diffusion-fundamentals

The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Understanding Water Diffusion in Concrete and Clays

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1. Introduction

Well-made concrete has a long service life, providing almost maintenance free structures, but many structures deteriorate prematurely. The key for making durable concrete is to limit its ability to transport fluids (like water). Devising realistic testing methods that determine the ability of concrete to withstand water penetration requires an understanding of water mobility. Concrete consists of a well-packed mixture of gravel and sand bound together with cement paste. This paste is made up of a number of components, including a surprisingly large amount of un-hydrated cement, and has a well developed pore structure containing water. The complex pore structure dominates many of concrete's properties; including the size, shape and connectivity of pathways for the ingress of ions and the exit of the corroded products that lead to a loss of durability. The complex nature of the pore structure is shown by the range in diameter (d), which can vary from mm's to Å's. Both ends of this range control the properties of the concrete. Capillary pores, $d > 100$ Å, typical of poor quality concrete, give easy access to aggressive fluids and dissolved ions. Gel pores, $d < 100$ Å, are thought to contain water in a similar fashion to the interlayers of layered silicate clay minerals, and will limit water diffusion in good concrete. Because of their size capillary pores control the transport of water in poor concrete, while in good quality concrete these pores become discontinuous and the gel pores will control the water transport.

The properties of water in gel pores are thought to be very similar to the interlayer water in the clay mineral smectite, which reversibly adsorb water because certain cations are present in the interlayer space. In this family of clays it is supposed that the cations act as gatekeepers as water is readmitted into the interlayer only when they are present. However, the clay halloysite has surface-bound interlayer water even though no cations are present. When halloysite is dried water cannot be readmitted to the interlayer space. Therefore it is of interest to compare water dynamics in these clays with cement paste.

Our study on the water mobility in concrete [1] and clays [2] aims to describe the reasons why water transport is hindered in durable concrete. In this work we show quasi-elastic neutron (QENS) results, measured using the spectrometers NEAT at HMI and OSIRIS at ISIS, contrasting the water diffusion in halloysite, smectite and cement pastes.

2. How can we describe water diffusion and rotation?

Protons are involved in both rotation and long-range translational motions of water molecules, occurring in different time and length scales [3]. Analysing the quasi-elastic (QE) region of the neutron scattering spectrum by using different energy resolutions allow these differences to be observed.

The simplest analytical model describing the diffusion of water assumes decoupled translational ($T(\omega,Q)$) and rotational ($R(\omega,Q)$) motions [4]. For our experimental Q-range, a random rotational diffusion is characterized by a relaxation time τ_R given by the first terms of the Sears expansion [5]. Translational motion, commonly modelled by a Lorentzian with half-width at half maximum $\Gamma_T(Q)$ [6], can be approximated by the random diffusion model [7]. From such analysis one obtains a self-diffusion coefficient (D_t) and an average residence time between jumps (τ_0), which are related to the mean jump distance (L).

Considering the "relaxing cage model" [8] some correction can be made for the correlation between translational and rotational motions of water molecules. This approximates to the cessation of both the isotropic nature of the diffusion and the localization of the water molecule by interaction with neighbours. Here the water dynamics is described by a structural relaxation time, τ, and an exponential parameter β, whose deviation from unity indicates that the "decoupling approximation" is no longer valid.

3. Conclusion

From the QENS data on the cement paste we could differentiate: (1) chemically bound water, (2) "glassy water" - that interacts with the surface of the gel pores in the paste, and (3) unbound water molecules confined within the larger capillary pores of the paste. The "glassy water" shows several anomalies, which required further studies that are now underway. The role of cations in water dynamics is shown by the lower temperature activation of the motions in the smectite compared to the halloysite interlayer water.

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