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In-situ X-ray tomographic monitoring of gypsum plaster setting

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ABSTRACT

The first *in-situ* monitoring of plaster hydration using X-ray tomography is reported in this paper. Dissolution of hemihydrate particles and formation of a network of gypsum needles can be observed in 3D. A 3D quantitative analysis based on the microstructure evolution allows the determination of the degree of reaction. In particular, the size of hemihydrate particles is shown to have an influence both on the hydration kinetics and on the final microstructure of the set plaster. This work paves the way to the understanding of the relationship between microstructure evolution, chemical degree of reaction and mechanical strength development for material processed through a setting reaction.

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1. Introduction

Gypsum plaster is one of the most ancient materials used for construction, with evidence of its use over several thousands of years and is still largely used for dry wall.

Its production starts from the calcination of gypsum, calcium sulfate dihydrate ($CaSO_4.2H_2O$), at around 120–130°C. During calcination calcium sulfate dihydrate loses ¾ of its crystallization water and turns into calcium sulfate hemihydrate ($CaSO_4.0.5H_2O$) also referred to as plaster powder. Hemihydrate is found in two forms, traditionally referred to as "α" or "β", depending on their mode of preparation; the α-form is prepared by wet methods (e.g. autoclaving) and the β-form by dry methods (e.g. calcining at around 120–130°C). The β-form is by far the most commonly encountered. Plaster is then prepared as a solid binder through a hydration reaction in water, as shown in Eq. 1.

$$CaSO_4.\frac{1}{2}H_2O+\frac{3}{2}H_2O\rightarrow CaSO_4.2H_2O \quad (1)$$

The driving force for this hydration reaction is the higher solubility of hemihydrate in water as compared to gypsum. This phenomenon has been described first by Lavoisier in 1765 and is largely reviewed in the literature. Authors often consider three steps in the hydration of plaster: dissolution of calcium sulfate hemihydrate leading to a supersaturated solution with respect to gypsum, nucleation and growth of gypsum needles and final formation of a solid material by entanglement of gypsum needles with complete depletion of hemihydrate.

The water to plaster (W/P) ratio, *i.e.* the amount of water used to hydrate the hemihydrate powder, is always well above the theoretical stoichiometric mass ratio of 0.186. This higher amount of water increases the fluidity of the paste, which becomes moldable. This also explains the high level of porosity in the final material obtained after complete drying of the excess water.

Monitoring and controlling the kinetics of the setting reaction are critical for industrial purposes. During the setting process, the plaster evolves both from a chemical point of view (monitored by the degree of reaction) and from a mechanical point of view (from a liquid paste to a solid material). Therefore, both aspects have been thoroughly investigated. For instance, the monitoring of the hydration reaction can be carried out using a large number of techniques [1–4], the most popular being calorimetry [1]. Other analytical techniques, such as differential thermal analysis or particle size analysis can only be implemented *ex-situ*; thus, they require stopping the reaction at different times, and demand one sample per studied time. Nevertheless, all these methods allow the determination of the degree of the hydration reaction.

On the other hand, the solidification of plaster can be monitored by the measurement of rheological and mechanical properties during setting. The evolution of the elastic modulus versus time can also be monitored by the measurement of ultrasound speed through the setting paste [5].

Some authors tried to link the degree of reaction and the development of strength, based on a description of the development of the microstructure at different times. Lewry and Willamson [1] suggested an evolution of the microstructure in three stages: firstly, development of a matrix of gypsum needles providing the initial strength, then relief of internal stresses caused by the built-up of pressure associated with needles growth and finally increase of strength due to water evaporation after complete hydration.

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The detailed description of the bonds formation between gypsum crystals is beyond the scope of our study, but it is admitted that the contact forces are highly dependent on the crystallographic orientation of the crystal faces in contact, as measured by atomic force microscopy [6,7].

The morphological description of the entanglement of gypsum crystals after hydration, as well as its structuring during the hydration phase, is critical as it directly controls the final strength of the material. The observation of the evolution of plaster microstructure with time is seldom described in the literature and mainly after having stopped the hydration at different times. The only study where the setting process has been monitored *in-situ* is, to the best of our knowledge, the work by Ridge [8] showing the growth of gypsum needles under an optical microscope. Lewry and Willamson carried out a *post-mortem* analysis based on scanning electron micrographs of fractured surfaces after having stopped the hydration at different stages. This allowed them to show the concomitant presence of partly solubilized hemihydrate particles and newly formed gypsum crystals [9]. This method is widely used to characterize the local organization of crystals due to its relative easiness and the high resolution one can reach. However, it is not possible to monitor *in-situ* the evolution of the microstructure during setting. Besides, the protocol used to stop the hydration might impact on the observed microstructure.

X-ray tomography has a strong potential for the study of plaster hydration, the presence of water and solid ensuring a good contrast between the phases. Historically, the first scans of gypsum were acquired at high resolution using synchrotron source on a hydrated plaster [10]. Recent work was done on the *in-situ* mechanical loading of fully hydrated plaster in a X-ray tomograph [11]. Only two studies deal with the use of X-ray tomography during hydration of plaster [4, 10]. In Bentz 2002 [10], 3D scans showed the concomitant existence of gypsum crystals and hemihydrate particles, but without any quantitative analysis. In Song 2009 [4], 3D scans with a voxel size of 3 μm of hydrating plaster were acquired after having stopped the hydration reaction after 60 min. Pore volume distribution was analyzed but no specific analysis of the pore evolution with hydration time was performed. One example of an *in-situ* monitoring of hydration was carried out on a cement paste [12]. The hydration was studied with X-ray synchrotron tomography from 1 to 60 days, as the hydration kinetics is much slower than for gypsum. The resolution of the X-ray device used in this study, even if as low as 0.7 μm, was a limitation to monitor the cement hydration as the calcium silicate hydrates controlling the strength are nanosized.

3D volumes with a voxel size of a few micrometers can be acquired and reconstructed in about 3 min on a lab scale apparatus thanks to recent development in microtomography. The present study focuses on the *in-situ* monitoring of plaster setting using X-ray tomography, for the first time to the best of our knowledge. The volume of material studied here is sufficiently large to give an average behavior of the setting of a plaster paste, with a statistical number of hemihydrate particles dissolving and with fluidity typical of an industrial process. 3D quantitative volume analysis is reported and the influence of hemihydrate particle size on the setting process and on the final microstructure of plaster is investigated; the evolution of hemihydrate particle size versus hydration time is also examined.

2. Materials and methods

2.1. Plaster preparation

A beta calcium sulfate hemihydrate was used for this study, supplied in store retail, with the following composition: hemihydrate 94.4%, gypsum 4.0% and impurities (mainly calcium carbonate) 1.6%. The hydration reaction was prepared using a W/P ratio by weight of 0.72. In general, the powder was used as received. In order to understand the influence of the granulometry of the raw powder on the microstructure of the set plaster, some plaster samples were prepared with hemihydrate sieved powder (either between 63 and 40 μm or below 40 μm).

The time when the hemihydrate powder was put in contact with water is referred to as the initial time in the paper. Hemihydrate and tap water, stored at 23 °C, were mixed by hand for 3 min. For X-Ray tomography observation, the paste was injected with a syringe into a drinking straw with a diameter of 3 mm, directly mounted on the X-ray tomograph. Knowing that surfaces of the setting paste were not observed, it was assumed that the bulk of the sample remained saturated with respect to water during the time period required for the complete examination of a sample (1 h 44 min). Setting time was separately characterized on the remaining paste, which was not injected into the straw, by the knife setting time method: setting time was then defined as the time after which a cut made in the paste with a blade remained opened.

2.2. X-ray tomography – in-situ setting of the plaster

The internal observation of the microstructure evolution during the plaster setting was carried out by means of X-ray tomography using a vtomex device (GE Phoenix|X-Ray GmbH) equipped with a 160 kV nano-focus tube, a tungsten transmitting target, and a 1920 × 1536 pixel Varian detector – see [13] for more details. The X-ray tube produces a polychromatic conical beam. The experiments were performed at a voltage of 80 kV and a current of 280 μA, with a voxel size of 2.5 μm^3. Based on Fourier Shell Correlation technique [14], the spatial resolution could be estimated to 5 μm. Due to the fast changes during hydration, the acquisition parameters were optimized in order to reduce the scan time. A continuous rotation was used and the integration time was 333 ms for each of the 600 projections acquired over 360°. These parameters resulted in measurement periods of 200 s for a complete scan. For technical reasons, a standard sequential tomographic acquisition has been utilized. However, new methods based on nonsequential projection angle sequences could also have been used [15] to monitor dynamic processes with a convenient balance of temporal and spatial resolutions. In the present study, for quantitative analysis purpose, the main motion artefacts were linked to the hemihydrate dissolution. The estimated dissolution rate (see section 3.2.2) was of the same order of magnitude as the resolution (few μm during a scan). Thus, it should not have given rise to significant errors. Beside, motion-related artefacts were not observed around hemihydrate particles.

The first scan was acquired 13 min (800 s) after the initial time; these 800 s corresponded to the time needed to prepare the sample (600 s) and to perform the first acquisition (200 s). In total, plaster setting was monitored during 6200 s after the initial time. After this time period, no more modification in the microstructure could be noted. The sample was then dried outside of the tomograph at 45 °C until constant weight was reached. It was re-scanned afterwards to observe the final set and dried microstructure. Because fast acquisition was no longer an issue, this last scan (denoted 24 h in the paper) was achieved with improved conditions *i.e.*, 900 projections and averaging 3 images at each step angle.

For coding the absolute value of the attenuation coefficient of each voxel, 32 bit volumes were reconstructed from the projections. Therefore, the voxel gray value corresponded to the measured attenuation coefficient (AC). The reconstructed volumes were first subjected to a median filtering. Then, the different scans were spatially registered: each scan was aligned with respect to the previous one to better follow-up the evolution of the microstructure versus time.

2.3. X-ray tomography – final microstructure

X-ray tomography was also used to perform 3D analysis at very high resolution (0.4 μm^3 per voxel, estimated resolution of 1.2 μm based on Fourier Shell Correlation technique) of the final microstructure of small pieces of plaster once set *ex-situ* (~ 0.5 × 0.5 × 5 mm). Note that such a very small voxel size requires very much care in doing the

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