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The mechanical performance of polymer-modified cement pastes at early ages: Ultra-short non-aging compression tests and multiscale homogenization

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HIGHLIGHTS

- Three-minutes-long creep tests are performed on polymer-modified cement pastes.
- Their early-age elastic stiffness is considerably smaller than of neat cement pastes.
- A semi-analytical multiscale model for polymer-modified cement pastes is proposed.
- The model can be applied to cement pastes modified with different polymers.
- The consideration of entrapped air is prerequisite for the modeling success.

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ABSTRACT

Polymer-modified cementitious materials exhibit improved durability, adhesion strength, and impermeability in comparison to unmodified competitors. Because polymer-modified cementitious materials are nowadays increasingly used in construction, there is the need for a reliable mathematical description of their mechanical behavior, particularly so at early-ages. This study focuses on the evolution of the elastic and the non-aging creep properties of polymer-modified cement pastes during the first week after production, combining experimental and computational methods. As regards experimentation, three-minutes-long creep tests are performed once every hour, starting 21 h after production and continuing up to material ages amounting to eight days. This allows for the quasi-continuous quantification of the early-age evolutions of the elastic stiffness and of the non-aging creep properties. As regards computational modeling, an existing multiscale model for the homogenization of the elastic stiffness of cement pastes is extended towards consideration of polymers and entrapped air. The satisfactory agreement between computed and experimentally determined stiffness evolutions shows that the extended model can reliably describe the elastic stiffness of polymer-modified cement pastes, provided that entrapped air is appropriately considered.

1. Introduction

Latex and powder polymers are used for cement-based mortars and concretes in order to improve their workability, impermeability, adhesion to substrates, and ductility [1–3]. While polymer-modified mortars and concretes have been mainly used in repair and restoration since the early 1980s, these materials are nowadays increasingly applied also in construction [4–6]. This includes the use of sprayable polymer-modified concrete [7,8] and polymer-modified self-compacting concrete [9,10]. In this context, it is noteworthy that the advantages described above go along with a lower elastic stiffness and a more pronounced creep activity in comparison to conventional cement-based materials [9,11,12]. This raises the need for improved knowledge regarding their material behavior both at early ages and during long-term applications. Clearly, long-term durability can only be achieved, if cementitious materials are not damaged at early ages. Thus, research regarding the early-age behavior of cementitious materials is highly relevant for the durability design of concrete structures [13–15]. The

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required advances concerning polymer-modified cementitious materials call for combined experimental-computational research approaches, following routes that have proven to be successful in the context of classical cementitious materials.

As for innovative experimental characterization of the early-age evolutions of the elastic stiffness and the creep properties of conventional cementitious materials, regularly repeated ultra-short loading–unloading tests were developed quite recently [16–21]. In more detail, Irfan-ul-Hassan et al. [18,21] performed three-minutes-long creep tests once every hour, spanning material ages from 21 h up to eight days. This provides access to mechanical properties of specific early-age microstructures, because three minutes are so short that the chemical reaction between cement and water cannot make a significant progress, i.e. the microstructure remains essentially the same during a three-minutes-long test. On the contrary, two subsequent tests already refer to different microstructures, because the ongoing hydration reaction does make a significant progress within one hour, at least at early ages.

The evaluation strategy for ultra-short creep tests performed on cementitious materials at *early ages* deserves special attention. Standard test protocols consider deformations developing during the loading processes to be of quasi-elastic nature, and they consider creep deformation to start once the load plateau is reached, see, e.g. [22,23]. However, it was shown in [18] that *significant* creep deformation develops already *during* the short loading phase of an early-age creep test, and that consideration of this effect provides access to mechanically rigorous elastic and creep properties. The latter are an important prerequisite for the success of follow-up activities regarding multiscale modeling of cementitious materials, see [24,21].

Continuum micromechanics [25] provides a powerful framework for homogenization of microheterogeneous materials, provided that their hierarchical organization is adequately represented. The quest for the most realistic representation of the hierarchical organization of cement pastes dates back to the early 2000s. The first three types of models were developed for predicting the hydration-induced *stiffness* evolution of cement pastes.

- Bernard et al. [26] considered at the scale of cement paste that the material is a highly disordered arrangement of unhydrated cement particles, capillary pores, C-S-H gel, Portlandite, and aluminates. All five constituents were considered to exhibit spherical phase shapes. At the next smaller scale of the C-S-H gel, Bernard et al. envisaged a matrix-inclusion morphology with a matrix made of outer "low-density" C-S-H gel and spherical inclusions made of inner "high-density" C-S-H gel.
- Constantinides and Ulm [27] improved the microstructural representation at the scale of cement paste by considering the material to be a matrix-inclusion composite with a C-S-H gel matrix and spherical inclusions representing unhydrated cement particles, capillary pores, Portlandite, and aluminates. At the next smaller scale of the C-S-H gel, they took over the developments of Bernard et al. described above.
- Sanahuja et al. [28] considered cement paste to be a matrix-inclusion composite with an outer "low-density" C-S-H gel matrix and spherical inclusions representing unhydrated cement particles and inner "high-density" C-S-H gel, respectively. Their improvement refers to the next smaller scale of the C-S-H gel: they introduced *oblate* solid C-S-H particles in direct mutual interaction with different amounts of gel and capillary pores. The *non-spherical* phase shapes allowed for controlling percolation thresholds, i.e. hydration degrees at which homogenization starts to deliver nonzero elastic stiffness.

In the context of modeling the hydration-induced evolutions of both the elastic stiffness *and* the uniaxial compressive strength, the following two types of models were developed:

- Pichler et al. [29] considered at the scale of cement paste that the material is a highly disordered arrangement of spherical unhydrated cement particles, spherical capillary pores, and hydrate gel *needles* which are isotropically oriented in all space directions. The needle shape was essential for satisfactory strength predictions, because the stress concentration into a hydrate gel needle is significantly more realistic for quantifying strength-related stress peaks, compared to the stress concentration into a hydrate gel sphere.
- Pichler and Hellmich [30] finally considered explicitly that capillary pores and hydrate gel needles are significantly smaller than unhydrated clinker grains. Therefore, they modeled cement paste as a matrix-inclusion composite with unhydrated cements particles embedded in a matrix called "hydrate foam". At the next smaller scale of this hydrate foam, they considered a highly disordered arrangement of capillary pores and hydrate gel needles which are isotropically oriented in all space directions.

In addition to elasticity and strength, the model by Pichler and Hellmich [30] is also capable of predicting the hydration-induced evolution of non-aging *creep* properties. In this context, two creep constants of the hydrate gel needles were identified in the context of a top-down analysis involving some 1000 non-aging creep functions of cement pastes, measured during the first week after production in hourly-repeated 3-min-lasting creep tests [18,24].

Recently, the state-of-the-art representation of conventional cement pastes, see [30], was extended towards consideration of polymers [31]. The extended model was applied to the analysis of dynamic moduli of polymer-modified cement pastes, measured 2, 7, and 28 days after production [31].

The present study refers to combined experimental-computational research concerning the early-age evolution of the mechanical properties of one plain cement paste (=reference mix) and of two different polymer-modified cement pastes. As for experimentation, the creep testing protocol of Irfan-ul-Hassan et al. [18] is followed, i.e. the cement pastes are subjected to hourly repeated three-minutes-long uniaxial compressive creep tests. In total, 168 creep tests are performed on each specimen, spanning material ages from 21 h up to eight days. The tests are evaluated following the strategy of Irfan-ul-Hassan et al. [18], i.e. it is considered that significant creep strains are already developing during the loading phase of each creep test. In addition, the ultrasonics pulse velocity test method is used to characterize the stiffness properties of the used polymers, and quasi-isothermal differential calorimetry is used to get access to the reaction kinetics of polymer-modified cement pastes. Modeling focuses on the early-age stiffness evolutions of polymer-modified cement pastes. The micromechanical representation of polymer-modified cement pastes according to [31] is further enriched by consideration of entrapped air. The further extended model is used to explain the stiffness evolutions from 24 h after production to material ages of 8 days. Predictive capabilities of the developed model are assessed quantitatively, by comparing model predicted stiffness values with experimentally determined counterparts.

The paper is organized as follows: Section 2 refers to the tested materials, to the employed experimental methods, and to corresponding results. Section 3 presents the extended multiscale homogenization method for polymer-modified cement pastes and the comparison of model predictions with the experimental