

# High durability cementitious material with mineral admixtures and carbonation curing

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## Abstract

Nuclear waste repositories need highly durable cementitious materials to function for over thousands of years while resisting leaching and degradation. The durability of cementitious material can be effectively improved by reducing permeability and by changing cement hydrates to a less soluble matrix. This paper describes the properties of carbonated new cementitious materials containing belite-rich cement and  $\gamma$ -2CaO · SiO<sub>2</sub> as main components. In addition, the long-term leaching properties are investigated and compared with ordinary Portland cement by using a predictive leaching model.

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## 1. Introduction

The use of cementitious materials in various barriers is being considered in the construction of radioactive waste disposal facilities, anticipating good mechanical performance and shielding properties. Very high durability is demanded of cementitious material since the service life of important structures, such as radioactive waste disposal facilities, may be tens of thousands of years. Leaching, by which cementitious material gradually dissolves because of long-term contact with groundwater, becomes an important topic in structures constructed at considerable depths, such as waste disposal facilities. Leaching occurs in which ions such as calcium (Ca) exit the cementitious material, dissolve in the pore solution, and after percolating the pore, increase the pore volume of the cement matrix. Controlling mass transfer of concrete by reducing pores and reducing dissolution of products by improving the quality of the cement matrix are effective measures to improve

durability against leaching for concrete. More specifically, the method of making the hydrate composition difficult to dissolve while reducing pores by accelerated carbonation of the cement composites may be considered.

Previous reports (Sakai et al., 1999; Yokozeki et al., 2002a) have been confirmed that pores can be reduced, leaching resistance improved, and strength increased by accelerated carbonation of belite-rich cement (low heat Portland Cement). This paper describes the effects of  $\gamma$ -phase dicalcium silicate ( $\gamma$ -2CaO · SiO<sub>2</sub>; hereafter referred to as “ $\gamma$ -C<sub>2</sub>S”), which reacts with carbon dioxide during carbonation curing and adds to the strength although no hydration reaction occurs. The physical and chemical properties of the cement matrix, anti-leaching performance and ability to inhibit mass transfer were experimentally evaluated; the long-term durability was studied by a numerical analysis methods based on this evaluation.

## 2. Materials

Table 1 shows the materials used in this research.  $\gamma$ -C<sub>2</sub>S is an important cement compound and is a crystalline form

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Table 1  
Materials

Type	Notation	Name	Specific surface area (cm <sup>2</sup> /g)	Density (g/cm <sup>3</sup> )	Properties							
					Chemical composition (mass %)							
					CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	SO <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O
Cement	OPC	Ordinary Portland cement	3310	3.16	64.6	21.3	5.3	2.9	1.5	2.0	0.3	0.5
	LPC	Low heat Portland cement	3380	3.22	63.0	26.1	2.8	2.7	0.9	2.1	0.3	0.2
Mineral admixtures	$\gamma$ -C <sub>2</sub> S	Dicalcium silicate ( $\gamma$ phase)	1500	2.99	61.9	35.0	1.7	0.1	0.5	–	–	0.1
	FA	Fly ash	5450	2.40	3.7	55.2	28.6	7.8	0.7	1.5	0.8	1.6
	SF	Silica fume	200,000	2.20	0.1	97.7	0.4	0.1	0.4	–	0.1	0.6
Fine aggregate	S	Pit sand	–	2.65	FM 2.63, percentage of water absorption 1.88%							
Coarse aggregate	G	Crushed stone	–	2.65	FM 6.63, percentage of water absorption 0.65%, $G_{\max}$ 20 mm							

Table 2  
Chemical compositions of raw materials for  $\gamma$ -C<sub>2</sub>S

Raw material	LOI	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	TiO <sub>2</sub>
Limestone powder	43.9	0.1	0.1	0.0	55.4	0.4	0.0	0.0	0.1
Silica powder	0.5	95.8	2.7	0.3	0.0	0.0	0.0	0.5	0.2

of dicalcium silicate (hereafter referred to as “C<sub>2</sub>S”). C<sub>2</sub>S may occur as  $\alpha$ ,  $\alpha'$ ,  $\beta$ ,  $\beta'$ , but the type used in this research,  $\gamma$ -C<sub>2</sub>S is not part of normal cements. On the other hand, Kondo (1974) has reported that  $\gamma$ -C<sub>2</sub>S when reacted with CO<sub>2</sub> forms calcium carbonate (CaCO<sub>3</sub>), and expands in volume to form a dense matrix.  $\gamma$ -C<sub>2</sub>S was produced by calcination of industrial raw materials; limestone powder, and silica powder shown in Table 2, in an actual rotary kiln at 1450 °C, kept for 20 min and cooled slowly. When the product is cooled,  $\gamma$ -C<sub>2</sub>S powder was obtained by “dusting”. Dusting is a phenomenon whereby the  $\alpha$  or  $\alpha'$  phase inverts spontaneously in the course of cooling; the resulting increase in volume causes the sintered cake to lose coherence, hence it is said to “dust.”

Table 3 shows the mineral compositions of ordinary Portland cement (OPC) and low heat Portland cement (LPC). The contents of C<sub>3</sub>S and C<sub>3</sub>A in LPC are less, and the content of C<sub>2</sub>S in LPC is larger than in OPC. By using LPC, thermal cracking and autogenous shrinkage of concrete can be reduced. In addition, the amount of

Table 3  
Mineral composition of Portland cement

	Compound			
	C <sub>3</sub> S	C <sub>2</sub> S	C <sub>3</sub> A	C <sub>4</sub> AF
OPC	52	24	9	9
LPC	29	54	3	9

Table 4  
Product solubility

	Ca(OH) <sub>2</sub>	CaCO <sub>3</sub>	C–S–H <sup>a</sup>	Monosulfate <sup>b</sup>	Ettringite <sup>c</sup>
Product of solubility logK <sub>sp</sub>	–5.05 [4]	–8.41 [5]	–8.16 [6]	–29.43 [4]	–44.55 [4]

<sup>a</sup> mCaO · SiO<sub>2</sub> · mH<sub>2</sub>O + H<sub>2</sub>O → mCa<sup>2+</sup> + 2mOH<sup>–</sup> + SiO<sub>2</sub>.<sup>b</sup> 3CaO · Al<sub>2</sub>O<sub>3</sub> · CaSO<sub>4</sub> · 12H<sub>2</sub>O → 6Ca<sup>2+</sup> + 2Al(OH)<sub>4</sub><sup>–</sup> + 3SO<sub>4</sub><sup>2–</sup> + 4OH<sup>–</sup> + 6H<sub>2</sub>O.<sup>c</sup> 3CaO · Al<sub>2</sub>O<sub>3</sub> · 3CaSO<sub>4</sub> · 32H<sub>2</sub>O → 6Ca<sup>2+</sup> + 2Al(OH)<sub>4</sub><sup>–</sup> + 3SO<sub>4</sub><sup>2–</sup> + 4OH<sup>–</sup> + 26H<sub>2</sub>O.

formed Ca(OH)<sub>2</sub> (calcium hydroxide ; CH) in LPC is small. Since the amount of CH generated in LPC is small compared to that generated in OPC, it is likely to improve the anti-leaching performance. Furthermore, fly ash (FA) and silica fume (SF) were both used as mineral admixtures.

### 3. Material design

Table 4 shows the solubility product of cementitious products (Atkinson et al., 1990; Readon, 1992; Chemical Society of Japan, 1984). The comparison of solubility product of the hydration products of cement, and the effects of improvement of anti-leaching performance by carbonation and replacement of mineral admixture are given below. Fig. 1 also shows a sketch of the effects.

- CH is changed to C–S–H with small solubility by pozzolanic reaction of SF or FA with formed CH.
- The amount of CH generated is reduced by reducing the unit content of cement by replacement of mineral admixtures, and the anti-leaching performance is improved (Watanabe et al., 2000).
- CH is converted to CaCO<sub>3</sub> with low solubility by carbonation.
- The watertightness is improved by using CaCO<sub>3</sub> together with  $\gamma$ -C<sub>2</sub>S, which reacts with carbon dioxide and forms a dense matrix.

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