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Multi-scale investigation on gas transport behaviour of compacted granular bentonite under partially saturated states

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Compacted bentonites with high swelling potential and low permeability are under consideration as buffer/backfill materials in deep geological repositories of radioactive nuclear waste [1]. Within the short-term service period, the bentonite undergoes progressive saturation developing the swelling pressure, due to the water received from the host rock. Since the progressive saturation first occurs at the boundaries near the host rock, the bentonite close to the fuel canister could remain partially saturated for a long time. Within the long-term running stage, bentonite will withstand the accumulation and transport of gases such as H_2 (mainly generated by the anaerobic corrosion of the metallic overpack of canisters) [2]. The accumulated volume of gas can produce a gas pressure build-up that could break through the bentonite, damaging its microstructural integrity and isolation capacity for radionuclides and contaminants [3]. This issue may be improved by adopting granular bentonite (GB) with an extended particle size distribution (maximum grain sizes of the order of mm) [4]. The compacted GB has a high initial macroporosity, which might permit gas transport at low pressures with less impact on the microstructure. However, up to now, few investigations have focused on the gas transport behaviour of partially saturated compacted GB within a multi-scale perspective.

To this end, gas injection tests were performed on partially saturated compacted MX-80 type GB at different hydro-mechanical states. Effective gas permeability was measured in samples at the as-compacted state and in samples prepared at the as-compacted state and then partially saturated under isochoric conditions without additional vertical stress apart from the generated due to swelling. Additionally, the tests were also conducted on these two types of samples but subjected to initial vertical stress of 3.8 MPa. X-ray micro-tomography in combination with mercury intrusion porosimetry was employed to observe the microstructural characteristics of samples before and after gas injection. The results highlight that the pore network of compacted GB is constituted of inter-granular, inter-aggregate and intra-granular/aggregate pores. Increasing the as-compacted degree of saturation S_r reduces the proportion of inter-granular pores and the effective gas permeability $K_{a.eff}$ (Figure 1(a)). For the as-compacted sample, gas transport can induce the size of inter-granular/aggregate pores to extend (Figure 1(b)). When the initial vertical stress is applied on the as-compacted sample, $K_{a.eff}$ decreases, with the closure of inter-granular/aggregate pores. Although the progressive saturation of the as-compacted samples under constant volume causes the reduction of inter-granular/aggregate pores, they are mainly filled with the low-density bentonite gel and the accumulated gas pressure is able to reopen and connect them to form fissures (Figure 1(b)). Thereby, at a given S_r resulting from this saturation process, $K_{a.eff}$ is similar to that at the same as-compacted S_r (Figure 1(a)). However, the decrease in $K_{a.eff}$ is significant, when the progressive saturation is conducted under vertical stress. This is because many inter-granular/aggregate pores collapse during the saturation process, and the vertical stress restrains the formation of fissures driven by gas pressure. On the other hand, gas transport does not affect the distribution of intra-granular/aggregate pores within the studied hydro-mechanical states.

The current outcomes provide multi-scale insights into the gas transport behaviour of partially saturated compacted GB and emphasise its microstructural response to gas transport under different hydro-mechanical states. The values of $K_{a.eff}$ measured in the

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tested GB are higher than those obtained in powder bentonites but comparable to those of sand/bentonite mixtures at an equivalent porosity [5, 6]. The reason is the GB material presents the granular-type microstructure, with many interconnected inter-granular/aggregate pores, even at high S_r. Once the progressive saturation increases S_r to a value close to 1, $K_{a.eff}$ will sharply drop, attributed to the significant microstructural modification [7]. Therefore, the initial microstructure (before gas injection) of compacted GB plays a critical role in the gas transport behaviour. This microstructural effect was also confirmed in previous investigations of other soil types [8]. Furthermore, the present work underlines the microstructure of compacted GB is modified during gas transport, dependent on hydro-mechanical states, which can further affect the gas transport behaviour.

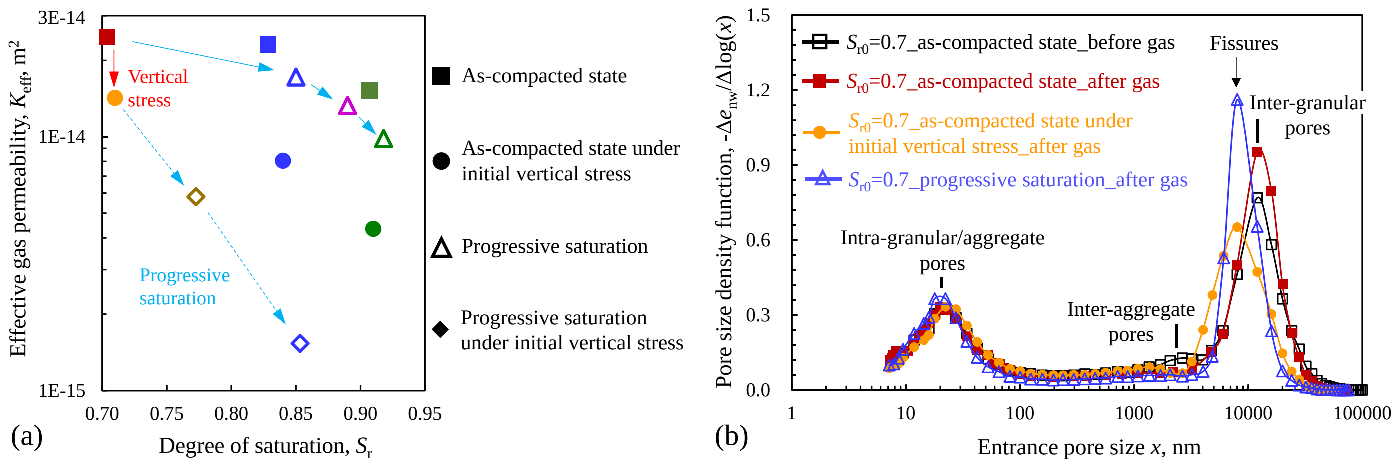

Figure 1: a) Effective gas permeability of GB samples under various hydro-mechanical states, b) Microstructural features of as-compacted GB and evolution after gas transport at different initial conditions.

Contributor statement

Hao Zeng: Conceptualisation, Data Curation, Formal analysis, Investigation, Visualization, Writing-Original Draft;
Laura Gonzalez-Blanco: Conceptualisation, Methodology, Funding acquisition, Supervision, Resources, Validation, Writing - Review & Editing;
Enrique Romero: Conceptualisation, Methodology, Funding acquisition, Supervision, Resources, Validation, Writing - Review & Editing.

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