



# Development of carbon-capture binder using stainless steel argon oxygen decarburization slag activated by carbonation

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## ABSTRACT

The study aims at evaluating the possibility of using stainless-steel argon oxygen decarburization (AOD) slag containing  $\gamma$ -C<sub>2</sub>S as a carbon-capture construction material. For this purpose, the physicochemical changes of paste specimens with different levels of AOD slag replacement up to 70% were investigated by means of carbonation curing.  $\gamma$ -C<sub>2</sub>S is non-hydraulic, and hence, does not react with water at room temperature. After carbonation, however,  $\gamma$ -C<sub>2</sub>S yields reaction products such as calcite and silica gel that fill up the pores within the cement paste creating a high-density microstructure. As a result, the carbonated specimens demonstrate increased compressive strength. Due to its relatively insoluble property under typical carbonation curing conditions, silica gel is formed on the surfaces of reacting  $\gamma$ -C<sub>2</sub>S particles. Therefore, crystalline calcite and highly polymerized silica, formed as a result of CO<sub>2</sub> curing, play an important role in achieving the desired strength. Their role could be considered similar to that of calcium silicate hydrates (C-S-H), which are formed when concrete produced from Portland cement is hardened.

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## 1. Introduction

Carbon capture and storage (CCS) refers to the technology of extraction, capture, pressurization, and storage of CO<sub>2</sub> emitted from industrial sites. This technology essentially prevents release of CO<sub>2</sub> into the atmosphere. Currently, many countries are actively conducting studies to develop CCS technology. Mineral carbonation is one of the CO<sub>2</sub>-fixation technologies. It captures CO<sub>2</sub> from emission sources, such as thermoelectric power plants and ironworks, which is then reacted with inorganic industrial by-products discharged from industrial activities or naturally occurring minerals to fixate the greenhouse gas within crystal structures of carbonate minerals (Lee and Koo, 2016). Carbonate minerals are the most stable in terms of thermodynamics among substances containing CO<sub>2</sub> (Lackner, 2002). Thus, mineral carbonation is considered an appealing technology for the fixation of CO<sub>2</sub> produced from man-made industrial activities, and many countries are investigating this process (Chae et al., 2009; Chang et al., 2013). Inorganic industrial by-products that could be used for storing greenhouse gases include, coal fly ash, steelmaking slag, and cement kiln dust

etc (Ukwattage et al., 2013, 2015; Eloneva et al., 2010; Huntzinger et al., 2009). Approximately 50 million tons of these industrial by-products are produced annually in South Korea, and utilizing them for mineral carbonization could theoretically fixate approximately 12 million tons of greenhouse gas annually (Lee and Koo, 2016). AOD slag accounts for approximately 5–6% of the total annual industrial by-product.

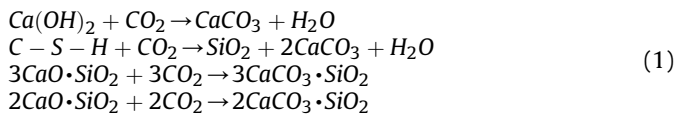
The source of calcium oxide or magnesium oxide capable of CO<sub>2</sub> fixation is alkali solid industrial by-products such as stainless steel slag. Stainless steel slag has high potential for applications and low cost, making it an ideal material for capturing CO<sub>2</sub> (Chang et al., 2012). However, there exist challenges such as volume instability, heavy-metal leaching, and low cementitious activity in the utilization of steel slag as supplementary cementitious materials (SCMs) (Pan et al., 2016). Several studies (Monkman et al., 2009; Pan et al., 2013) have reported utilization of carbonation curing for steel slag treatment as a novel approach for overcoming the above barriers. Prior to utilization of steelmaking slags as SCMs, carbonation curing serves to improve physicochemical properties of slags by combining them with CO<sub>2</sub> for stabilization (Li et al., 2016). Steelmaking by-products could be categorized into basic oxygen furnace slag (BOFS) and electric arc furnace (EAF) slag. Basically, EAF slag is largely divided into EAF oxidizing slag and AOD

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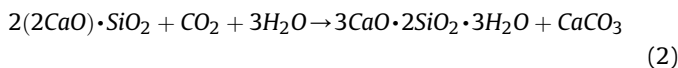
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slag depending on the processing methods involved. AOD slag incurs an expansion due to the crystal phase transition of 2 CaO·SiO<sub>2</sub>(C<sub>2</sub>S) when molten slag at high temperature is cooled such that it contains a large amount of γ-C<sub>2</sub>S (Kriskova et al., 2012; Tossavainen et al., 2007; Watanabe et al., 2006).

Carbonation curing involves the use of CO<sub>2</sub> as a curing agent in concrete manufacturing. CO<sub>2</sub> curing refers to a curing method that produces a large amount of carbonates through injection of CO<sub>2</sub>. In the process, H<sub>2</sub>CO<sub>3</sub> is produced from CO<sub>2</sub> dissolving in the pore solution that exists within the concrete matrix. Then, it is converted to CO<sub>3</sub><sup>2-</sup> and reacts with Ca<sup>2+</sup> in the pore solution to produce calcium carbonate (CaCO<sub>3</sub>). During this reaction process, the pH of the pore solution drops. CO<sub>2</sub> not only reacts with typical hydration products of cement such as Ca(OH)<sub>2</sub> and calcium silicate hydrates (C-S-H), but also reacts with C<sub>3</sub>S and C<sub>2</sub>S that were not consumed during hydration as shown in Eq. (1) below. This produces CaCO<sub>3</sub> and prevents leaching of Ca<sup>2+</sup>, which increases the resistivity to a further pH decrease. Moreover, it contributes to reducing the setting time and achieving compressive strength (Chang and Chen, 2005; Dias, 2000; Qian et al., 2016).



γ-C<sub>2</sub>S is non-hydraulic and does not react with water under typical room temperature conditions (Taylor, 1990; Yoshioka et al., 2013). However, it is highly reactive in the presence of both water and CO<sub>2</sub> (Saito et al., 2010; Santos et al., 2013). The carbonation of γ-C<sub>2</sub>S can be defined with Eq. (2) and the main by-products in the production of stainless steel slag from carbonation curing are calcium silicate hydrates (C-S-H) and calcium carbonate (Young et al., 1974).



When γ-C<sub>2</sub>S is used as an admixture for cement, it reacts with CO<sub>2</sub> to produce calcium carbonate (Eq. (2)), which increases in volume to form a dense matrix (Watanabe et al., 2006). Consequently, when cement and materials containing γ-C<sub>2</sub>S are used in cementitious materials their porosity decreases with the carbonation of γ-C<sub>2</sub>S. This improves the durability owing to increased strength, the shielding effect of chlorine ions, and the prevention of calcium ion leaching (Chang et al., 2013; Lee and Koo, 2016).

In recent years, certain researchers suggested concrete products produced by implementing the reaction process of accelerated mineral carbonation (Thiery et al., 2013; Xuan et al., 2016; Zhan et al., 2013). This is a curing process that accelerates reactions between components in cement and CO<sub>2</sub> under a certain pressure. This process can quickly achieve the strength required for concrete products by improving their resistance against surface permeation, freezing and thawing, and sulfate attack (El-Hassan and Shao, 2014). Zhan et al. (2016) achieved strengths of approximately 24 MPa or higher for concrete blocks that were subjected to pressurized CO<sub>2</sub> curing at 23 ± 1 °C, 60 ± 10%, and pressure of 0.1 bar for 2 h. Furthermore, it was confirmed that these strengths were

similar to that of concrete blocks exposed to wet curing for 28 days. El-Hassan and Shao (2014) used initial curing on carbonation curing of lightweight concrete. They showed that 22–24% CO<sub>2</sub> based on the cement content could be taken up by concrete blocks within 4 h of carbonation curing.

According to Salman et al. (2014), curing paste mixed with AOD stainless steel (0.15 water to slag ratio by weight) for 7 days at atmospheric pressure under the conditions of 22 °C, 5 vol% CO<sub>2</sub>, and 80% relative humidity provided an average compressive strength of 20 MPa or higher. Mo et al. (2016) performed accelerated carbonation curing for PC blend paste with 100% steel slag and 80% steel slag replacement under conditions of 99.9% CO<sub>2</sub> and pressure of 0.1. Fourteen days of CO<sub>2</sub> curing resulted in a compressive strength of 44.1 MPa and 72.0 MPa for each specimen, respectively. Watanabe et al. (2012a, 2012b) mixed γ-C<sub>2</sub>S and various pozzolan materials in low-heat Portland cement to perform accelerated carbonation curing. As a result, the porosity was decreased substantially due to the reaction of γ-C<sub>2</sub>S with CO<sub>2</sub>, which is promoted by the pozzolanic admixture. Guan et al. (2016) reported that the maximum amount of CO<sub>2</sub> uptake could vary according to the curing temperature, pressure, and humidity.

This study aims at evaluating the possibility of using stainless-steel AOD slag containing γ-C<sub>2</sub>S as carbon-capture construction materials. For this purpose, AOD slag was used as replacement in ordinary Portland cement (OPC) in various ratios by weight, and specimens were accordingly manufactured to investigate the changes that occur in the physiochemical properties of the mixture owing to carbonation curing. Heat flow of hydration as well as specific heat flow were utilized in order to analyze the effects of the use of AOD slag on OPC hydration. X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyses were performed in order to study the changes in chemistry and microstructures within the paste. Moreover, the compressive strength performance and micro-pore distribution were analyzed to investigate the changes in physical properties on day 7 and day 28 of CO<sub>2</sub>- and non-CO<sub>2</sub>-curing cases. A thermogravimetric analysis (TGA) was performed to quantify the CO<sub>2</sub> uptake of the paste using AOD slag as replacement.

## 2. Experiment

### 2.1. Materials

This study used cement and stainless steel AOD slag as raw materials. For cement, ordinary Portland cement (OPC) was used with density of 3.14 g/cm<sup>3</sup>, and fineness of 332 m<sup>2</sup>/kg. AOD slag manufactured by POSCO of South Korea was used. The slag is characterized by the special process of air-cooling to 500 °C. Through this process, 2CaO·SiO<sub>2</sub>(C<sub>2</sub>S) transforms into the gamma phase alongside an expansion in volume. Density and fineness are 2.91 g/cm<sup>3</sup> and 359 m<sup>2</sup>/kg, respectively. Table 1 provides the chemical compositions of the AOD slag and OPC obtained through X-ray fluorescence (XRF) analysis. As shown in the table, CaO, SiO<sub>2</sub>, MgO, and Al<sub>2</sub>O<sub>3</sub> are the main components of AOD slag each constituting 54.3%, 17.7%, 9.2%, and 6.4%. The Bogue phase composition of OPC on a weight basis was 58.1%, 15.0%, 8.1%, and 9.2% for C<sub>3</sub>S, C<sub>2</sub>S, C<sub>3</sub>A, and C<sub>4</sub>AF, respectively.

**Table 1**  
Chemical composition of raw materials.

	Chemical compositions (%)													Sum
	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	Cr <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	SO <sub>3</sub>	MnO	LOI		
AOD slag	54.3	17.7	6.4	3.0	9.2	2.8	0.9	0.2	0.1	0.4	0.5	1.7	97.2	
OPC	62.4	20.8	4.9	3.5	1.6	–	0.1	0.9	0.3	2.2	0.1	2.7	99.5	

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