Supplementary material : Study on the effect of amorphous silica from waste granite powder on the strength development of cement-treated clay–stone powder composites

## Chemically bound water content

 $\frac{Chemically bound water =}{\frac{\{m_{0^{\circ}C} (TG value) - m_{105^{\circ}C} (TG value)\}^{-(11.76\% m_{clay} + 1.96\% m_{stone powder}) (TG Value)}{m_{0^{\circ}C} (TG value)} \times 100 [\%]$ (S1)

The TG-DTA results for untreated Tokuyama clay and stone powder indicated mass losses of 11.76% and 1.96% at 0°C–1000°C, respectively These losses for untreated clay and stone powder should be accounted for when calculating the  $Ca(OH)_2$  content of cement-treated clay. If the TG-DTA sample is assumed to have the same composition as that of the prepared sample, the initial mass of the sample before the test is given by

initial sample mass, 
$$m_{0^{\circ}C} = m_{cement} + m_{clay} + m_{stone \ powder} + m_{water}$$
. (S2)

The mass of clay is obtained by substituting the masses of cement, stone powder and water given by Eqs. 15, 16 and 17, respectively, in Eq. 18.

$$m_{cement} = \frac{C^*}{1 - C^*} \times m_{clay},\tag{S3}$$

$$m_{stone\ powder} = \frac{SP}{1-SP} \times m_{clay}, \qquad (S4)$$

 $m_{water} = 1.5LL \times (m_{clay} + m_{stone \ powder}), \tag{S5}$ 

$$m_{clay} = \frac{m_{0} \circ_{C}}{\left[1 + \frac{C^{*}}{1 - C^{*}} + \frac{SP}{1 - SP} + 1.5LL(1 + \frac{SP}{1 - SP})\right]}.$$
(S6)

The values of  $C^*$ , SP and LL are divided by 100.

## Ca(OH)<sub>2</sub> content due to cement hydration

Because TG-DTA detects the water dehydrated from  $Ca(OH)_2$  at 400°C–500°C, the equivalent mass of Ca(OH)<sub>2</sub> was determined by stoichiometry. The dehydroxylation of  $Ca(OH)_2$  produces CaO and water:

$$Ca(OH)_2 \to CaO + H_2O. \tag{S7}$$

By stoichiometry, the equivalent mass of Ca(OH)<sub>2</sub> can be calculated while considering the clay loss:

$$Ca(OH)_2 = \frac{Molar \ mass \ of \ Ca(OH)_2}{Molar \ mass \ of \ H_2O} \times \ mass \ of \ H_2O - (2.448\% m_{clay} + 1.45\% m_{clay})$$

$$0.308\%m_{stone\ powder})\ (TG\ Value),\tag{S8}$$

where the molar masses of Ca(OH)<sub>2</sub> and H<sub>2</sub>O are 74.092 and 18.015 g/mol, respectively.

The TG-DTA results for untreated Tokuyama clay indicated mass losses of 2.448% and 0.308% at 400°C–500°C and 600°C–800°C, respectively.

## 4.2.2 CaCO<sub>3</sub> content

TG-DTA detects the mass of  $CO_2$  decomposed from CaCO<sub>3</sub> at 600°C–800°C. The equivalent mass of CaCO<sub>3</sub> must be determined by stoichiometry. The decomposition of CaCO<sub>3</sub> produces CaO and CO<sub>2</sub>:

$$CaCO_3 \to CaO + CO_2. \tag{S9}$$

By stoichiometry, the equivalent mass of CaCO<sub>3</sub> is

Mass of 
$$CaCO_3 = \frac{Molar mass of CaCO_3}{Molar mass of CO_2} \times mass of CO_2 - (1.480\% m_{clay} + 1.480\% m_{clay})$$

$$0.243\% m_{stone \ powder})(TG \ Value), \tag{S10}$$

where the molar masses of CaCO<sub>3</sub> and CO<sub>2</sub> are 100.09 and 44.01 g/mol, respectively.

## $4.2.3 Ca(OH)_2$ consumption by carbonation

 $Ca(OH)_2$  is consumed by carbonation to produce  $CaCO_3$  and water:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O. \tag{S11}$$

The amount of Ca(OH)<sub>2</sub> consumed by carbonation is determined with stoichiometry:

$$Ca(OH)_2 = \frac{Molar \ mass \ of \ Ca(OH)_2}{Molar \ mass \ of \ CaCO_3} \times \ mass \ of \ CaCO_3.$$
(S12)