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Differential scanning calorimetry evaluation of autoclaved cement based building materials made with construction and demolition waste

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Abstract

The effect of adding finely ground construction and demolition (C&D) waste fines, containing a significant amount of clay-brick, to mixtures of Portland cement and quartz sand autoclaved at 180 °C was investigated by DSC. The incorporation of up to 41.25 mass% C&D waste fines enhanced the formation of aluminium-substituted tobermorite indicating that the mullite in the clay-brick fines is reactive under autoclaving conditions and supplied the aluminium required for the formation of this critical phase. C&D waste fine additions in excess of 41.25 mass% promoted the formation of aluminium-substituted C–S–H[1] over aluminium-substituted tobermorite. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Autoclaving; DSC; Tobermorite; Construction and demolition waste; Mullite

1. Introduction

In recent years there has been a great deal of interest and support worldwide in using construction and demolition (C&D) waste for the production of aggregate material which can then be reused for road base courses and other engineering applications [1,2]. In Australia, in particular in the Sydney metropolitan area, this type of aggregate material is comprised of concrete and mortar but also contains a significant amount of clay-brick (between 15 and 40 mass%) [1]. More importantly however a substantial amount of fine material, enriched with clay-brick, is generated during the crushing process of C&D waste. Preliminary work by the authors has demonstrated that the formation of 1.1 nm tobermorite,[2] the principal binder of most autoclaved calcium silicate based building materials, is enhanced by the addition of finely ground clay-brick to mixtures of Portland cement and quartz sand [3]. The objective of the current work is to study the effect of finely ground C&D waste additions to mixtures of Portland cement and quartz sand on 1.1 nm tobermorite formation. In this article we report DSC data which, in combination with XRD have been employed effectively in evaluating the effects of finely ground C&D waste additions on the formation of 1.1 nm tobermorite.

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[1] Cement chemistry nomenclature: C = CaO, H = H_2O, S = SiO_2.

[2] The limiting compositions of 1.1 nm tobermorite are $Ca_4Si_6O_{15}(OH)_2{\cdot}4H_2O$ to $Ca_5Si_{5.5}O_{15}(OH)_2{\cdot}4H_2O$.

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2. Experimental

The following raw materials were used.

(a) Type A Portland cement containing: SiO_2 (21.1%), CaO (64.4%), Al_2O_3 (4.15%), Fe_2O_3 (4.60%), SO_3 (2.60%), MgO (1.14%), TiO_2 (0.22%), produced by Blue Circle Southern Portland Cement, Berrima Works, NSW, Australia. Blaine surface area is 3550 cm^2/g.

(b) Ground quartz sand (quartz) containing: SiO_2 (99.1%), Al_2O_3 (0.36%), Fe_2O_3 (0.10)%, TiO_2 (0.07%) from Ballarat, Victoria, Australia and supplied by Commercial Minerals, Sydney, Australia. Blaine surface area is 3600 cm^2/g.

(c) C&D fines less than 6 mm, were generated by a recycling plant in the Sydney metropolitan area. This material was ground by dry ball milling to a Blaine surface area comparable to that of the quartz. The chemical composition of C&D waste fines, as determined by XRF is given in Table 1.

A control OPC–quartz mixture was prepared with 45 mass% OPC and 55 mass% quartz. C&D waste fines were introduced at 13.75, 27.5, 41.25 and 55 mass% as quartz replacement. The water to total solids ratio was 0.36 as this yielded comparable workability for all the mixtures. Mechanical mixing was conducted in accordance with ASTM C 305-082. Pastes were cast into stainless steel moulds and consolidated on a vibratory table, followed by a 24 h curing period in a moist cabinet. Demoulded specimens were autoclaved for 8 h, 6 h of which were at 180 °C under saturated steam. After autoclaving, samples were oven dried at around 105 °C overnight followed by disc milling. Powdered samples were examined by DSC using a SETARAM SetSys 1750 balance with DSC/TGA rod 1500 °C at a heating rate of 10 °C/min under flowing air (20 ml/min) from 20 to 1100 °C. Sample masses were between 15 and 20 mg and were packed into a Pt crucible with 20 taps. All curves were evaluated using the SETARAM software. X-ray diffraction (XRD) analysis was carried out on powdered specimens using a Siemens D5000 and Cu $K\alpha_1$ radiation from 3 to 50 ° 2θ at 0.02 ° 2θ s^{-1}. XRD patterns were $K\alpha_2$ stripped and corrected for instrumental drift.

3. Results and discussion

DSC curves of autoclaved specimens are shown in Fig. 1. The main observed endotherms (endo) and exotherms (exo) are ascribed in the following discussion to:

ca. 60–260 °C (endo)	Presence of calcium silicate hydrates including 1.1 nm tobermorite
ca. 260–400 °C (endo)	Presence of a member of the hydrogarnet series, $C_3AS_{3-x}H_{2x}$, $x = 0$–3
ca. 573 °C (endo)	Crystalline inversion due to unreacted quartz
ca. 600–700 °C (endo)	Decarbonation of calcium carbonate
ca. 850–890 °C (exo)	Crystallization of beta-wollastonite (β-CS) from calcium silicate hydrates including 1.1 nm tobermorite

Table 1
Major oxides and loss on ignition (LOI)[a] of C&D waste fines used in this study

Major oxides	%
SiO_2	75.2
Al_2O_3	8.7
CaO	5.9
Fe_2O_3	3.4
K_2O	1.2
LOI	5.9

[a] LOI: loss on ignition at 1050 °C.

The variation in the β-CS formation temperature versus C&D waste fine content, representing increasing replacement for quartz in the raw mix, is shown in Fig. 2. The variation in the β-CS formation temperature versus the total bulk Al_2O_3 content in the initial raw mix is depicted in Fig. 3.

When compared with the control sample (curve 1), the DSC curves of specimens made with C&D waste fines (curves 2–5) manifest the following (Fig. 1).

(i) Increasing amounts of Al were incorporated into the 1.1 nm tobermorite crystal lattice with C&D waste fine additions of up to 41.25 mass%. This

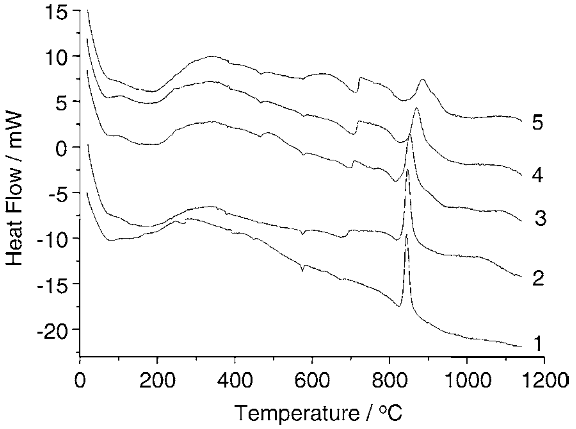

Fig. 1. DSC curves of autoclaved specimens made with: (1) 0; (2) 13.75; (3) 27.5; (4) 41.25 and (5) 55 mass% C&D waste fines (bottom to top).

is evident from the changes in the exotherm profile above 800 °C due to the crystallization of β-CS (Fig. 1) exhibiting an increase in the β-CS formation temperature (Fig. 2), a reduction in the height and the broadening of the exotherm around 840 to 870 °C. Previous work established that with increasing Al content into the 1.1 nm tobermorite crystal lattice, the β-CS formation temperature increases and the exotherm becomes flatter and broader [4,5].

(ii) The increase in the β-CS formation temperature (Figs. 1 and 2) correlates well with the increase in the total Al_2O_3 content in the initial raw mix (Fig. 3).

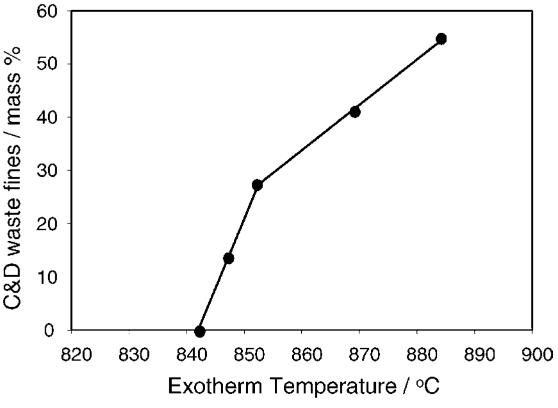

Fig. 2. Variation in the β-CS formation temperature with increasing C&D waste fine addition.

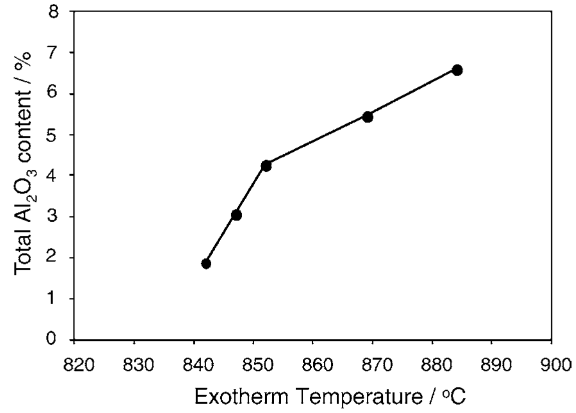

Fig. 3. Variation in the β-CS formation temperature vs. the total bulk Al_2O_3 content in the initial raw mix.

(iii) The incorporation of C&D waste fines in excess of 41.25 mass% promoted the formation of Al-substituted C–S–H (Al-(C–S–H)) (Figs. 1 and 2).

In relation to the thermal behaviour of Al-(C–S–H), in particular, the β-CS formation temperature, previous work has established that this exothermic peak occurs predominantly around 880–900 °C (Fig. 4) [6]. Results obtained by XRD confirmed the above observation. A more detailed account of the exotherm profiles in relation to C–S–H and 1.1 nm tobermorite containing Al may be found elsewhere [6,7].

In view of the fact that the C&D waste fines used in this study contained a substantial amount of clay-brick, with mullite ($3Al_2O_3{\cdot}2SiO_2$) as the predominant Al-containing phase, is it apparent that mullite is a reactive phase under hydrothermal conditions and supplied the Al required for the formation of Al-tobermorite for C&D waste fine additions of up to 41.25 mass%. In contrast, the incorporation of C&D waste fines in excess of 41.25 mass% promoted the formation of Al-(C–S–H) (Figs. 1 and 4). A possible explanation is as follows: Klimesch et al. [8] have shown previously that the formation of Al-(C–S–H), more specifically Si-rich Al-(C–S–H), is favoured when using excess amounts of reactive alumino–silicate based raw materials. In other words, as the amount of C&D waste fines, containing clay-brick fines, was increased from 13.25 to 55 mass%, the bulk Ca/(Al + Si) ratio increased only slightly from 0.49 to 0.59 while the bulk Al/(Al + Si) ratio increased substantially from 0.059 to 0.134 and

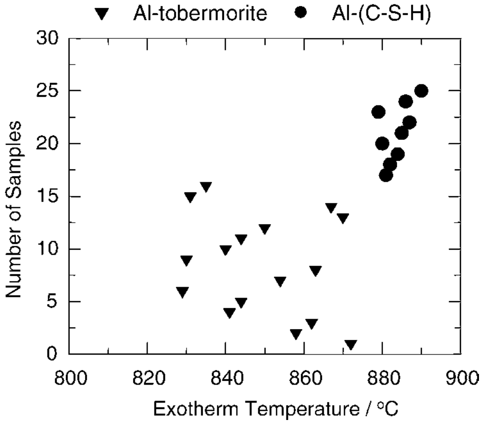

Fig. 4. Observed β-CS formation temperatures of Al-tobermorite and Al-(C–S–H) incorporating different levels of Al. Compiled from data by Klimesch [6].

the formation of a Si-rich Al-(C–S–H) appears to have been favoured. It is well known that this type of C–S–H impedes the formation of Al-tobermorite [9]. In addition, it may be noted that the total bulk composition, i.e. total bulk Ca/(Al + Si) and Al/(Al + Si), does not equal the total reacted composition, since some unreacted quartz and/or C&D waste fines remained, which is evident from Fig. 1. It is also noteworthy that the amount of calcium carbonate increased with increasing C&D waste fine addition. The calcium carbonate was already present in the C&D waste material and originated mainly from the carbonation of building products including concrete and mortar.

4. Conclusions

1. Replacing quartz sand with C&D waste fines, containing clay-brick fines, for the production of hydrothermally cured calcium silicate based materials is a viable option for the future.
2. The incorporation of up to 41.25 mass% C&D waste fines enhanced the formation of Al-substituted tobermorite indicating that the mullite in the clay-brick fines is reactive under hydrothermal conditions and supplied the Al required for the formation of Al-substituted tobermorite.
3. C&D waste fine additions in excess of 41.25 mass% promoted the formation of Al-(C–S–H).
4. DSC manifested the presence of Al-(C–S–H) more definitively than XRD.

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