



Advances in understanding hydration of Portland cement



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ABSTRACT

Progress in understanding hydration is summarized. Evidence supports the geochemistry dissolution theory as an explanation for the induction period, in preference to the inhibiting layer theory. The growth of C–S–H is the principal factor controlling the main heat evolution peak. Electron microscopy indicates that C–S–H “needles” grow from the surface of grains. At the peak, the surface is covered, but deceleration cannot be attributed to diffusion control. The shoulder peak comes from renewed reaction of C₃A after depletion of sulfate in solution, but release of sulfate absorbed on C–S–H means that ettringite continues to form. After several days space becomes the major factor controlling hydration.

The use of new analytical technique is improving our knowledge of the action of superplasticizers and leading to the design of molecules for different applications. Atomistic modeling is becoming a topic of increasing interest. Recent publications in this area are reviewed.

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1. Introduction

In this paper we summarize progress in understanding the hydration of Portland based cements since the Madrid conference in 2011 [1]. The aim is to highlight the most important contributions to the field rather than give an exhaustive list of everything published in the

area. In addition to published materials, we add input from discussions of a Nanocem workshop attended by many leading researchers in the field from Europe and North America [2].

Nowadays the overwhelming majority of cement sold outside North America consists of Portland cement clinker blended with supplementary cementitious materials (SCM). Nevertheless, early age strength is still mainly due to the reaction of the clinker phases. It is therefore important to understand this reaction, which may also be modified by the physical presence of the SCM (so called filler effect discussed in more detail below). Later on the SCMs themselves

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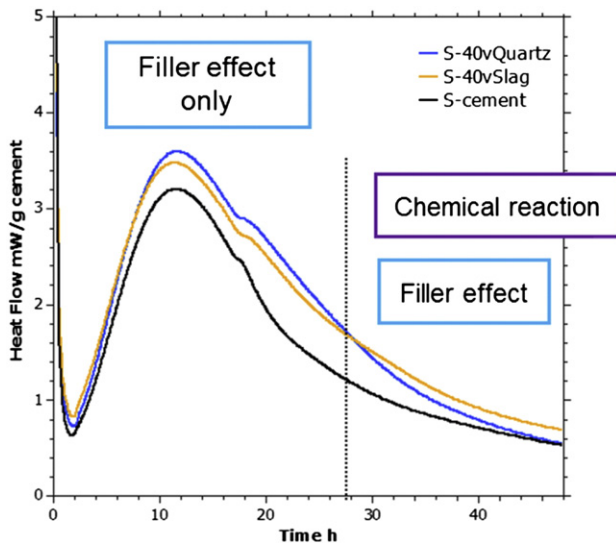


Fig. 1. Rate of heat evolution normalized to clinker content for a Portland cement and the same cement substituted 40% by quartz or by slag. From [4].

react and contribute to the hydration process. Consideration of the available options [3] shows that most cementitious materials used are likely to continue to be based on Portland cement clinker for the foreseeable future.

In this paper first the reaction of plain Portland cement and blended systems is compared. Then we discuss the hydration of the Portland cement clinker, focusing on the three main periods of reaction of alite/ C_3S :

- Up to the end of the induction period (about 3 h¹)
- Main heat evolution peak (approx. 3–24 h), including impact of fillers
- Continuing reaction after 1 day (particularly from 1 to 28 days.)

This is followed by a discussion of the impact of the aluminate containing phases and perspectives for microstructural modeling.

In the second part of the paper work on the impact of admixtures is discussed, followed by a discussion on atomistic level modeling.

1.1. Comparison of hydration kinetics between pure Portland systems and systems with SCMs

In blended (or composite) cements there is generally negligible reaction of the SCM in the first day or so. This is nicely illustrated in Fig. 1 from Berodier [4]. Here the rate of heat evolution is shown as a function of the

clinker phases for 3 systems: a pure Portland system, a blend of the same Portland with 40% quartz (inert) and a blend with 40% slag. For the first 28 h the curves for the blends with quartz and slag are essentially the same, confirming that the reaction of slag itself is not occurring in this period. Nevertheless, when the rate of heat evolution is normalized relative to the clinker component (as here) it is clear that the slag and quartz enhance the reaction of the clinker component due to their physical presence; this is the so called *filler effect* and can be attributed to two main factors. First, when SCM grains substitute clinker grains, as illustrated in Fig. 2, there is relatively more space available for the hydrates of the clinker phases to form in. Secondly, the surface of the SCM grains act as sites for the heterogeneous precipitation and growth of hydrates.

In the example of Fig. 1, after 28 h the heat evolution of the blend with slag is noticeably higher than that of the blend with quartz – this indicates the start of the reaction of the slag. Recent progress in understanding the impact of SCMs on hydration is discussed later, was reviewed in [5] and also forms part of another article in this volume [6].

2. Hydration of C_3S /alite

C_3S /alite constitutes between 50 and 80% of Portland cement and dominates the development of properties. So it is clear why many studies have concentrated on the reactions of this phase. It is also worth noting, that in a properly sulfated cement, in which the depletion of sulfate ions in solution occurs shortly after the maximum of the heat main heat evolution peak, the aluminate containing phases have little impact of the hydration of C_3S (see section 3). Furthermore the reaction of the C_2S /belite phase only really becomes significant after about 10 days or so [1].

Before discussing progress in understanding hydration during the different periods, it is worth noting a very important result published by Thomas [7]. Through the use of a novel method with a scanning calorimeter he could demonstrate that the activation energy of C_3S had an effectively constant value of 51 kJ/mol until well after the main heat evolution peak up to a degree of hydration of at least 65%. This suggests that the rate controlling step remains the same during this early hydration period, as different rate controlling mechanisms tend to have different activation energies. The activation energy for cements was slightly lower in the range 44–49 kJ/mol for the first 60 h or so of hydration. The difference between pure C_3S and cement can probably be attributed to the presence of minor elements in solid solution in the alite of cement and perhaps to the more rapid cooling conditions of industrial clinker.

2.1. Up to the end of the induction period

In the paper written for the Madrid conference [1], the mechanisms governing the induction period, in which the initially rapid rate of hydration decreases within the first few minutes to remain at a low rate,

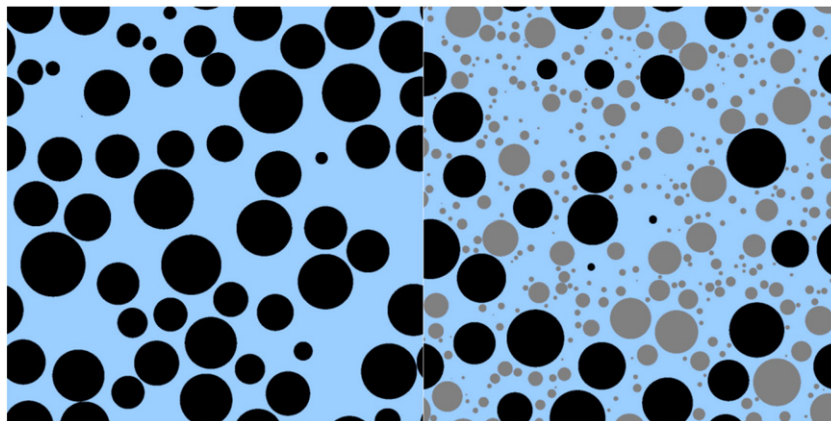


Fig. 2. Schematic of the particle distribution after mixing for a 100% Portland cement system at a water to cement ratio of 0.4 (left) and for the same system but with 40% of the clinker grains replaced by a filler (right). It is clear that replacement of cement grains by filler gives relatively more space for the hydrates of the clinker phases.

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