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## **Semi-permeable membrane experiment for unsaturated liquid permeability of building materials: potential and practice**

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### **ABSTRACT**

The unsaturated liquid permeability is a crucial hygric property of porous building materials. Its direct measurement is very challenging however, because currently available experimental protocols are either demanding in test facilities, complicated in data processing, or limited in moisture content range. Inspired by the osmosis phenomenon, we study the possibility to apply semi-permeable membranes for directly measuring the liquid permeability of porous building materials. The key principle is studied profoundly and a simple experimental set-up is constructed. Trial tests on calcium silicate insulation and autoclaved aerated concrete are performed. Results show that the experimental set-up can be used to measure the liquid permeability, but strongly underestimates its value in its current form. Based on an in-depth analysis, we believe that the underestimation should be attributed to the pore clogging to the semi-permeable membrane due to the extremely high concentration of salt in the solution. Hence, in a future iteration, we will turn to unsaturated solutions with lower salt concentrations to alleviate the problem.

### **KEYWORDS**

liquid permeability, building material, semi-permeable membrane, hanging water column

### **INTRODUCTION**

Hygric properties are key characteristics of porous building materials. They are indispensable for analyzing the hygrothermal performance of building envelopes and the built environment (Defraeye et al. 2013; Zhao and Plagge, 2015). Of all hygric properties, the liquid permeability ( $K_l$ ,  $\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}\cdot\text{Pa}^{-1}$ ) is one of the most important.

Liquid permeability depends on moisture content. For (nearly) saturated samples, the liquid permeability can be easily measured by the water column test (Pedescoll et al. 2011) and the tension infiltrometer test (Zhao and Plagge, 2015). However, if the moisture content is lower, the direct measurement of the liquid permeability becomes rather difficult. One solution is to perform the Boltzmann transformation of the moisture content profiles visualized during a capillary absorption test for obtaining liquid diffusivity and subsequently liquid permeability. However, this protocol requires expensive experimental facilities – such as the X-ray set-up (Roels and Carmeliet, 2006) – and the data processing is complicated with large scatters. The drying test is another option for obtaining the liquid permeability below the capillary moisture content (Zhao and Plagge, 2015). However, this is basically an inverse method, requiring a lot of parameter tuning on the permeability model to approximate the measurement. It is therefore not straightforward and large uncertainties also exist.

From the review above, it's clear that current experimental protocols are either demanding in facilities, complicated in data processing, or limited in the moisture content range. Thus the

simple, reliable and full-range measurement of liquid permeability remains a challenging task. Inspired by the osmosis phenomenon, we investigate the possibility to employ the semi-permeable membrane for directly measuring the liquid permeability of porous building materials in a wide moisture content range. In this method, saturated salt solutions are used to exert a capillary pressure gradient across the sample, and semi-permeable membranes are used to protect against salts' interference. The permeability of the sample can hence be obtained from the measured liquid flux and the capillary pressure difference. Specifically, in the following section we will introduce the osmosis phenomenon. Next, our new experimental set-up with a semi-permeable membrane is described, and trial tests on calcium silicate insulation and autoclaved aerated concrete are explained. After that the experimental results are presented and analyzed. Finally, the future improvement of this method is discussed.

## METHODS

In this section, we will first introduce the osmosis phenomenon. Then our new experimental set-up with a semi-permeable membrane is described, and trial tests on calcium silicate insulation and autoclaved aerated concrete are presented.

### Osmosis phenomenon and the semi-permeable membrane

Osmosis is a common phenomenon in nature and has been widely adopted in many disciplines. As illustrated in Figure 1, when a solution and a pure solvent are separated by a proper semi-permeable membrane, the solvent particles can diffuse through the membrane in both directions while the solute particles are blocked. Due to the concentration difference of the solvent particles, a net flow of solvent from the pure solvent side towards the solution can be observed. This is the so-called osmosis phenomenon (Feher, 2017).

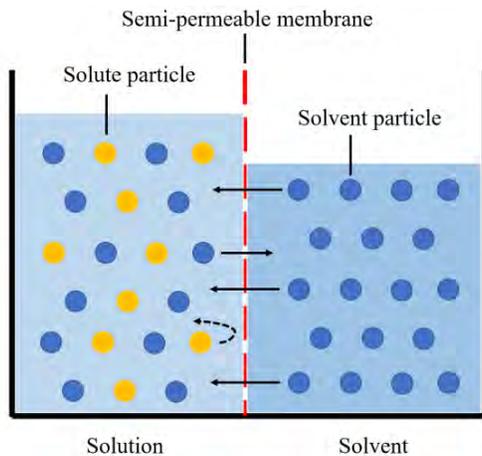


Figure 1. The osmosis phenomenon.

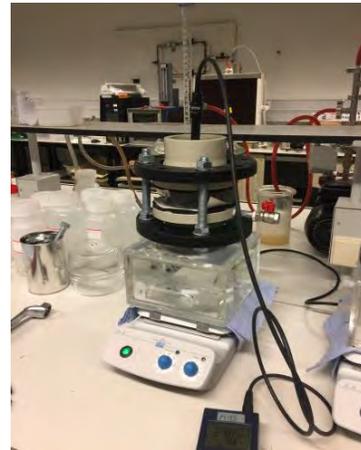


Figure 2. Photo of our semi-permeable membrane set-up

The osmosis phenomenon shows that a semi-permeable membrane exclusively allows the flow of solvent. It is therefore possible to use saturated salt solutions to exert a constant driving force for liquid water flowing through a sample, with semi-permeable membranes to protect the sample against salts' interference. The liquid permeability can thus be derived from the water flux and the capillary pressure difference. By altering the saturated salt solutions, the capillary pressure difference over and the moisture content in the sample can be controlled. This method is therefore in principle applicable to a wide moisture content range.

### Experimental set-up

Based on the principle explained in the previous section, a simple experimental set-up is designed and constructed (Figure 2 and 3). In this set-up, a sample of known cross-sectional area ( $A$ ,  $m^2$ ) and thickness ( $T$ ,  $m$ ) is laterally sealed with epoxy to obtain a 1-D liquid flow. The top of the sample is in direct contact with pure water, for which the capillary pressure ( $p_c$ , Pa) is assumed 0 Pa. The sample bottom sits on a semi-permeable membrane in contact with a saturated  $K_2SO_4$  solution with an RH of 97% at  $22 \pm 1^\circ C$  corresponding to a  $p_c$  of  $-3.6 \cdot 10^6$  Pa. The additional water head difference on both sides of the sample, originating from different water levels in the container and burette, is negligibly small relative to the used  $p_c$  difference ( $\Delta p_c = 3.6 \cdot 10^6$  Pa). Consequently, the capillary pressure difference is assumed the only driving force for the water flow through the sample.

When the flow is ongoing, the pure water in the water tank passes through the sample and the semi-permeable membrane, entering the solution tank, where undissolved salt exists and a magnetic stirring system is installed to keep the solution saturated all the time. In this way a constant  $\Delta p_c$  can be maintained and a steady flow can be reached after an initial period. The volumetric flow rate ( $G$ ,  $m^3 \cdot s^{-1}$ ) can be easily measured by reading the burette regularly. Given that the burette is part of the container with the saturated solution, the volumetric flow rate thus relates to the inflowing pure water and the additionally dissolved salt.

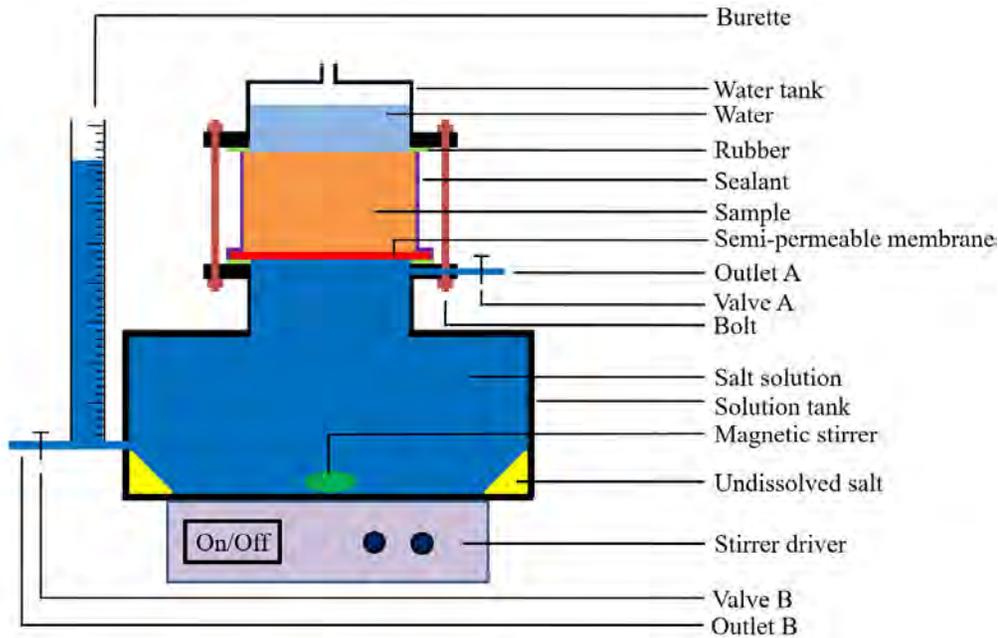


Figure 3. Schematic of our semi-permeable membrane set-up.

Assuming a certain amount of pure water ( $\Delta m_{water}$ , kg) passes through the sample and the membrane into the solution tank. The  $K_2SO_4$  dissolved by it ( $\Delta m_{K_2SO_4}$ , kg) amounts to  $s \cdot \Delta m_{water}$ , where  $s$  is the solubility of  $K_2SO_4$ ,  $kg(K_2SO_4) \cdot kg(water)^{-1}$ . Consequently, in the solution tank the increased saturated  $K_2SO_4$  solution ( $\Delta m_{solution}$ , kg) occupies a volume of  $\Delta V_{solution}$  ( $m^3$ ):

$$\Delta V_{solution} = \frac{\Delta m_{solution}}{\rho_{solution}} = \frac{\Delta m_{water} + \Delta m_{K_2SO_4}}{\rho_{solution}} = \frac{\Delta m_{water} + s \cdot \Delta m_{water}}{\rho_{solution}} \quad (1)$$

where  $\rho_{solution}$  is the density of saturated  $K_2SO_4$  solution,  $kg \cdot m^{-3}$ . Since previously undissolved  $K_2SO_4$  is now dissolved, its volume ( $\Delta V_{K_2SO_4}$ ,  $m^3$ ) should therefore be accounted for when calculating the net volume change ( $\Delta V_{net}$ ,  $m^3$ ) in the solution tank:

$$\Delta V_{net} = \Delta V_{solution} - \Delta V_{K_2SO_4} = \frac{\Delta m_{water} + s \cdot \Delta m_{water}}{\rho_{solution}} - \frac{s \cdot \Delta m_{water}}{\rho_{K_2SO_4}} \quad (2)$$

where  $\rho_{K_2SO_4}$  is the density of  $K_2SO_4$ ,  $kg \cdot m^{-3}$ . Now we define a coefficient  $c$  ( $kg \cdot m^{-3}$ ) by:

$$c = \frac{\Delta m_{water}}{\Delta V_{net}} = \frac{1}{\frac{1+s}{\rho_{solution}} - \frac{s}{\rho_{K_2SO_4}}} \quad (3)$$

At 22°C we obtain  $c=1009 \text{ kg} \cdot \text{m}^{-3}$ , meaning that 1  $m^3$  increase in the net volume of the solution tank (indicated by the burette) reflects 1009 kg of inflowing pure water. With the help of the known  $c$ , we can describe the mass balance of the transport process by:

$$G \cdot c = \frac{\Delta p_c}{R_{total}} \cdot A \quad (4)$$

where  $R_{total}$  is the total liquid transport resistance,  $m^2sPa \cdot kg^{-1}$ .  $R_{total}$  is the sum of the membrane's resistance  $R_{membrane}$  ( $m^2sPa \cdot kg^{-1}$ ) and the sample's resistance  $R_{sample}$  ( $m^2sPa \cdot kg^{-1}$ ):

$$R_{total} = R_{membrane} + R_{sample} = R_{membrane} + \frac{T}{K_l} \quad (5)$$

Combining Eq.(4) and (5), the liquid permeability of the sample can be finally derived:

$$K_l = \frac{T}{\frac{\Delta p_c \cdot A}{G \cdot c} - R_{membrane}} \quad (6)$$

After the test, the moisture content of the sample ( $w$ ,  $kg \cdot m^{-3}$ ) can be obtained gravimetrically, while  $R_{membrane}$  can be obtained from similar measurements without the sample in the set-up.

To completely fulfill the measurements in a wide moisture content range, a double-membrane set-up with two semi-permeable membranes separating two different saturated salt solutions on both sides of the sample is needed. At this trial stage, we simplify the set-up to the single-membrane system described above. This set-up restricts the applicable moisture content range to near saturation, but is adequate for preliminary validation. Its success will call for a slightly more complicated double-membrane set-up.

### Trial measurements

As two representative porous building materials, calcium silicate insulation and autoclaved aerated concrete are cut into duplicate cylindrical samples, with a diameter of 10 cm and a thickness of 2 to 4 cm. As a reference, falling-head water column tests are also performed for comparison. Due to the page limit of this paper, we will not explain the water column method specifically, and details can be found in (Pedescoll et al. 2011).

## RESULTS

Figure 4 illustrates the experimental results obtained from the semi-permeable membrane approach described in the previous section. Results from water column tests are also included for reference. As is clearly shown, for both calcium silicate insulation and autoclaved aerated concrete, the measured  $K_l$  reflects an expected moisture content dependence – the higher the  $w$  is, the larger the  $K_l$  is. However, it is very noticeable that the measured  $K_l$  values from two methods differ about 4 orders of magnitude. This is far beyond what common experimental errors can explain, and more profound reasons must exist.

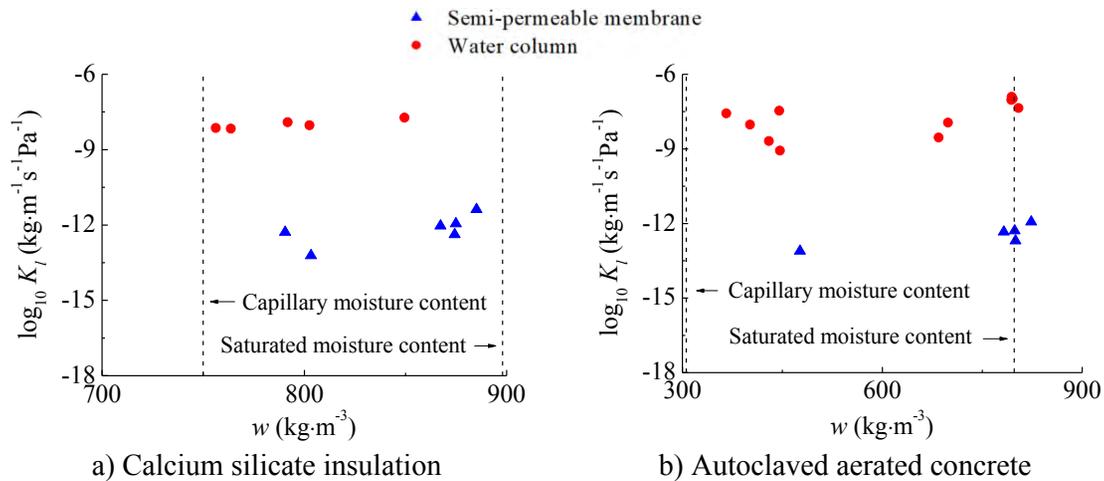


Figure 4. Experimental results of the semi-permeable membrane and water column tests.

## DISCUSSIONS

The water column test is a well-established method. Our water column measurements on other materials agree nicely with other data sources. Thus we believe the water column results in Figure 4 are trustworthy and the semi-permeable membrane approach underestimates  $K_l$ .

The first explanation for the underestimation could be that the transport area of the sample should be corrected for the masked sample edge in the set-up. However, the total diameter of the sample is 10 cm while the unmasked diameter is roughly 9 cm. Simple estimation reveals that the underestimation of  $K_l$  caused by the masked edge should be less than  $1-(9/10)^2 \approx 20\%$ , while from Figure 4 we should focus on the difference amounting to 4 orders of magnitude. Consequently, the masked edge is not a primary reason for the underestimation.

The second possibility is the change of driving force. Ideally no salt can pass the semi-permeable membrane, while in practice a small amount of salt may still penetrate through the membrane and the sample into the tank for pure water, reducing the real driving force and causing the underestimation of the final  $K_l$ . However, reducing the  $\Delta p_c$  to  $1/10^4$  of the assumed original value ( $3.6 \cdot 10^6$  Pa) means that the pure water tank above the sample should almost get saturated. The membrane used in this study has a NaCl rejection capability as high as 99.5%, while the salt used in our test is  $\text{K}_2\text{SO}_4$ , relatively more difficult to pass through the membrane. Consequently, it is unimaginable to assume an almost saturated solution in the upper tank due to the negligible salt transfer through the membrane. Direct measurements of the capillary pressure and electrical conductivity in both tanks also provide support.

Another explanation could be that the overall transport resistance is underestimated due to the existence of air layers on both sides of the membrane, which destroys the hydraulic continuity

of the system. An evidence is that the membrane resistance  $R_{membrane}$ , when measured separately by the semi-permeable membrane method and the water column method, comes to values of  $1.4 \cdot 10^{10} \text{ m}^2\text{sPa}\cdot\text{kg}^{-1}$  and  $1.1 \cdot 10^8 \text{ m}^2\text{sPa}\cdot\text{kg}^{-1}$  respectively. When there is only the membrane without the sample in the semi-permeable membrane set-up, the potential air layer between the membrane and the sample no longer exists, and this clearly reduces the difference between the measured  $R_{membrane}$  values from both methods. To furthermore validate this hypothesis, we turned the presently vertical system into a horizontal one for complete elimination of the air layers. Unfortunately, the newly measured  $R_{membrane}$  remained almost unchanged, indicating that the air layer should not be the most crucial reason.

The last and most plausible reason is that the pore clogging due to the extremely high salt concentration could happen to the semi-permeable membrane and hence increases the overall transport resistance. In our recent trials, we switched to unsaturated  $\text{K}_2\text{SO}_4$  solution, obtaining  $R_{membrane}$  around  $2 \cdot 10^9 \text{ m}^2\text{sPa}\cdot\text{kg}^{-1}$ , 10 times smaller than the saturated case. Encouraged by this supporting evidence, we plan to use unsaturated solutions of different salts and concentrations for further study and hopefully can succeed in the near future.

Albeit the semi-permeable membrane method is still under development, its principle is completely plausible. Theoretically, the liquid permeability below the capillary moisture content could be measured directly with two semi-permeable membranes on both sides of the sample, hence filling the gap of currently available experimental protocols.

## CONCLUSIONS

This paper proposes a novel experimental approach – the semi-permeable membrane method – for determining the liquid permeability of porous building materials. A simple set-up has been constructed and trial tests on calcium silicate insulation and autoclaved aerated concrete are performed. Traditional water column tests are also conducted as a reference. Results show that our current semi-permeable membrane set-up strongly underestimates the liquid permeability, most plausibly due to the pore clogging to the semi-permeable membrane. In the future we are going to use unsaturated salt solutions to solve this problem.

## ACKNOWLEDGEMENT

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