# A model for predicting the coefficient of thermal expansion of cementitious paste

# Pongsak Choktaweekarn<sup>a</sup>, Somnuk Tangtermsirikul<sup>b</sup>

<sup>a</sup> School of Civil Engineering and Technology, Sirindhorn International Institute of Technology, Thammasat University, Pathum Thani 12120, Thailand

<sup>b</sup> Construction and Maintenance Technology Research Centre (CONTEC), Sirindhorn International Institute of Technology, Thammasat University, Pathum Thani 12120, Thailand

\* Corresponding author, e-mail: pongsak@siit.tu.ac.th

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**ABSTRACT**: The coefficient of thermal expansion (CTE) of cementitious paste at various ages was studied. Pastes were prepared with various water to binder and fly ash to binder ratios. The CTE of paste increased with age, and decreased with fly ash content, particularly at an early age. A model for predicting the CTE of paste was proposed as a time, material, and mix proportion dependent function. The model was verified with various experimental results and the verification results were satisfactory.

KEYWORDS: mass concrete, fly ash, autogenous shrinkage

#### **INTRODUCTION**

Mass concrete is defined as "any volume of concrete with dimensions large enough to require that measures be taken to cope with generation of heat from hydration of the cement and attendant volume change to minimize cracking."<sup>1</sup> During the construction of massive concrete structures such as dams and mat foundations, temperature gradients occur inside the structures due to heat of hydration. Consequently, thermal stress is developed which may reach its critical value. Since concrete possesses a low thermal conductivity, cumulative heat from hydration which results in temperature gradients can induce cracks, especially at an early age. To predict the conditions under which thermal cracking will occur, quantitative evaluation of heat evolution during hardening as well as thermal properties and related mechanical properties at an early age are necessary. During hydration, the amount of unreacted cementitious materials and water decreases, but the amount of hydrated products increases, causing the thermal properties of concrete to vary with the degree of reaction and then with time<sup>2,3</sup>. The properties used in thermal cracking analysis of mass concrete are the heat of hydration of the cementitious materials, thermal conductivity, specific heat, coefficient of thermal expansion (CTE), modulus of elasticity, tensile strain capacity or tensile strength, creep, and shrinkage<sup>1</sup>. The change of all of these properties with time should be predictable.

One significant property is the CTE which is used to compute the strain due to temperature variation in mass concrete<sup>4</sup>. CTE is defined as

$$\alpha = \frac{\varepsilon}{\Delta T} = \frac{\Delta L}{L \,\Delta T},\tag{1}$$

where  $\varepsilon$  is the strain due to temperature change,  $\Delta L$  is the length change due to temperature change, L is the initial length, and  $\Delta T$  is the temperature change.

The CTE of concrete depends on the CTEs of the ingredients, which are cementitious materials, water, hydration products, and aggregates. The CTE of concrete is mainly affected by the CTEs of cement paste and aggregates. It was found that the CTE increased with the increase in density of concrete<sup>5</sup>. For mass concrete, it is important to evaluate its CTE at early age because thermal cracking usually occurs at early age. Sellevold and Bjøntegaard<sup>6</sup> found that CTE at early age is time-dependent. Before setting, both cement paste and concrete have high CTE values since no solid framework exists and the continuous water phase controls the expansion. The CTE decreases as the solids form, reaching a minimum value around the final set. After that, the CTE increases as selfdesiccation proceeds. Various types of materials are used on different occasions. Fly ash is widely used in mass concrete but the data on the effect of fly ash on the CTE of concrete are extremely limited. Wesche<sup>7</sup> found that the use of fly ash reduced the CTE of concrete. However, the magnitude of the decrease was not reported.

The measuring methods of CTE used in some studies require sophisticated equipment and tests were usually done at a late age<sup>8,9</sup>. Some dried the specimens before test<sup>10</sup>. As a result, the moisture condition was different from that of real mass concrete. Autogenous shrinkage strain during measurement is one of the important parameters affecting the values obtained for the CTE. Yang and Sato<sup>11</sup> avoided autogenous shrinkage strain during the measurement of thermal strain by reducing the hydration that occurred during the test period. The CTE excluding autogenous shrinkage could be obtained by performing the tests at low temperature (-1 to 5 °C). However, this requires sophisticated equipment. In the method proposed by Bjøntegaard and Sellevold<sup>12</sup>, the CTE was measured by using various temperature histories. The specimens were heated up by 7-10 °C per step until reaching the required temperature history. Thermal deformation was measured in between each temperature step and autogenous deformation was measured directly at each temperature step. The method is reasonable for measurement of the CTE by automatically deleting the effect of autogenous shrinkage, but it requires relatively complicated facilities. Kada et al<sup>13</sup> proposed a test method of applying a temperature shock in the range of 10-50 °C which can be carried out in less than one hour. The effect of autogenous shrinkage was neglected in that study because the test duration was short. However, the high temperatures may have caused moisture movement to the surface inside the plastic wrap during the test. In the present study, our CTE test method was designed to minimize the effects of autogenous shrinkage and prevent moisture transfer during testing.

Various expressions have been proposed for calculating the CTE of concrete  $^{14-16}$  all of which require a value for the CTE of paste. The CTE of paste can not be predicted solely from the concrete mix proportion, but requires an accurate prediction model made with different materials and mix proportions. In this study we propose a model for predicting the CTE as a function of time, material, and mix proportion. Then the proposed model is used for simulating thermal cracking in mass concrete. If the modulus of elasticity of concrete at the age before setting is small, then the stress induced by the rise in temperature is insignificant, even in zones of full restraint<sup>17</sup>. Thermal cracking usually occurs at an age after which the concrete has gained enough stiffness to generate restraint. Hence the CTE before hardening and a few hours after hardening (12 h after mixing) are not considered in this study.

### MATERIALS AND METHODS

The measuring method of Kada et al<sup>13</sup> and standard test method ASTM C531<sup>18</sup> were adopted and modified for use in this study due to their simplicity. The CTE test method was designed to minimize the effect of autogenous shrinkage deformation by ensuring that the measuring duration was minimal. The temperature range was controlled to prevent moisture transfer during testing. Although the test duration was short and autogenous shrinkage strain can be minimized, the autogenous shrinkage strain was calculated and deducted from the measured strain by applying a model to compute the paste autogenous shrinkage strain<sup>19</sup>.

#### Materials and mix proportions

Cement-fly ash pastes were produced and tested at various ages (1, 3, 7, and 28 days). Chemical compositions and physical properties of the cement and fly ash used in the tests are given in Table 1. Cement-fly ash pastes were tested to observe the effect of water and fly ash content. Various values of water to binder ratio by weight (w) and fly ash to binder ratio by weight, r, were used. We denote a w of 0.25 and r of 0.30 by w25r3. The CTE was tested at 1, 3, 7, and 28 days of age and mixtures w25r0 and w25r5 of the cement-fly ash paste were tested at 12 h.

#### Specimen preparation and testing

Prism specimens (25 mm  $\times$  25 mm  $\times$  285 mm) were used in this study. It is known that moisture content of concrete is an important factor influencing the CTE. Inside mass concrete the moisture content is approximately constant<sup>20</sup>. To emulate this, water loss

Table 1 Chemical compositions and physical properties of ordinary Portland cement and fly ash.

| Material | SiO <sub>2</sub> (%) | $Al_2O_3(\%)$ | $Fe_2O_3$ (%) | CaO (%) | MgO (%) | SO <sub>3</sub> (%) | LOI <sup>a</sup> (%) | BF <sup>b</sup> (cm <sup>2</sup> /g) | Specific gravity |
|----------|----------------------|---------------|---------------|---------|---------|---------------------|----------------------|--------------------------------------|------------------|
| cement   | 20.20                | 4.70          | 3.73          | 63.40   | 1.37    | 1.22                | 2.72                 | 3190                                 | 3.15             |
| fly ash  | 36.10                | 19.40         | 15.10         | 17.40   | 2.97    | 0.77                | 2.81                 | 2510                                 | 2.10             |

<sup>a</sup> Loss on ignition

<sup>b</sup> Blaine fineness

in the specimens was minimized by firmly wrapping them in aluminium foil immediately after casting, gluing the seams with adhesive tape, and performing the tests without removing the foil. The specimens were kept at room temperature until the test date. Each data value was the average of the value measured from two specimens. A thermocouple was placed at the centre of each specimen to measure the specimen temperature.

The CTE of concrete is approximately constant within the normal temperature range experienced by mass concrete<sup>21</sup>. The temperature range used in the experiment was adopted from Kada et al <sup>13</sup>. The specimens were tested by placing them in a refrigerator to reduce the temperature from room temperature (about  $30 \pm 2$  °C) to 10 °C, then moving them out of the refrigerator to return to room temperature. For every 5 °C change of temperature, the length change was measured using a length comparator.

#### **Calculation of CTE**

The CTE was calculated using

$$\alpha = \frac{\varepsilon \pm \varepsilon_{\rm AS}}{\Delta T},\tag{2}$$

where  $\alpha$  is the tested thermal expansion coefficient,  $\varepsilon$ is the measured strain due to temperature change,  $\varepsilon_{\rm AS}$ is the strain due to autogenous shrinkage occurring during the temperature change, and  $\Delta T$  is the temperature change. The plus sign is used for the heating stage and the minus sign is used for the cooling stage. For computing the paste autogenous shrinkage strain as a function of age t we used<sup>19</sup>

$$\varepsilon_{\rm AS}(t) = \varepsilon_{\rm AS,Chem}(t) + \varepsilon_{\rm AS,Phy}(t) + \varepsilon_{\rm exp}(t),$$
 (3)

where  $\varepsilon_{AS,Chem}(t)$  and  $\varepsilon_{AS,Phy}(t)$  are the autogenous shrinkage strains contributed by chemical and physical effects and  $\varepsilon_{exp}(t)$  is the chemical expansion strain resulting from the fly ash in the cement-fly ash paste.

The four major oxide compounds in cement are tricalcium silicate (C<sub>3</sub>S), dicalcium silicate (C<sub>2</sub>S), tricalcium aluminate (C<sub>3</sub>A), and tetracalcium aluminoferrite (C<sub>4</sub>AF)<sup>14</sup>. The autogenous shrinkage strain resulting from the hydration reaction of these along with the pozzolanic reaction of fly ash is given by

$$\varepsilon_{\rm AS,Chem}(t) = \sum_X \kappa_X m_X \psi_X \lambda_X(t)$$
 (4)

where X stands for C<sub>3</sub>A, C<sub>4</sub>AF, C<sub>3</sub>S, C<sub>2</sub>S, and fly ash, and  $\kappa_X$ ,  $m_X$ ,  $\psi_X$ , and  $\lambda_X$  are, respectively, the volume reduction per unit mass of X, the mass of X per unit volume of paste, the retardation factor of X hydration by fly ash, and the degree of hydration of X in cement. Note that  $\psi_X = 1$  except when  $X = C_3S$ , and  $\lambda_{FA}(t)$  is the degree of pozzolanic reaction.

The autogenous shrinkage strain by physical effect is given by <sup>19</sup>

$$\varepsilon_{\rm AS, Phy}(t) = \frac{1.962\gamma A_{\rm S}(t)}{r_{\rm av}(t)E_{\rm S}}$$
(5)

where  $\gamma$  is the surface tension of water,  $A_{\rm S}(t)$  is the area factor,  $r_{\rm av}(t)$  is average radius of the pores in the cement paste, and  $E_{\rm S}$  is elastic modulus for capillary stress. In this study,  $E_{\rm S}$  is assumed to be about 1/4 of the ordinary value of the static elastic modulus of cement paste.

The chemical expansion strain of fly ash-cement paste is given by <sup>19</sup>

$$\varepsilon_{\exp}(t) = (258s + 109.42r + 200w - 60) \times 10^{-6} \times \tan^{-1} \left[ (0.4159 + 3.37s) t \right]$$
(6)

where s is the proportion by weight of  $SO_3$  in fly ash and t must be given in days.

It was found that the amount of autogenous shrinkage in each step of temperature change was small because the test period in each step was short (about 10 min), which is consistent with a previous study<sup>13</sup>.

#### Thermal expansion coefficient model of paste

After the formation of a strong paste structure, the CTE of the paste varies according to the CTE and volume fraction of non-reacted cementitious materials and hydrated product. During the reaction, the amount of unhydrated cement and non-reacted fly ash decreases with time but the amount of hydrated product increases. An equation for estimating the CTE of pastes is

$$\alpha_{\rm p}(t) = a n_{\rm uc}(t) \alpha_c + b n_{\rm ufa}(t) \alpha_{\rm fa} + c n_{\rm hp}(t) \alpha_{\rm hp}$$
(7)

where  $\alpha_c$ ,  $\alpha_{fa}$ , and  $\alpha_{hp}$  are the CTEs of cement, fly ash, and the hydrated and pozzolanic reaction products, respectively, and  $n_{uc}(t)$ ,  $n_{ufa}(t)$ , and  $n_{hp}(t)$ are the volumetric ratios of unhydrated cement, nonreacted fly ash, and the hydrated and pozzolanic reaction products, respectively. The constants *a*, *b*, *c*, and the CTE of hydrated and pozzolanic products were obtained by using the method of regression analysis from the authors' test results of pastes (with and without fly ash). The values of *a*, *b*, and *c* obtained were 0.284, 1.230, and 1.499, respectively.

For simplicity at this stage, the proposed model can be applied to predict the CTE of paste at an age greater than 12 h. All hydrated and pozzolanic reaction products are assumed to have the same CTE. This CTE was found to be about  $20 \times 10^{-6}$  K<sup>-1</sup>. The effect of air on the volume change of hardened concrete due to temperature change is negligible after the formation of a strong paste structure. The volume of hydrated product is composed of the volume of hydrated cement, reacted fly ash, and water consumed by hydration and pozzolanic reaction, and is hence given by

$$n_{\rm hp}(t) = 1 - n_{\rm uc}(t) - n_{\rm ufa}(t) - n_{\rm wfree}(t) - n_{\rm air}$$
 (8)

where the the degree of hydration of cement, the degree of pozzolanic reaction of fly ash, and the volume of free water are given by  $^{19}$ 

$$n_{\rm uc}(t) = n_{\rm uc0} \left(1 - \lambda_{\rm hy}(t)\right) \tag{9}$$

$$n_{\rm ufa}(t) = n_{\rm ufa0} \left(1 - \lambda_{\rm poz}(t)\right) \tag{10}$$

$$n_{\rm wfree}(t) = \frac{M_{\rm fw0} - M_{\rm whp}(t) - M_{\rm wgel}(t)}{\rho_{\rm w} V_{\rm p}} \qquad (11)$$

where

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$$M_{\rm whp} = \theta_{\rm hpc} M_{\rm uc0} \lambda_{\rm hy} + \theta_{\rm hpf} M_{\rm ufa0} \lambda_{\rm poz} \qquad (12)$$

$$M_{\text{wgel}} = \left(0.0126 + \frac{0.0026}{-1.009 + \exp(0.1414w)}\right) M_{\text{uc0}}\lambda_{\text{hy}} + (6.177r^2 + 9.681r)(8.87w^{8.44} + 0.1156)M_{\text{ufa0}}\lambda_{\text{poz}},$$
(13)

with  $r \leq 0.5$ , where  $n_{\rm uc}(t)$ ,  $n_{\rm ufa}(t)$ ,  $n_{\rm wfree}(t)$ , and  $n_{\rm hp}(t)$  are the volumetric ratios at age t of unhydrated cement, non-reacted fly ash, free water, and the hydration and pozzolanic reaction products, respectively,  $n_{\rm air}$  is the volumetric ratio of air, and  $n_{\rm uc0}$  and  $n_{\rm ufa0}$  are volumetric ratios of cement and fly ash at the time of mixing (at t = 0).  $M_{\rm whp}(t)$  and  $M_{\text{wgel}}(t)$  are the masses at the considered age of water consumed by hydration and pozzolanic reactions, and gel water in the mixture, respectively.  $M_{\rm fw0}$ ,  $M_{\rm uc0}$ , and  $M_{\rm ufa0}$  are the masses of water, cement, and fly ash at t = 0, respectively.  $\rho_w$  is the density of water and  $V_{\rm p}$  is the volume of paste.  $\lambda_{\rm hy}(t)$  is the degree of hydration of cement in the paste,  $\lambda_{poz}(t)$  is the degree of pozzolanic reaction of fly ash in the paste,  $\theta_{
m hpc}$  is the minimum weight ratio of water to cement for completing hydration reaction, and  $\theta_{hpf}$  is the



Fig. 1 Average degree of hydration of cement paste with w = 0.40 at constant temperature (30 °C and 60 °C) and varied concrete temperature.



Fig. 2 Degree of pozzolanic reaction of paste with fly ash replacement ratio of 0.50 and w = 0.35 at constant temperature (30 °C and 60 °C) and varied concrete temperature.

minimum weight ratio of water to pozzolanic material for attaining the maximum pozzolanic reaction.

The details of free water determination are briefly described in this paper (for more details see Ref. 22). The average degree of hydration of cement is defined as the weight average of the degree of hydration of all cement compounds in the paste mixture, whereas the degree of pozzolanic reaction of fly ash is defined as the weight fraction of already reacted fly ash to total fly ash. The average degree of hydration and degree of pozzolanic reaction are affected by the water to cement ratio, concrete temperature, and age. The rate of change of both of them increase with the increase in concrete temperature. Mass concrete temperature changes with time because of the heat of hydration and pozzolanic reaction. Figs. 1 and 2 show the examples of the effect of temperature histories, two constant temperature (30 °C and 60 °C) and a varied

temperature cases, on the degree of hydration and the degree of pozzolanic reaction, respectively. The change in concrete temperature affects the rate of hydration and pozzolanic reaction. The temperature history dependent degree of hydration and degree of pozzolanic reaction were used in the model for predicting autogenous shrinkage and the CTE of pastes. The computed autogenous shrinkage changes according to the temperature change. The details of the degree of hydration and degree of pozzolanic reaction are provided elsewhere<sup>19, 23</sup>.

# **RESULTS AND DISCUSSION**

#### Effect of age

The CTE of paste is clearly a time-dependent property and increases with time (Figs. 3, 4, and 5). This is because the continuity of the paste structure increases with age as a result of increasing hydrated products. Berwanger and Sarkar<sup>24</sup> also reported that CTE decreased with increasing w and increased with age. Yang and Sato<sup>11</sup> reported that CTE of high strength concrete decreased rapidly after setting and reached a minimum value at an age of about 1 day. Thereafter, the CTE increased until an age of up to 7 days, depending on the development of selfdesiccation. Sellevold and Bjøntegaard<sup>6</sup> found that the CTE decreased rapidly after mixing and reached a minimum value after setting. Note that the CTE at the age below 12 h is not included in this study.

# Effect of water to binder ratio

The effect of w on the CTE of paste is not significant (Fig. 3). This is because after the formation of a strong paste structure, water behaves differently from the solid structure. When the temperature is increased, the volume of water in the capillary pores of paste can also increase. However, unlike the solid structure, water can migrate from one capillary pore to other pores, causing an insignificant effect on the thermal expansion of paste.

#### Effect of fly ash

From the experiments, the CTE of cement-fly ash paste increased from the first day and continued to increase in the long term (Fig. 4). The use of a higher replacement percentage of fly ash reduced the CTE of paste, especially at an early age. Unlike in the case of cement-fly ash paste, with cement paste, the CTE is almost constant after 7 days. It was also found that the CTE of cement-fly ash paste was lower than that of the cement paste with the same w (Fig. 5). Wesche<sup>7</sup> also found that the use of fly ash reduced



Fig. 3 Test and predicted results of CTE of cement pastes with w = 0.25, 0.35 and 0.40.



Fig. 4 Test and predicted results of cement-fly ash pastes with r = 0, 0.3 and 0.5, w = 0.25.

the CTE of concrete at a high replacement percentage. Arshad et al<sup>5</sup> found that the CTE of concrete was approximately equal to the volumetrically weighted average of the coefficients of its ingredients. Fly ash has a lower CTE than cement and so using fly ash to replace cement reduces the CTE, especially at early age when the reaction of fly ash is still not active. The use of fly ash delays the reaction of concrete at an early age but continues in the long term. Consequently, the CTE at an early age of cement-fly ash pastes is lower but tends to reach the value of cement-only paste or even higher in the long term.

# Verification of thermal expansion coefficient model of paste

The CTEs of cement, fly ash, quartz sand, and lime-stone are  $14.4\times,~6.45\times,~10.4\times,~and~4.5\times10^{-6}$ 

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Fig. 5 Test and predicted results of cement-fly ash pastes with r = 0, 0.3 and 0.5, w = 0.35.

 $K^{-1}$ , respectively<sup>21,25,26</sup>. By using these values and (6), we can now compare the model for the CTE of paste with the experimental results (Figs. 3, 4, and 5). It is shown that the proposed CTE model is satisfactory for predicting the CTE of the tested cement-fly ash pastes. The model shows that paste with a higher fly ash to binder ratio (r = 0.5) has a lower CTE than that with a lower fly ash to binder ratio (r = 0.3) at an early age but has a higher CTE at a later age. The model also shows that the CTE of paste is time-dependent and increases with age. Fig. 6 shows the comparison between the test results conducted by the authors and the computed results from the model. The model was also verified, as shown in Fig. 6, using the test results conducted by Liwu and  $\dot{\rm Min}^{27}$  where pastes were cast with w=0.3and r = 0, 0.4. The test results after 12 h of age were used in the verification. It was found that the prediction was satisfactory ( $R^2 = 0.88$ ).

Fig. 7 shows examples of the computed CTE of pastes under two constant temperatures,  $30 \,^{\circ}$ C and  $60 \,^{\circ}$ C, and a varied temperature history. It was found that the CTE changes according to temperature change due to differences in the degree of reactions. By using the proposed CTE model of paste with formulae for the CTE of concrete, the overall CTE of mass concrete with varied temperature conditions should be predictable.

#### CONCLUSIONS

After formation of strong paste structure, the CTE of paste is a time-dependent property which increases with age due to higher amounts of hydrated product and higher continuity of the paste structure. The effect of w on CTE of paste is not significant. The use of



**Fig. 6** Comparison between predicted and test results of authors' test results of CTE of cement-fly ash pastes, Liwu and Min's test results of cement-fly ash pastes.



Fig. 7 CTE of cement paste with w = 0.40 at constant temperature (30 °C and 60 °C) and varied concrete temperature.

higher replacement percentage of fly ash reduces the CTE of paste, especially at early age. The CTE of cement-fly ash paste increases from the first day and continues to increase in long term due to continuing pozzolanic reaction. The CTE model of paste was verified to be satisfactory for predicting the CTE of the cement-only and the cement-fly ash pastes.

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