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# An experimental study on the measurement of gas diffusivities in unsaturated clay based materials

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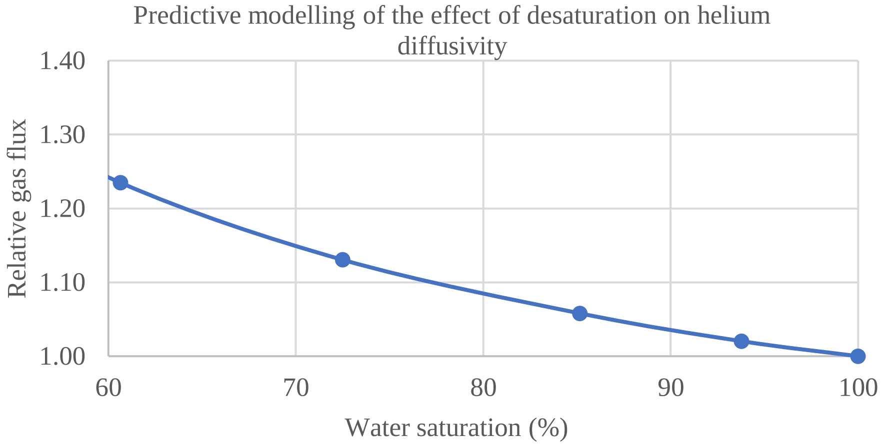
Gas migration in porous media in the context of nuclear waste disposal is dominated by diffusion. The availability of abundant data on diffusion coefficients of different gases in many clay based materials such as Boom Clay, Opalinus Clay and Callovo-Oxfordian Clays provides a good overview of the rates of gas transport through these different clay rocks [1]. Such formations are being considered as potential host rocks for the disposal of high and intermediate level radioactive wastes across Europe. Diffusion coefficients have been studied with particular interest due to the unavoidable gas generation phase in the nuclear waste repository [2]. However, all the diffusion measurements performed so far have been under fully saturated conditions, with little experimental data on gas diffusivities in partially saturated conditions, which have been hypothesized to exist during the early part of the life cycle of the repository [3]. The objective of the current study is to provide an overview of the effect of desaturation on the rate of diffusive transport of gases in partially saturated clay based materials.

The experimental methodology is based on the double through diffusion setup used in previous studies to measure gas diffusivities in saturated clays [1]. However, the setup is modified so as to keep a fixed level of unsaturation in the clay sample. This is done by employing the vapour equilibrium technique, using oversaturated salt solutions in both upstream and downstream reservoirs to maintain a constant relative humidity in the entire setup [4].

Preliminary pore network modelling has already shown that the gas diffusivity increases with a decrease in water saturation of the clay material. However, the predicted trend also suggests that this increase is only marginal and should not normally exceed an order of magnitude. This is shown in the figure below.





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All the diffusion measurements are carried out on synthetic clay samples of dimensions 3.8 cm (diameter) and 2 cm (height). In Table 1, the Sw stands for water saturation. The compositions of the synthetic clay samples (60% clay, 20% sand, 20% silt) have been adjusted so as to most closely mimic the mineralogical compostion of Boom Clay. Synthetic samples are used instead of natural samples like Boom Clay because of their better physical response to drying than Boom Clay.

The selected control variable to lower the saturation of the material is suction, which is imposed by changing the relative humidity of the environment using oversaturated salt solutions [4]. Suction can be kept constant given constant ambient temperature and pressure. Thus, the clay sample is conditioned at a certain suction, which in turn leads to desaturation in the range of 70-100% of total saturation depending on the level of suction. For this study, the saturation range has been fixed in between 70-100% of total saturation. This range has been chosen because of what is expected in a real life nuclear waste repository. The first measured diffusion coefficients in two partially saturated samples are shown in Table 1. The initial measurements of gas diffusion coefficients in unsaturated synthetic clays show that the increase in diffusivity of helium is higher than that of argon. Helium is the second lightest known element and has an extremely small molecular size and hence the impact of desaturation on helium diffusivity could be more pronounced than the same for heavier gases.

**Table 1**: Diffusion coefficients overview.

| Sample | Suction (MPa) | $D_{eff}$ Helium ($x10^{-10}$ $m^2/s$) | $D_{eff}$ Argon ($x10^{-10}$ $m^2/s$) |
|---|---|---|---|
| **FPR-022-056** | 0 (100% Sw) | 3.48 | 2.50 |
| **FPR-022-057** | 3 (~90% Sw) | 4.61 | 2.70 |
| **FPR-022-063** | 9 (~75% Sw) | 5.43 | 2.88 |

Further measurements will provide a more comprehensive overview of the absolute effect of desaturation on the diffusive behaviour of gases in clay based materials. Thus far, a brief correlation can be drawn from the measurements made to illustrate this effect.

**Contributor statement**

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