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# ***Thermal Analysis of Cement Components Hydrated in the Presence of Calcium Carbonate***

by V.S. Ramachandran

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## RÉSUMÉ

On a hydraté du silicate tricalcique, de l'aluminate tricalcique et de l'aluminate tricalcique + gypse en présence de 5-50 % de  $\text{CaCO}_3$  pendant différentes périodes de temps (quelques minutes à 28 jours) et on a examiné les produits par calorimétrie différentielle à balayage (CDB), thermogravimétrie différentielle (TGD), analyse thermique différentielle (ATD) et calorimétrie par conduction.

L'hydratation du silicate tricalcique a été accélérée par le  $\text{CaCO}_3$  et on a constaté l'incorporation de  $\text{CaCO}_3$  dans la phase silicate de calcium hydraté. Lors de l'hydratation de l'aluminate tricalcique, on a identifié du complexe carboaluminate et on observé que la formation de la phase aluminat de calcium hydraté cubique était retardée. Dans le système aluminat tricalcique +  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  +  $\text{CaCO}_3$  +  $\text{H}_2\text{O}$ , la vitesse de formation d'ettringite (forte teneur en sulfoaluminate de calcium hydraté) et sa conversion au monosulfoaluminate hydraté ont été accélérées par le  $\text{CaCO}_3$ . En se basant sur ces résultats, on a proposé un mécanisme pour expliquer l'effet du  $\text{CaCO}_3$  sur l'hydratation du ciment.

## THERMAL ANALYSES OF CEMENT COMPONENTS HYDRATED IN THE PRESENCE OF CALCIUM CARBONATE

V.S. RAMACHANDRAN

*Building Materials Section, Institute for Research in Construction, National Research Council, Montreal Road, Ottawa, Ontario K1A 0R6 (Canada)*

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### ABSTRACT

Tricalcium silicate, tricalcium aluminate and tricalcium aluminate + gypsum were hydrated in the presence of 5–50%  $\text{CaCO}_3$  for different lengths of time (a few minutes to 28 days) and the products were examined by differential scanning calorimetry (DSC), differential thermogravimetry (DTG), differential thermal analysis (DTA) and conduction calorimetry.

The hydration of tricalcium silicate was accelerated by  $\text{CaCO}_3$  and there was evidence of incorporation of  $\text{CaCO}_3$  into the calcium silicate hydrate phase. In the hydration of tricalcium aluminate, carboaluminate complex was identified and the formation of the cubic calcium aluminate hydrate phase was found to be retarded. In the tricalcium aluminate +  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{CaCO}_3 + \text{H}_2\text{O}$  system the rate of formation of ettringite (high calcium sulphaaluminate hydrate) and its conversion to the monosulphaaluminate hydrate was accelerated by  $\text{CaCO}_3$ . On the basis of these results a mechanism has been proposed to explain the effect of  $\text{CaCO}_3$  on the hydration of cement.

### INTRODUCTION

In concrete technology, limestone is used for various purposes, namely, as raw material for clinker production, for partial replacement for gypsum and as coarse or fine aggregate. Recently, limestone dust produced in quarrying operations has been suggested for use as an additive to portland cement. Because of the concern that properties and durability of concrete may be affected, addition of only a small amount of limestone is permitted in some standards. Even in the limited amounts of work carried out on the effect of limestone on cement and concrete properties, there is divergence of opinion on the relative effects of different amount of calcium carbonate [1]. Variations in the nature of calcium carbonate and cement, the conditions of hydration, interpretation of results and limitations of techniques adopted to measure the hydration properties may account for this disagreement.

A better understanding of the effect of limestone addition on cement may be obtained by first studying its effect on the individual components of cement and then extending this knowledge to cement itself. Such an ap-

proach has been applied successfully to explain the effect of calcium lignosulphonate on the hydration of cements [2].

Thermal analysis techniques such as differential thermal analysis (DTA), differential thermogravimetry (DTG), differential scanning calorimetry (DSC) and conduction calorimetry have been applied successfully to investigate the role of admixtures and additives in cement [3]. The objective of this study was to apply thermal analysis techniques to gain a better understanding of the effect of calcium carbonate ( $\overline{CC}$ ) on the hydration of individual components of cements, namely, tricalcium silicate ( $C_3S$ ), tricalcium aluminate ( $C_3A$ ) and tricalcium aluminate + gypsum ( $C_3A + \overline{CSH}_2$ ) \*.

## EXPERIMENTAL

### *Materials*

The following cement components were used: tricalcium silicate, with a Blaine surface area of  $3600 \text{ cm}^2 \text{ g}^{-1}$  supplied by the Portland Cement Association, tricalcium aluminate with a  $N_2$  specific surface area of  $0.5 \text{ m}^2 \text{ g}^{-1}$  supplied by Tetrattech Co., and gypsum of reagent quality obtained from Fisher Scientific Co.

Three types of  $\overline{CC}$  were used: (a) that obtained from Anachemica Chemical Co. was designated Ac  $\text{CaCO}_3$  and had a surface area of  $6.5 \text{ m}^2 \text{ g}^{-1}$  with particles in the size range  $< 10 \text{ } \mu\text{m}$ ; (b) that supplied by Fisher Scientific Co. was designated Fr  $\text{CaCO}_3$  and had a surface area of  $0.4 \text{ m}^2 \text{ g}^{-1}$  with particles in the size range  $< 25 \text{ } \mu\text{m}$ ; (c) four samples were obtained by grinding pure limestone to different particle size ranges. Particles were in the size ranges,  $< 10 \text{ } \mu\text{m}$ ,  $10\text{--}20 \text{ } \mu\text{m}$ ,  $20\text{--}45 \text{ } \mu\text{m}$  and  $74\text{--}149 \text{ } \mu\text{m}$  and were designated Ls  $< 10 \text{ } \mu\text{m}$ , Ls  $10\text{--}20 \text{ } \mu\text{m}$ , Ls  $20\text{--}45 \text{ } \mu\text{m}$  and Ls  $74\text{--}149 \text{ } \mu\text{m}$ , respectively.

### *Methods*

The following techniques were adopted. DSC and DTG curves were obtained using a Du Pont 1090 thermal analysis system. In the DSC mode each sample was heated up to  $600^\circ\text{C}$  and in the DTG mode up to the maximum temperature of  $1200^\circ\text{C}$ . DTA was used only on the  $C_3S$  systems for the estimation of  $C_3S$ . In each experiment a 30 mg sample was run at a

\* Cement Chemistry Notation:  $\overline{CC}$  = calcium carbonate;  $3\text{CaO} \cdot \text{SiO}_2 = C_3S$ ;  $3\text{CaO} \cdot \text{Al}_2\text{O}_3 = C_3A$ ;  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} = \overline{CSH}_2$ ; calcium silicate hydrate =  $C\text{--}S\text{--}H$  or  $\text{CSH}$ ;  $\text{Ca}(\text{OH})_2 = \text{CH}$ ; hexagonal calcium aluminate hydrate =  $(C_2\text{AH}_x\text{--}C_4\text{AH}_y)$ .

heating rate of  $20^{\circ}\text{C min}^{-1}$  in a flow of  $\text{N}_2$  gas. The rate and amount of heat developed by the sample in the first 24 h was measured by a conduction calorimeter supplied by the Institute of Applied Physics, Delft, Holland.

### *Sample preparation*

The cement components containing 0, 5, 10, 15 and 50%  $\text{CC}$  were prepared at a water/solid ratio of 0.4 or 4.0 as follows. The solid mixture was transferred to a polyethylene vial. Glass spheres 5 mm in diameter were added to the mixture and vibrated on a spex high speed mixer for 20 min. A required amount of carbon dioxide-free distilled water was added and the mixture was capped and vibrated for 20 s. Samples hydrated for periods less than 7 h were subjected to cold methanol treatment and then dried for 24 h in vacuo. To obtain samples hydrated for longer periods, the polyethylene vial was placed in a plexiglas tube, put on rollers and rotated for different periods. At a specific interval of time the sample in the vial was dried by continuous vacuum pumping for one day and subjected to analysis.

The water/solid ratio used for the  $\text{C}_3\text{S}$  system was 0.4 or 0.7, whereas that for the  $\text{C}_3\text{A}$  or  $\text{C}_3\text{A} + \text{gypsum}$  systems was 4.

## RESULTS AND DISCUSSION

### *$\text{C}_3\text{S}-\text{CaCO}_3-\text{H}_2\text{O}$ system*

The DTG curves of  $\text{C}_3\text{S}$  hydrated in the presence of different amounts of  $\text{CaCO}_3$  indicated three inflections at  $100-120^{\circ}\text{C}$ ,  $460-475^{\circ}\text{C}$  and  $730-740^{\circ}\text{C}$ . They were caused by the loss of water from the C-S-H phase, dehydration of CH and decomposition of  $\text{CC}$ , respectively. The intensity of the CH peak may be used to follow the kinetics of hydration of  $\text{C}_3\text{S}$ . Figure 1 shows the amount of CH formed at different times during the hydration of  $\text{C}_3\text{S}$  in the presence of 0, 5 and 15%  $\text{CC}$ . Results based on the total amount of the paste ( $\text{C}_3\text{S} + \text{CSH}$  and  $\text{CC}$ ) indicate that the addition of  $\text{CC}$  accelerates the hydration of  $\text{C}_3\text{S}$  at ages up to 7 days (Fig. 1a). The values calculated on the basis of  $\text{C}_3\text{S}$  (to avoid dilution effects) demonstrate that addition of  $\text{CC}$  increases the amount of CH at all hydration times (Fig. 1b). The accelerating effect is greater for larger additions of  $\text{CC}$  and is particularly significant at short hydration times. At 28 days, the addition of 0, 5 and 15%  $\text{CC}$  yields 23.2, 24.0 and 25.5% CH, respectively; the corresponding values at 1 day are about 9, 11 and 14%, respectively.

The DTG technique was also applied to determine the effect of  $\text{CC}$  on the non-evaporable content of the pastes hydrated for various times. The non-evaporable water content, a measure of the degree of hydration, refers

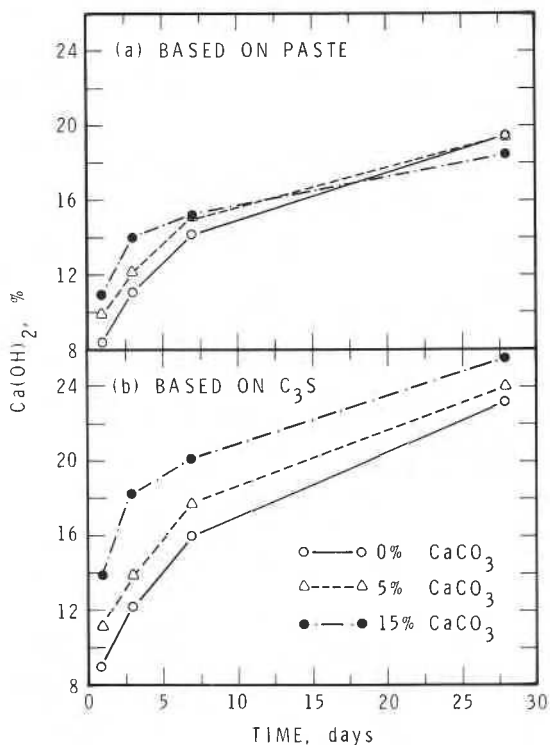


Fig. 1. Formation of  $\text{Ca(OH)}_2$  in  $\text{C}_3\text{S}$  pastes containing different amounts of  $\text{Ac CaCO}_3$ .

to the amount of chemically combined water in the C-S-H and CH phase. Results similar to those based on CH determination were obtained.

The degree of hydration may also be followed by estimating the amount of  $\text{C}_3\text{S}$  remaining unhydrated at different times. Ramachandran [4] has proposed a DTA method in which the endothermal peak area of the triclinic-monoclinic transition of  $\text{C}_3\text{S}$  occurring at  $915^\circ\text{C}$  is determined. In

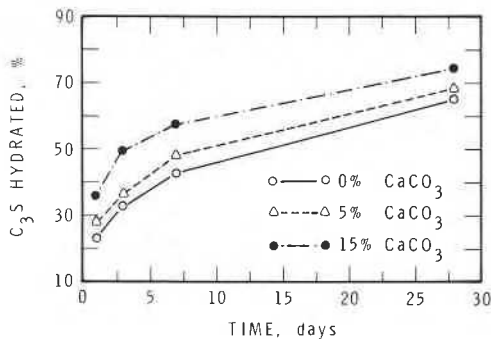


Fig. 2. Degree of hydration of  $\text{C}_3\text{S}$  in the presence of  $\text{CaCO}_3$ .

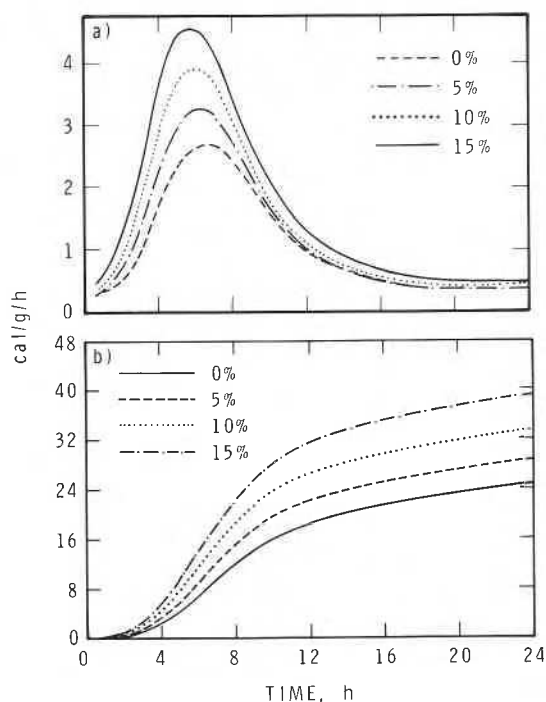


Fig. 3. Conduction calorimetric curves of  $\text{C}_3\text{S}$  containing different amounts of  $\text{CaCO}_3$ : (a) rate of heat produced; (b) the amount of heat produced at different times.

Fig. 2 the effect of 0, 5 and 15%  $\text{CC}$  on the degree of hydration of  $\text{C}_3\text{S}$  is plotted as a function of time. The substantial accelerating effect of  $\text{CC}$  is confirmed at short hydration times. The results show similar trends to those based on CH and non-evaporable water content determinations.

It is tacitly assumed in the literature that  $\text{CC}$  does not influence the rate of hydration of  $\text{C}_3\text{S}$ . In order to confirm the unexpected accelerating effect of  $\text{CC}$  on the hydration of  $\text{C}_3\text{S}$ , a few more experiments were conducted using conduction calorimetry. The rate of evolution of heat and the amount of heat produced in 1 day in the presence of 0, 5, 10 and 15%  $\text{CC}$  were determined (Fig. 3). The intensity of the peak occurring over a range of about 6–7 h, representing the degree of hydration of  $\text{C}_3\text{S}$ , increases as the amount of added  $\text{CC}$  increases. The integral heat developed in the mixes also shows the accelerating effect of  $\text{CC}$ .

The rate of hydration of  $\text{C}_3\text{S}$  not only depends on the amount, but also on the particle size, of added  $\text{CC}$ . It is evident that the larger the particle size the lower the accelerating effect (Fig. 4). Pronounced differences in acceleration effects are indicated especially at short hydration times.

Ramachandran has proposed a theory for the acceleration of hydration of  $\text{C}_3\text{S}$  in the presence of  $\text{CaCl}_2$ . A complex formation between the hydrating  $\text{C}_3\text{S}$  and  $\text{CaCl}_2$  is envisaged [2]. If such a phenomenon were also to occur in



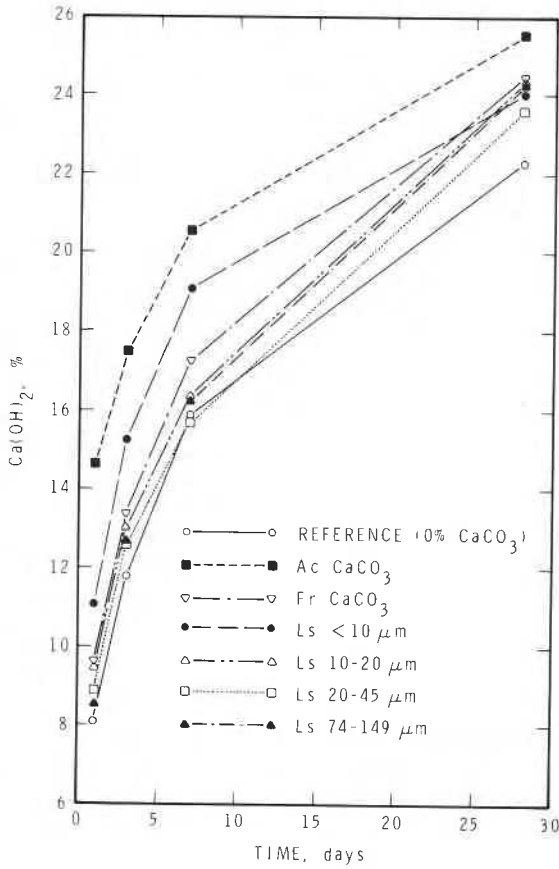


Fig. 4. Effect of particle size of  $\text{CaCO}_3$  on the hydration of  $\text{C}_3\text{S}$ .

the hydration of  $\text{C}_3\text{S}$  in the presence of  $\text{CC}^-$ , a complex should form between  $\text{CC}^-$  and the hydrating  $\text{C}_3\text{S}$ . The DTG method was adopted to determine if the amount of  $\text{CC}^-$  added initially decreased as the hydration progressed. The

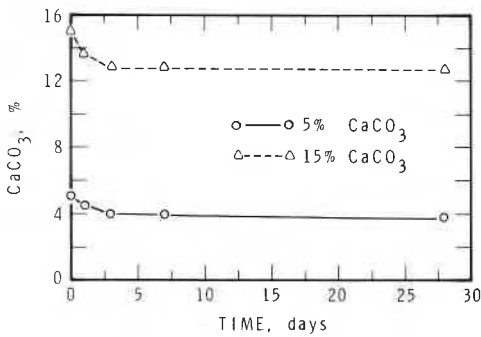


Fig. 5. Amount of free  $\text{CaCO}_3$  remaining in the hydration of  $\text{C}_3\text{S}$  admixed with 5% or 15%  $\text{CaCO}_3$ .

DTG peak due to  $\text{CaCO}_3$  diminished as hydration proceeded, indicating that some  $\text{CC}$  was consumed during hydration. In the  $\text{C}_3\text{S}-\text{CC}$  mixture hydrated with 15%  $\text{CC}$  and 5%  $\text{CC}$ , the amounts of  $\text{CC}$  that reacted with the hydrating  $\text{C}_3\text{S}$  were 2.38 and 1.34% respectively in 28 days (Fig. 5). Most  $\text{CaCO}_3$  reacted within 3 days. X-ray diffraction (XRD) could not identify the type of complex formed between the hydrating  $\text{C}_3\text{S}$  and  $\text{CC}$ , possibly because it is amorphous and present only in small amounts.

### $\text{C}_3\text{A}-\text{CaCO}_3-\text{H}_2\text{O}$ system

DSC of the  $\text{C}_3\text{A}-\text{H}_2\text{O}$  system shows endothermal effects even at 5 min, signifying the presence of hydration products (Fig. 6a). Peaks at about 145–150°C and 265–280°C represent the presence of metastable hexagonal phases ( $\text{C}_2\text{AH}_x-\text{C}_4\text{AH}_y$ ). The endothermal effect at about 150°C increases up to 4 h and that at 265–300°C continues to increase up to 2 days. The large peak at about 300°C (4 h to 2 days) is mainly due to the presence of cubic phase ( $\text{C}_3\text{AH}_6$ ). The exact time of conversion of the hexagonal to the cubic phase is not easy to determine because they co-exist and their peaks overlap. Both DSC and XRD results indicate that at 7 days and after, the major component in this system is the cubic phase.

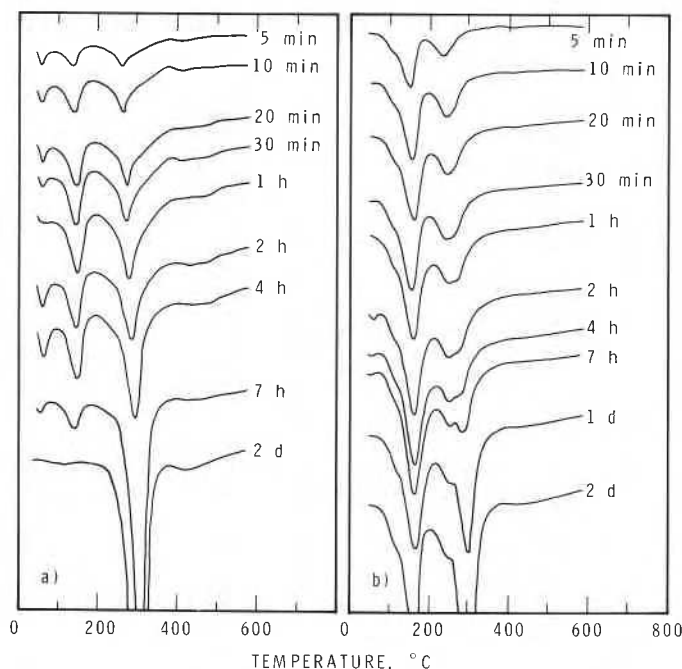


Fig. 6. DSC curves of  $\text{C}_3\text{A}$  hydrated for different periods: (a) 0%  $\text{CaCO}_3$ ; (b) 12.5%  $\text{CaCO}_3$ .

In the  $C_3A$ - $CaCO_3$ - $H_2O$  system (Fig. 6b), at short hydration times, similar effects to those observed in Fig. 6a are evident, although the peaks are more intense. These peaks however, represent calcium carboaluminate or a solid solution [3]. No hexagonal phase of the type  $(C_2AH_x-C_4AH_y)$  could be identified by XRD in this system. A small amount of the cubic phase  $C_3AH_6$  ( $260-280^\circ C$ ) formed at 2 h intensifies as hydration progresses. At 7 h this peak is of much lower intensity than that formed in the  $C_3A$ - $H_2O$  system (Fig. 6a). Thus, it can be concluded that the addition of CC of  $C_3A$  retards the formation of the cubic phase. The presence of carboaluminate complex is identified at 2 days by a peak at about  $160^\circ C$ .

#### $C_3A$ - $C\bar{S}H_2$ - $CaCO_3$ - $H_2O$ system

In this system, when no  $CaCO_3$  is present, a small endothermal effect at about  $100^\circ C$  and another of larger intensity at about  $130^\circ C$  appear after only 5 min (Fig. 7a). The first small peak intensifies as the hydration progresses (up to 1 h) and represents the presence of ettringite of formula  $C_3A \cdot 3C\bar{S} \cdot 31H_2O$ . The second peak caused by the dehydration of  $C\bar{S}H_2$  decreases with time because of its progressive interaction with  $C_3A$ . At 2 h the two main peaks at about  $100^\circ C$  and  $165-180^\circ C$  are indicative of the

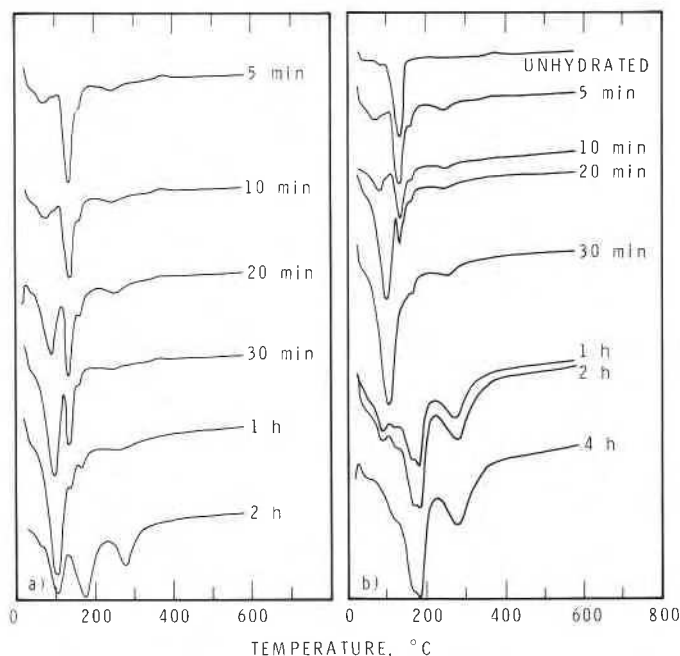


Fig. 7. DSC curves of  $C_3A$ + $C\bar{S}H_2$  hydrated in the presence of  $CaCO_3$ : (a)  $C_3A$ +25%  $C\bar{S}H_2$ ; (b)  $C_3A$ +25%  $C\bar{S}H_2$ +12.5%  $CaCO_3$ .

presence of ettringite and monosulphoaluminate hydrate ( $C_3A \cdot \bar{C}\bar{S} \cdot 11H_2O$ ), respectively. The decrease in the ettringite peak after 1 h is due to the partial conversion of ettringite to the monosulphoaluminate phase. These results are confirmed by XRD. A comparison of Figs. 7a and 7b shows that gypsum is consumed more rapidly (within 20 min) in the mixture containing  $CaCO_3$ . A comparison of the curves at 30 min also suggests that the formation of ettringite is accelerated by the addition of 12.5%  $\bar{C}\bar{C}$  to the  $C_3A + 25\% \bar{C}\bar{S}H_2$  mixture. Conversion of ettringite to monosulphoaluminate is also accelerated by incorporation of  $\bar{C}\bar{C}$ . In the absence of  $\bar{C}\bar{C}$  practically no monosulphoaluminate is formed at about 1 h (Fig. 7a), whereas at the same period in the presence of  $\bar{C}\bar{C}$  a substantial amount of aminosulphoaluminate is identified (Fig. 7b). XRD and scanning electron microscopic investigation indicated similar interconversion effects in this system.

#### ROLE OF CALCIUM CARBONATE IN CEMENTS

The application of thermal analysis techniques has shown that the hydration of  $C_3S$  is accelerated especially in the presence of  $CaCO_3$  with a large surface area. XRD and microstructural investigations also confirmed this observation.

It was evident that at short hydration times some  $CaCO_3$  is incorporated into the C-S-H phase. If this complex occurs on the surface of  $C_3S$  it could enhance the formation of a permeable C-S-H membrane, or this phase might itself be a nucleating agent for the formation of hydration products. Either possibility would result in an acceleration of  $C_3S$  hydration. In addition, microstructural studies showed the presence of hydrated products (C-S-H and CH) on  $CaCO_3$  particles. This suggests that carbonate particles act as nuclei. Such growth of CH and C-S-H would decrease the concentration of Ca and Si ions in the solution phase and promote their transfer from the  $C_3S$  phase to the solution phase.

In the  $C_3A$ -gypsum- $CaCO_3$ - $H_2O$  system the accelerated formation of ettringite and its conversion to the monosulphoaluminate may be visualised as follows. In this system the  $CaCO_3$  particles ( $6.5 \text{ m}^2 \text{ g}^{-1}$ ) are much finer than those of  $C_3A$  or gypsum (approximately  $0.5 \text{ m}^2 \text{ g}^{-1}$ ). The  $CaCO_3$  particles surround the  $C_3A$  and gypsum particles and on contact with water, react to form carboaluminate complex on the surface of  $C_3A$  particles. This layer may be less compact and more permeable than the sulphoaluminate layer normally occurring in the  $C_3A$ -gypsum system. Thus the interaction of Ca, Al and  $SO_4$  ions is enhanced in the presence of  $CaCO_3$ . The conversion of ettringite to monosulphoaluminate hydrate takes place after gypsum is exhausted. Calcium carboaluminate, by enhancing the consumption of gypsum, also facilitates conversion of ettringite to monosulphoaluminate.

The effect of  $\text{CaCO}_3$  on the  $\text{C}_3\text{S}$  and  $\text{C}_3\text{A} + \text{gypsum}$  systems should be reflected in the hydration behaviour of cement. The acceleration effects should produce early setting, and early strength development in cements. The formation of complexes would be expected to influence the durability of cements.

## CONCLUSIONS

High surface area  $\text{CaCO}_3$  not only accelerates the hydration of tricalcium silicate but also forms a complex with its hydrated products. Addition of  $\text{CaCO}_3$  to the tricalcium aluminate–water system results in the formation of carboaluminate hydrate that retards the formation of cubic aluminate hydrate. The normal mode of hydration of tricalcium aluminate that occurs in the presence of gypsum is modified by  $\text{CaCO}_3$ . The rate of ettringite formation and its conversion to the monosulphoaluminate is accelerated by  $\text{CaCO}_3$ . The complex formation of  $\text{CaCO}_3$  with the hydrating components of cements can explain its influence on these components in the cement system (acceleration or retardation).

## ACKNOWLEDGEMENTS

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