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# Relation between ultrasound measurements and phase evolution in accelerated cementitious matrices

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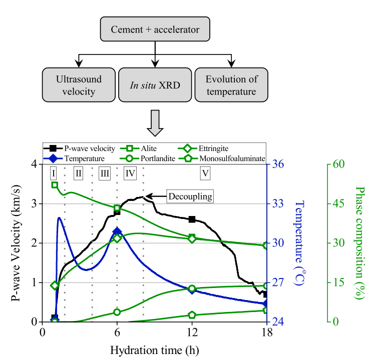
## HIGHLIGHTS

- Ultrasound velocity was sensitive to cement composition, accelerator type and dosage and phase composition of the matrix.
- Increase of P-wave velocity is directly proportional to hydration rates (mainly $C_3A$ hydration).
- Correlations between US velocity in mortars and their chemical properties and phase composition were established.

## GRAPHICAL ABSTRACT

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## ABSTRACT

This paper focuses on the characterization of setting and hardening of accelerated cementitious matrices by ultrasound propagation velocity, correlating these processes with chemical parameters and the phase evolution obtained by *in situ* XRD. Evolution of temperature and determination of setting times complemented this analysis. The technique employed provided a continuous monitoring of the setting and hardening of the hydrating matrix and was susceptible to changes in accelerator reactivity and phase composition. Results showed that ettringite formed by accelerator reaction improves the solid-phase interconnectivity and increases initial ultrasound velocity. P-wave propagation during the acceleration period is directly proportional to alite and $C_3A$ degrees of hydration. The influence of AFm phases to increase ultrasound velocity is stronger than ettringite and C-S-H. Based on an extensive statistical analysis, multivariate linear regressions were established between ultrasound velocity and the main chemical properties influencing its evolution, leading to a better comprehension of how these parameters are related.



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## 1. Introduction

Ultrasound measurements are a versatile tool to characterize cementitious matrices. Several ultrasound techniques have been used to evaluate the setting process [1–3], mechanical strength [4,5], porosity [6,9], permeability [6–9] and durability [10,11] of pastes, mortars and concrete during the past years. The influence of different superplasticizers and additions on early strength development has been analyzed by [12–15].

This technique may also be used to characterize the early age hydration reactions in cementitious matrices containing set accelerators [16, 17], in which the determination of initial stiffness and of early age strength are crucial for an adequate application. It may provide a continuous evaluation of the setting and hardening processes of these mixes, being more representative than Vicat needle and pin penetration tests, which are discontinuous and have a limited application range [18].

Table 1 summarizes the most recent work conducted in the field of ultrasound measurements applied to cementitious matrices and the main tests performed in each work. Note that the majority of the studies focus on the characterization of conventional concrete by ultrasound propagation velocity and only a few also assess the kinetics and mechanisms of hydration. None of them evaluates how accelerated chemical reactions and the resulting microstructure influence the propagation of ultrasound waves from a quantitative point of view.

The lack of such relations may compromise the characterization of accelerated cementitious matrices and the applicability of ultrasound measurements in practice. Therefore, it is fundamental to better comprehend how the chemical reactions occurring when accelerators are added and the resulting phase evolution influence the overall response to ultrasound propagation throughout the whole hydration process. By doing so, a more complete characterization of the evolution of mechanical properties of the matrix at early ages may be achieved, improving their design and quality control.

The main objective of this study is to characterize the hydration behavior of accelerated cement pastes and mortars by ultrasound propagation velocity, correlating these measurements with the chemical parameters and phase evolution of the matrix from a quantitative standpoint. For that, an experimental program was conducted with two types of cement and four types of accelerator, either alkali-free or alkaline. The setting and hardening processes as well as the development of microstructure were monitored by ultrasound propagation velocity. Setting times were determined by the Vicat needle test. Kinetics and mechanisms of hydration were characterized by *in situ* X-ray diffraction and evolution of temperature.

Multivariate regression analyses were conducted to better comprehend how ultrasound velocity and chemical properties of the matrix are related. For these analyses, the phase evolution during hydration and the chemical composition of cements and accelerators were considered as independent variables, whereas ultrasound velocity was the dependent variable.

Results indicate the main chemical parameters that influence microstructure development and ultrasound propagation velocity, explaining their origin. Correlations obtained are a simple useful tool to evaluate how the ultrasound velocity is affected by the chemical characteristics of the matrix. Therefore, this study promotes the rational use of this technique for characterization and control of matrices containing accelerators and for the evaluation of accelerator reactivity and compatibility with cement.

## 2. Experimental methodology

The experimental program was conducted at the Laboratory of Technology of Structures Luis Agulló at Universitat Politècnica de Catalunya (UPC) and at the Scientific and Technological Center from Universitat de Barcelona (CCIT-UB). Software development and data analysis for ultrasound wave propagation tests were carried out at the Institute of Physical and Information Technologies Leonardo Torres Quevedo from the Spanish National Research Council.

Ultrasound measurements were performed in cement pastes and mortars. In cement pastes, kinetics and mechanisms of hydration are not influenced by aggregates and may provide a clearer evaluation of the chemical processes occurring during hydration. However, accelerated pastes are very reactive and shrinkage may limit the progression of the test. Therefore, accelerated mortars were also evaluated because aggregates provide more space for the precipitation of hydrated phases and reduce reaction rates. As a result, the negative effects of chemical shrinkage are minimized and a continuous monitoring of setting and hardening may be obtained. In the following sections, materials, production processes and tests performed are described.

### *2.1. Materials*

Two types of Portland cement (CEM I 52.5R and CEM II/A-L 42.5R) were used in this study. These cements were selected among the commonly used in sprayed concrete applications. CEM II/A-L is widely used in countries from northern Europe, while CEM I is common in Spain and countries from Asia and America. Tables 2 and 3 show their mineralogical composition determined by XRD-Rietveld refinement and their chemical composition determined by XRF spectrometry, respectively. Besides, Table 4 summarizes their chemical and physical properties.

For the fabrication of the mortars, standard siliceous sand following the requirements from UNE EN 196-1:2005 [21] was used. Distilled water and a superplasticizer based on a polycarboxylate solution (34% of solid content) were used to prepare all pastes and mortars.

Three alkali-free accelerators composed by aluminum hydroxysulfate solutions were selected to cover the types commonly found in

**Table 1**
Summary of recent studies on ultrasound measurements applied to cementitious matrices.

| Reference | Samples tested | | | Characterization | | | Quantitative relation between ultrasound and phase evolution |
|---|---|---|---|---|---|---|---|
| | Conventional (fresh) | Conventional (hardened) | Accelerated (fresh) | Ultrasound | Kinetics | Phase evolution | |
| [1,12,14,15][a] | • | | | • | • | | |
| [2–5,13][b] | • | | | • | | | |
| [6–10][c] | | • | | • | | | |
| [11][d] | | • | | • | | • | |
| [16][a] | | • | | • | • | | |
| [17][b] | | • | | • | | | |
| This study | | | • | • | • | • | • |

[a] Characterization of setting, hardening and microstructure development of fresh cementitious matrices. Kinetics of hydration characterized by calorimetry.
[b] Characterization of setting, hardening and microstructure development of fresh cementitious matrices.
[c] Determination of porosity, permeability and mechanical strength of hardened concrete.
[d] Determination of durability in hardened concrete.