capture of  $CO_2$  included in the exhaust gas from thermal power stations. After this concrete is manufactured, it is set in a chamber. Exhaust gas is drawn into the chamber, and  $CO_2$  included in exhaust gas is captured in the concrete.  $CO_2$  emissions for this concrete can be reduced by half compared to the ordinary concrete by using

 $\gamma$ -C<sub>2</sub>S and coal-ash. Also, amounts of CO<sub>2</sub> exceeding emission amounts are captured in the hardening process of the concrete. In total, CO<sub>2</sub> emissions of this newly developed concrete material can be below zero.

For SUICOM, which has environmental performance as one of its advantages, the method used to properly evaluate the CO2 content contained in the concrete is important. Calcium carbonate is the main compound generated through the reaction between concrete and CO<sub>2</sub>. Thermal analysis is generally used as the technique to quantify calcium carbonate. In this method, the quantity is calculated from the mass decrease rate corresponding to decarbonation when heating the sample between 500 and 800 °C. However, within calcium carbonate generated through carbonation, there exists a phase with low crystallinity, which sometimes decreases in mass through decarbonation at lower temperatures. In this case, discrimination between hydrates such as calcium hydroxide and organic admixtures often becomes difficult. Also, the material that most contributes to the fixation of CO<sub>2</sub> among concrete components is the paste phase containing  $\gamma$ -C<sub>2</sub>S, and the contribution of aggregates is small. However, depending on the size of a sample piece of concrete used as the analytical sample, the ratio of paste phase and aggregates may differ in the concrete mix [8], which may affect the evaluation of  $CO_2$  content.

In this paper, the characteristics of the carbonation reaction of  $\gamma$ -C<sub>2</sub>S is investigated in detail, and the analytical instruments used for the evaluation of CO<sub>2</sub> content, as well as results examining the effect of the size of concrete specimens on CO<sub>2</sub> content are reported. In addition, the results of evaluation of CO<sub>2</sub> content by applying this analytical method to concrete cured with exhaust gas, as well as the CO<sub>2</sub> balance related to concrete production in consideration of the estimation of amounts of CO<sub>2</sub> emission resulting from the materials used are investigated and reported.

#### 2. Materials and proportions

### 2.1. Special additives $\gamma$ - $C_2S$

Table 1 shows the chemical and physical properties of  $\gamma$ -C<sub>2</sub>S.  $\gamma$ -C<sub>2</sub>S consists of CaO and SiO<sub>2</sub> as its principal components. Its main mineral is gamma type calcium silicate. It was manufactured using a by-product containing Ca(OH)<sub>2</sub> mainly, and SiO<sub>2</sub> powder. Fig. 3 shows the appearance of  $\gamma$ -C<sub>2</sub>S, which is a powdery material. Its specific surface area suggests that it is slightly coarser than Portland cement. Fig. 4 shows CO<sub>2</sub> emissions arising from the manufacture of ordinary Portland cement and  $\gamma$ -C<sub>2</sub>S from limestone and a by-product containing mainly Ca(OH)<sub>2</sub>. There are many manufacturing methods, but the quantity of CO<sub>2</sub> generated during manufacture of the special admixture can be reduced by using by-product which generates smaller quantities of CO<sub>2</sub>. In this case, the quantity of CO<sub>2</sub> emitted is about one-fifth that of ordinary cement.

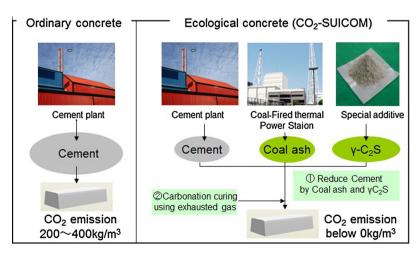


Fig. 2. CO<sub>2</sub>-SUICOM concept.

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