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Le Châtelier's Conjecture: Measurement of Colloidal Eigenstresses in Chemically Reactive Materials

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Abstract

Volume changes in chemically reactive materials, such as hydrating cement, play a critical role in many engineering applications that require precise estimates of stress and pressure developments. But a means to determine bulk volume changes in the absence of other deformation mechanisms related to thermal, pressure and load variations, is still missing. Herein, we present such a measuring devise, and a hybrid experimental–theoretical technique that permits the determination of colloidal eigenstresses. Applied to cementitious materials, it is found that bulk volume changes in saturated cement pastes at constant pressure and temperature conditions result from a competition of repulsive and attractive phenomena that originate from the relative distance of the solid particles – much as Henry Louis Le Châtelier, the father of modern cement science, had conjectured in the late 19th century. Precipitation of hydration products in confined spaces entails a repulsion, whereas the concurrent reduction in interparticle distance entails activation of attractive forces in charged colloidal particles. This cross-over from repulsion to attraction can be viewed as a phase transition between a liquid state (below the solid percolation) and the limit packing of hard spheres, separated by an energy barrier that defines the temperature-dependent eigenstress magnitude.

Keywords: Poromechanics, Colloidal Stresses, Cement-Based Materials, Expansion, Shrinkage.

1 Introduction

In summary of his seminal works on cement hydration in 1898, Henry Louis Le Châtelier (1850–1936) concluded that "il résulte donc de ces recherches que l'hydratation de tous les liants hydrauliques est accompagnée à la fois d une augmentation du volume apparent et d'une diminution du volume absolu"; and that "l'augmentation du volume apparent ne peut dans l'état actuel recevoir aucune explication c'est à dire être rattachée à un phénomène plus simple et d'ordre plus general."[1][1] The absolute volume reduction, also coined chemical shrinkage

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[1]It follows from these researches that the hydration of all the hydraulic binders is accompanied both by an increase in the apparent volume and a decrease in the absolute volume (...) Our current state-of knowledge falls short, to explain the increase in the apparent volume, that is to say to be attached to a simpler and more general phenomenon.