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Diffusive behavior of tritiated water and ^{36}Cl through partially-saturated cement-based materials

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Diffusion into cement-based materials has been extensively investigated since this transport phenomenon is a key parameter for modelling processes related to contaminant transport in cement-based barriers, assessment of rebar corrosion in civil concrete structures and long-term behavior of nuclear waste disposal systems based on cement-based engineered barriers. Moreover, there are many situations where cement-based materials can be partially water-saturated, like in deep geological facility for the disposal of long-lived intermediate and high level nuclear waste. In this case, the saturation state of cement-based materials will be controlled during the exploitation period by the surrounding relative humidity imposed by ventilation of the underground drifts and shafts and during the post-closure period by gas generation (radiolysis, anoxic corrosion...). However, few data are available in literature on radionuclide diffusion behavior in cement-based materials according to saturation. Since determining the solute diffusion coefficients through cement-based material remains a challenging work for fully-saturated conditions, acquiring them for controlled partial saturation conditions constitutes thus a knotty task.

In the current study, we present three innovative and complementary techniques allowing an accurate determination of diffusive parameters for tritiated water (HTO) and ^{36}Cl through hardened cement pastes (HCP) undergone water-saturation ranging from 20% to 100%. The first technique is based on an adapted through-diffusion experiment with a control of the suction (up to 9 MPa) using osmosis process. The two other techniques perform diffusion experiments in humidity chambers controlled by salt solutions. One allows ^{36}Cl to diffuse from a spiked fresh cement paste into a cementitious material having a given saturation. After a controlled diffusion time, the ^{36}Cl activity profiles within the samples are acquired by means of the abrasive peeling method. The last technique is based upon a vapor exchange method where HTO is added in the salt solution and penetrates the studied material under its gas form. Using this principle, steady-state through-diffusion experiments can be performed for HTO allowing the determination of its effective diffusion coefficient and retention properties for several saturation states.

The comparison of the data acquired by means of the three techniques shows a good consistency (Figure 1). The evolution of the effective diffusion coefficients with saturation are discussed as a function of species type using empirical models such as the Archie's second law.

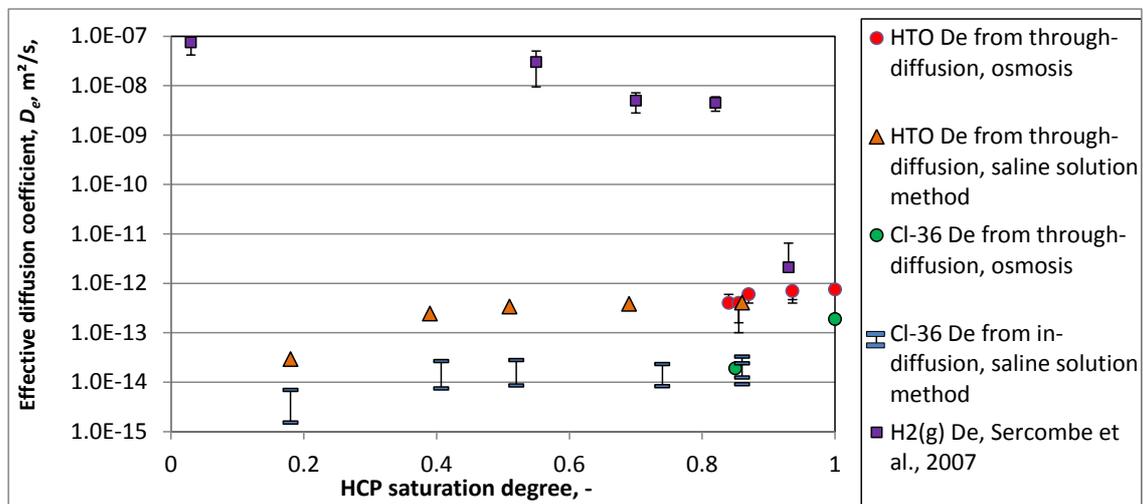


Figure 1: Effective diffusion coefficient (D_e) values normalized by the values obtained at full-saturation for HTO and ^{36}Cl as a function of the water saturation of the cement pastes.