

A METHOD FOR THE DETERMINATION OF GAS DIFFUSION COEFFICIENTS IN UNDISTURBED BOOM CLAY

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The main mechanisms by which gas will be generated in deep geological repositories are: anaerobic corrosion of metals in wastes and packaging; radiolysis of water and organic materials in the packages, and microbial degradation of various organic wastes. Corrosion and radiolysis yeald mainly hydrogen while microbial degradation leads to methane and carbon dioxide.

The gas generated in the near field of a geological repository in clay will dissolve in the ground water and be transported away from the repository by diffusion as dissolved species. However if the gas generation rate is larger than the diffusive flux, the porewater will get oversaturated and a free gas phase will be formed. This will lead to a gas pressure build-up and finally to an advective gas flux. The latter might influence the performance of the repository.

Therefore it is important to assess whether or not gas production rates can exceed the capacity of the near field to store and dissipate these gases by dissolution and diffusion only. The current available gas diffusion parameters for hydrogen in Boom Clay, obtained from the MEGAS project (Volckaert *et al.*, 1994), suffer from an uncertainty of 1 to 2 orders of magnitude. Sensitivity calculations performed by Weetjens *et al.* (2006) for the disposal of vitrified high-level waste showed that with this uncertainty on the diffusion coefficient, the formation of a free gas phase cannot be excluded. Furthermore, recent re-evaluations of the MEGAS experiments by Krooss (2008) and Aertsens (2008) showed that the applied technique does not allow precise determination of the diffusion coefficient.

Therefore a new method was developed to determine more precisely the gas diffusion coefficient for dissolved gases (especially dissolved hydrogen) in Boom Clay. This should allow for a more realistic assessment of the gas flux evolution of a repository as function of the estimated gas generation rates.

The basic principle of the set-up is shown in figure 1:

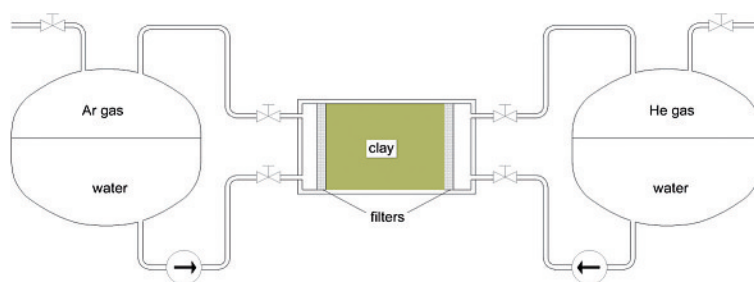


Figure 1: Basic principle for a set-up to determine of diffusion coefficients of dissolved gasses in a low permeability clay system.

The basic idea is to perform a kind of a through diffusion test with dissolved gasses.

A clay core is sealed in a stainless steel cell and connected at both sides with water vessels that are pressurized with 2 different gasses at the same pressure. In this way no advective gas flux can occur and the clay sample remains fully water saturated. The water at both sides is pumped around. Gas dissolves into the water and because of the gas concentration gradient, these dissolved gasses diffuse through the

clay. The dissolved gas that diffuses into the other water vessel will equilibrate with the gas atmosphere and the changes in the gas composition can be determined through e.g. gas chromatography.

As such, the diffusion parameters of two gasses can be determined in a single experiment.

Tests will be conducted with the following gas combinations: He and Ar, He and CH₄, Ar and Hytec (5% H₂ in Ar).

These tests are interpreted with a simple diffusive transport model. The model solves the diffusive transport equation in a 1D geometry. The porosity of the clay core is set to 30%. At one end, a fixed gas concentration was imposed as boundary condition, corresponding to the dissolved fraction at the prevalent pressure in the vessel following Henry's law. At the other end, the concentration of the considered gas is assumed to be 0. As model output, fluxes at both faces, as well as concentration profiles at regular time intervals are obtained.

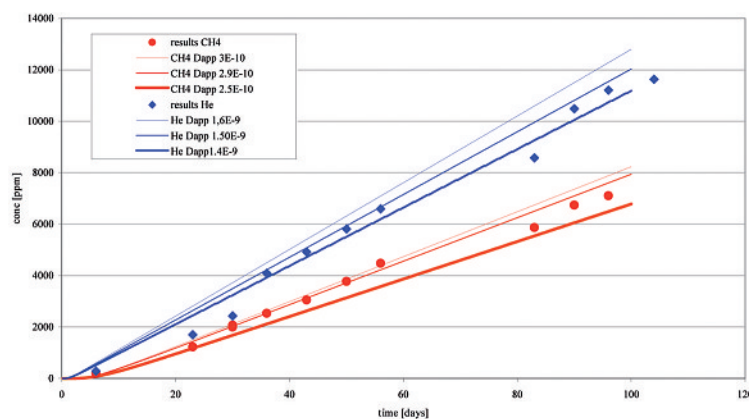


Figure 2: Measured gas concentrations of the gasdiffusion test with He and CH₄ and corresponding simulations with a narrow range of diffusion coefficients.

Figure 2 shows that for both He and CH₄ the evolution of the measured gas concentrations in the downstream vessel for each gas fall within simulation curves using rather narrow ranges of the D_{app} : $1.4 - 1.6 \cdot 10^{-10} \text{ m}^2/\text{s}$ for He and $2.5 - 3 \cdot 10^{-10} \text{ m}^2/\text{s}$ for CH₄. It is clear that the method is very sensitive and that quite accurate measurements of the diffusion coefficients can be obtained, far better than the previous results obtained in the MEGAS project (Volckaert *et al.*, 1994). Currently an experiment to determine the diffusion coefficient for hydrogen is on-going.

This method allows for an accurate determination of diffusion parameters of dissolved gasses in the Boom Clay and can be applied to other low permeability porous medium.

References:

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