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Influence of purified attapulgite clays on the adhesive properties of cement pastes as measured by the tack test

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ABSTRACT

This study evaluates the influence of small additions of highly-purified attapulgite clays (0.2% and 0.5% addition by mass of cement) on the adhesive properties of cement pastes. Adhesive properties are measured by the tack test, a novel method of evaluating the rheological properties of granular materials. To better understand the results of the tack test as they pertain to cementitious materials, a highly concentrated material that is evolving due to thixotropic rebuilding and hydration, they are supplemented with a measure of the viscoelastic properties over time obtained through low-amplitude oscillatory shear rheometry. The influence of different preshear conditions and resting times (age of paste) on the adhesive properties are determined. Results show the tack test to be a suitable method for obtaining useful information about the adhesive properties and structural evolution of the material in the fresh state.

1. Introduction

The fresh-state properties of cementitious materials impact construction productivity and the eventual properties of the material in service. With the addition of chemical and mineral admixtures, it is possible to manipulate the fresh state to improve concrete processing. This has led to such innovations as self-consolidating concrete (SCC) – a highly flowable concrete with superior segregation resistance. These properties effectively eliminate the need to apply external vibration during casting, which increases construction efficiency. However, the high flowability and faster casting rates achieved with SCC compared to normal vibrated concrete results in potentially higher lateral pressure exerted on formwork. Due to this effect, and the lack of a full understanding of the pressure transmitted to formwork from SCC, formwork is typically designed to withstand the full hydrostatic pressure that the SCC could exert. As formwork is one of the largest components of the cost of concrete construction, it is desirable to safely reduce the formwork design criteria through suitable materials selection and proper construction techniques.

The pressure exerted by SCC on formwork is closely tied to its fresh-state rheological properties. Recent work has shown the potential of further manipulating the rheological properties of SCC with clays. Small additions of clays (<1% by mass of binder) have been found to significantly increase the green strength (fresh-state stiffness) and shape stability of SCC with little compromise to the initial flowability [1]. The most pronounced effect was achieved with nanoclays (clays dispersed into their individual constituents). These properties were demonstrated to be effective in reducing SCC formwork pressure. In a study that evaluated the influence of different mineral admixtures on the formwork pressure response of SCC, an SCC mix with 0.33% nanoclay was found to experience reduced lateral pressure while still exhibiting comparable initial slump flow as the control [2].

The physical origin of this effect of clays is not fully understood. Flocculation studies have shown that clays increase floc size and flocculation strength [3,4], which can increase the viscosity and level of thixotropy of concrete. Reduction in SCC formwork pressure has been tied to increases in thixotropy [5–7]. In a series of studies determining the influence of various admixtures on SCC formwork pressure, thixotropy-enhancing admixtures were found to decrease initial pressure and increase rate of pressure drop over time [8–10]. Although thixotropy is undoubtedly tied to the clay effect, the cohesiveness within the material is expected to have a significant effect on stress transfer and subsequently on the formwork pressure. This issue can be investigated by considering the adhesive properties of the material. A standard method of evaluating the adhesive properties of a material is the *probe* tack test, which is widely used to

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complement rheological characterization of soft polymer adhesives [11–13]. The test can provide a measure of cohesion (the internal strength of the material at rest) and adherence (the tendency of the material to stick to a surface). High cohesion has been found to increase the rate of lateral pressure drop of SCC over time [14]. And high adherence can help to transfer lateral stress to vertical stress, making it possible for SCC formwork pressure to be less than hydrostatic [15]. Therefore, the influence of clays on the adhesive properties of cementitious materials is of interest.

Thus far, only a few select studies have implemented the tack test on granular-type materials [16,17]. Due to the limited number of studies that have been done to evaluate the adhesive properties of cementitious materials, the information that can be obtained from the results of the tack test is not fully resolved. In addition, due to the nature of the material: a highly concentrated suspension that is evolving due to thixotropic rebuilding and hydration, there is a need to interpret the results while taking the aging of the material into careful consideration. This current study aims to evaluate the influence of nanoclays on the adhesive properties of cement pastes through the tack test. In addition, to better understand the results as they pertain to cementitious materials in general, they are supplemented with a measure of the viscoelastic properties over time through low-amplitude oscillatory shear rheometry. It is found that the tack test is a suitable method for obtaining useful information about the adhesive properties and structural evolution of the material in the fresh state.

2. Materials and experimental procedures

2.1. Materials

Tap water and type I Portland cement with a Blaine fineness of 385 m^2/kg are used in all mixes. A highly-purified attapulgite clay, refined from bulk attapulgite, is the clay selected for this study [18]. They are 1.75 μm in average length and 3 nm in average diameter, thereby considered to be nano-sized. They have been chemically exfoliated to preserve their uniform shape and size while removing all impurities (such as quartz and swelling clays). The aspect ratio (average length divided by average diameter = 583) of the clay particles is very high. Therefore, they may form a highly entangled gel even at a small volume concentration, provided they are properly dispersed into individual particles.

All paste mixes have a water-to-cement (w/c) ratio of 0.43 by mass and are prepared by hand-stirring for 60 s. For mixes with a nanoclay addition, they are introduced as an aqueous suspension, where the nanoclay is blended with the mixing water in a household blender for 5 min to facilitate dispersion. All tests start immediately after mixing.

2.2. Experimental methods

All rheological tests are performed on a Paar Physica MCR rheometer with a parallel-plate geometry. The top plate has a diameter of 50 mm and the bottom plate is temperature-controlled with a circulating water bath set to 20 °C. To prevent slip, the surfaces of the plates are covered with 150-grit adhesive sandpaper. The rate of data acquisition is 10 Hz for higher plate velocities and 5 Hz for lower plate velocities. Further details of the experimental method can be found elsewhere [19].

2.2.1. Tack test

The tack test is performed on cement pastes with 0%, 0.2% and 0.5% nanoclay addition by mass of cement (0 NC, 0.2 NC and 0.5 NC). The fresh cement paste sample is placed on the bottom plate, then the top plate is lowered to an initial measuring gap of 1 mm. Once the top plate is in position, the sample is trimmed to match the diameter of the plate. During the test, the top plate moves up vertically at a constant velocity, subjecting the sample to stretching, and the normal force experienced by the top plate is recorded over time. The normal force evolution is recorded for the following plate velocities: 10, 50, 200, 500, and 1000 μm/s. The test is run until the sample reaches complete separation between the two plates. A shear rate of 50 s^{-1} (measured at the circumference of the sample) is applied for 60 s before the start of the tack test to ensure all samples start at the same state after mixing.

Cementitious materials are both time and shear history dependent. Therefore, the influence of a higher preshear condition and various resting times on the adhesive response of pastes is evaluated. The tack test is performed at 10 μm/s for cement paste mixes with a 0% and 0.5% nanoclay addition subjected 300 s^{-1} for 60 s. Following this preshear, the tack test is performed after the following resting periods: 0, 150, and 300 s.

A new sample is prepared for each test. Three measurements are taken for each mix for each testing protocol and the average is taken to be the representative measurement.

2.2.2. Low-amplitude oscillatory shear rheometry

To better interpret the results of the tack test, they are supplemented with a measure of the evolution of the paste's structure. This is done through low-amplitude oscillatory shear rheometry. This method provides information about the viscoelastic properties of suspensions and it has been demonstrated to be applicable to fresh cement paste [20,21]. The theory is briefly explained here.

An oscillatory strain is applied as a sine function:

$$\gamma = \gamma_0 \sin \omega t \tag{1}$$

where γ_0 is maximum strain amplitude, t is time, and ω is frequency. If the strain is sufficiently low so that the particles in the suspension remain close to each other, the microstructure is not disturbed and the material can recover elastically. In this case (linear regime), the measured response in terms of stress is as follows:

$$\tau = \gamma_0 (G' \sin \omega t + G'' \cos \omega t) \tag{2}$$

where τ is shear stress, G' is storage modulus, and G'' is loss modulus. G' is the elastic (or in-phase) component while G'' is the viscous (or out-of-phase) component. By monitoring the time evolution of G', it is possible to measure the structural building of cement pastes over time. The method has been applied to determine the effect of admixtures, such as nanosilica and cellulose ethers, and w/c ratio on the storage modulus of cement pastes and has been tied to flocculation and hydration mechanisms [22–24].

The first step in performing oscillatory shear rheometry is to find the linear viscoelastic region (LVR), where G' (and also G'') is independent of applied frequency and strain. To find the LVR, a strain sweep is performed on cement pastes with 0% and 0.5% nanoclay addition. Strain amplitudes ranging from 10^{-4} to 1 are applied at a fixed frequency of 1 Hz. The amplitude sweep results are reported in Fig. 1a. The strain sweep curves exhibit two main branches – a plateau region corresponding to the LVR regime and a decreasing branch indicating that the sample is experiencing a shear-induced breakage. The LVR regime of the plain cement paste ends at a critical strain of about 6×10^{-4}, which corresponds to the same order of magnitude as that reported by previous authors [21]. The critical strain for the clay-modified cement paste is significantly higher than that of a plain cement paste. This is in agreement with previously reported results indicating that the addition of clays increases flocculation within the