



## A test method for determining adhesion forces and Hamaker constants of cementitious materials using atomic force microscopy

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### ABSTRACT

A method for determining Hamaker constant of cementitious materials is presented. The method involved sample preparation, measurement of adhesion force between the tested material and a silicon nitride probe using atomic force microscopy in dry air and in water, and calculating the Hamaker constant using appropriate contact mechanics models. The work of adhesion and Hamaker constant were computed from the pull-off forces using the Johnson–Kendall–Roberts and Derjagin–Muller–Toropov models. Reference materials with known Hamaker constants (mica, silica, calcite) and commercially available cementitious materials (Portland cement (PC), ground granulated blast furnace slag (GGBFS)) were studied. The Hamaker constants of the reference materials obtained are consistent with those published by previous researchers. The results indicate that PC has a higher Hamaker constant than GGBFS. The Hamaker constant of PC in water is close to the previously predicted value  $C_3S$ , which is attributed to short hydration time ( $\leq 45$  min) used in this study.

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### 1. Introduction

Design of a concrete mixture with desirable workability, especially proper flow ability, is essential in every step of concrete construction, from the fresh concrete manufacturing process and quality control to the subsequent hardened concrete performance [1]. Recent advances in rheological characterization of cement-based materials have permitted engineers to formulate optimal concrete mix design and control mixture homogeneity during the concrete manufacturing and construction processes [2].

The rheological behavior of a cement-based material is primarily controlled by interparticle forces and spatial particle distribution. The interparticle forces in a flowing cement paste system consist of lubrication, adhesion, and collision forces between cement particles and/or between a cement particle and a boundary [3]. All these forces are influenced by the hydration process of cementitious materials, which depends not only upon the material characteristics (such as particle size distribution, chemical composition, water-to-cementitious material ratio (w/cm), and admixtures) but also upon the hydration time, construction process (such as mixing and placement procedures), and environmental conditions (such as time, temperature and relative humidity) [4–7]. Although a great deal of work has been done on the interparticle forces of granular and/or suspension materials, limited research is conducted to study the interparticle

forces in a cement system, which is partially due to the complexity of cement hydration [8]. Roussel et al. [9] had provided general guidelines that identify the physical microstructure parameters that govern the macroscopic rheological behavior in the steady state flow of cement suspensions. The parameters covered were interaction forces (surface, Brownian, hydrodynamic and contact forces), yield stress (particle interactions, packing and yield stress model [10,11]) and flow (shear thinning and thickening). Upon discussion of the different parameters as related to cement paste flow, a classification of different flow behaviors based on predominant interactions under simple shear with varying volume fractions and shear rates was presented.

One important parameter that depicts particle interactions is the Hamaker constant—a force constant used for describing the van der Waals force between two particles or between a particle and a substrate. Using this force constant, the particle interactions in a granular or suspension system can be simulated and predicted [9,12]. A few researchers have attempted to measure adhesion forces and Hamaker constant of cement-based materials. Uchikawa et al. determined the steric repulsive force between polished clinker and silicon in solutions with different admixtures [13]. They found that the fluidity of fresh cement pastes was correlated to the repulsive forces of their particles. Kauppi et al. measured the interaction forces between spherical and flat MgO particles using an Atomic Force Microscope (AFM) in a solution containing superplasticizer [14]. They discovered that superplasticizers contributed to both electrostatic and steric repulsion. Lesniewska et al. [15,16] evaluated the forces between calcium silicate hydrate (C-S-H) layers in different solutions. They reported that in the solution similar to

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