A CRITIQUE ON “A BOLTZMANN TRANSFORMATION METHOD FOR INVESTIGATION OF WATER VAPOR TRANSPORT IN BUILDING MATERIALS”

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ABSTRACT

In 2011, this journal published the paper “A Boltzmann transformation method for investigation of water vapor transport in building materials” (Pavlik et al., 2011), proposing a dynamic measurement of building materials’ vapour permeability via the Boltzmann-Matano method. This critique points out that this publication is flawed, since it proposes an invalid approach and presents erroneous results. The analysis first shows that the presented vapour permeability values cannot be reproduced and instead far lower vapour transport properties are obtained. These corrected outcomes are however also much below the dry-cup reference values, and it is established that these deviations stem from the invalidity of the proposed approach, due to the disregard for the material’s hygroscopicity. Given Pavlik et al.’s (2011) erroneous results and invalid approach, it is finally recommended to have the paper officially retracted by the journal.

KEYWORDS

vapour permeability, Boltzmann-Matano, dynamic experiment, retraction

1. INTRODUCTION

In 2011, this journal published the paper “A Boltzmann transformation method for investigation of water vapor transport in building materials” (Pavlik et al., 2011), proposing a dynamic measurement of building materials’ vapour permeability via the Boltzmann-Matano approach. The paper has remained below the radar for quite some time, with no independent references to it in the first five years after publication. That changed when a Chinese research group adopted the method to assess the impacts of porosity and temperature (Yi et al., 2016) and of sample length (Tian et al., 2019) on the vapour permeability of aerated concrete. And while the results of their first study did not arouse suspicion, their second study demonstrated that the sample length has to be chosen carefully to obtain reliable findings. That conclusion is in contradiction to the theory though, based upon which no impact of sample length is expected, as long as the sample is long enough to be regarded as a semi-infinite medium.

That contradiction is due to the flawed nature of the Pavlik et al. (2011) method, as reprocessing of the results of Tian et al. (2019) with the correct method (Carmeliet et al., 2004) invalidates the impact of the sample length as expected (Tian, 2020). This critique aims at pointing out the errors in the Pavlik et al. (2011) paper, which comprise both the proposed approach and the presented results. It is shown below that the vapour permeabilities reported in the paper cannot be reproduced here. Instead hugely lower values for the vapour transport properties are obtained, and successful verification indicates that Pavlik et al.’s (2011) results are erroneous. However, the corrected vapour permeability values obtained here are two orders of magnitude below the dry-cup reference values. It is demonstrated beneath that these deviations stem from the invalidity of the approach of Pavlik et al. (2011). In conclusion, given Pavlik et al.’s (2011) erroneous results and invalid approach, it is suggested to have the paper officially retracted by the journal.

2. CONCISE SUMMARY OF PAVLIK ET AL. (2011)

Pavlik et al. (2011) suggest to determine a material’s vapour permeability with a hygroscopic adsorption test, from which measured relative humidity profiles are Boltzmann-Matano transformed to vapour permeabilities. Their sole validation case applies a 350 mm long bar of Ytong P2-350 aerated concrete, wherein 16 mini-hygrometers are placed to measure the relative humidity profiles. The bar is initially preconditioned to 1.5 %RH, and its lateral surfaces are wrapped to get a one-dimensional configuration. The test starts when exposing the bar’s left surface to an environment at 79 %RH, and continues for 9 days. Its primary results are the relative humidity profiles at the end of each day, shown in Pavlik et al.’s (2011) Figure 3, reproduced here in Figure 1.

Figure 1: measured RH profiles in aerated concrete, with reconstructions for 1, 3, 9 days (blue, orange, green) and the simplification for 1 day (gold) superimposed.

These relative humidity profiles are processed into vapour permeabilities via a Boltzmann-Matano approach. Pavlik et al. (2011) describe the isothermal moisture transfer process with:

$$\frac{d\rho_v}{dt} = \frac{d}{dx} \left(\delta_p \frac{d\rho_v}{dx} \right) \quad (1)$$

with t [s] time, x [m] coordinate, ρ_v [kg/m³] vapour density, and δ_p [m²/s] vapour permeability for gradients of vapour density. This δ_p corresponds with the “vapour diffusion coefficient D ” in Pavlik et al. (2011), but our notation is preferred for clarity: the symbol D with unit m²/s can also be interpreted as a diffusivity, which combines the moisture storage and transport properties of the material. Transformations in Pavlik et al. (2011), see below, do however confirm that their D is a pure transport parameter. Based on the Boltzmann-Matano approach (Cerny et al., 2004), they then propose to quantify the vapour permeability δ_p as:

$$\delta_p(\rho_v \text{ or } \varphi) = \frac{\int_{x_0}^{\infty} x \frac{d\rho_v}{dx} du}{2t_0 \left(\frac{d\rho_v}{dx} \right)_{x_0}} = \frac{\int_{x_0}^{\infty} x \frac{d\varphi}{dx} du}{2t_0 \left(\frac{d\varphi}{dx} \right)_{x_0}} \quad (2)$$

with φ [-] relative humidity, t_0 [s] time point of the used vapour density or relative humidity profile, and x_0 [m] location at which the targeted ρ_v or φ is found in that profile.

The outcomes of Pavlik et al.’s (2011) application of equation (2) are illustrated in their Figure 4, revealing vapour permeabilities δ_p between $5 \cdot 10^{-7}$ m²/s and $10 \cdot 10^{-7}$ m²/s for the dry-cup relative humidity range of 10 % to 60 %. These are then transformed to vapour permeabilities δ_p [s] (for gradients in vapour pressure) ranging from $4 \cdot 10^{-12}$ s to $8 \cdot 10^{-12}$ s as well as to diffusion resistance factors μ [-] ranging from 42 to 21, see their Figures 5 and 6. All these results do however deviate strongly from dry-cup measurement findings, being $15 \cdot 10^{-7}$ m²/s, $11 \cdot 10^{-12}$ s and 15 for δ_p , δ_p and μ respectively (Pavlik et al., 2011). Nevertheless, even with these deviations being (on average) a factor 2, the authors consider this “a reasonable agreement”.

3. ERRONEOUS RESULTS IN PAVLIK ET AL. (2011)

Unfortunately, the author of this critique cannot reproduce the outcomes referenced above, and instead finds values that are far lower than these reported by Pavlik et al. (2011). To reveal that, the relative humidity profiles after 1, 3 and 9 days are first digitally extracted from Figure 1, and they are then analytically fitted with equations of the form:

$$\varphi(x) = (1 + \exp((x - A)/B))^C + D \quad (3)$$

The recreated profiles are shown in blue, orange and green in Figure 1, demonstrating the good approximation. The values for parameters A, B, C, D are collected in Table 1:

	1 day	3 days	9 days
A	0.0525	0.0833	0.3019
B	0.0238	0.0438	0.0854
C	-2.91	-2.17	-9.80
D	3.5E-04	6.1E-05	5.5E-03

Table 1: fitting parameters for relative humidity profiles

Application of equation (2) on these recreated profiles leads to the results shown in Figure 2. To obtain these, the integrals and differentials appearing in equation (2) are calculated numerically, by use of a very finely grained x-discretisation. There are two clear deviations from the findings of Pavlik et al. (2011), see their Figure 4. Firstly, there is more disparity between the outcomes for the different profiles here, but more importantly, the order of magnitude here is also drastically smaller. In Figure 2, vapour permeabilities δ_p range from $7 \cdot 10^{-9}$ m/s² to $13 \cdot 10^{-9}$ m/s² on average for the 10 % to 60 % relative humidity range. When transformed to vapour permeabilities δ_p and resistance factors μ , ranges from $5 \cdot 10^{-14}$ s to $10 \cdot 10^{-14}$ s and from 2000 to 4000 are obtained. Relative to Pavlik et al. (2011), the values here are almost two orders of magnitude lower.

Figure 2: vapour permeabilities δ_p in function of φ , from the RH profiles after 1, 3 and 9 days

Such large deviations demand further verification. To that aim, the relative humidity profile after 1 day is simplified to a straight line between two (x, φ) couples: (0, 0.73) and (0.065, 0), depicted in Figure 1 in gold. Applying equation (2) for a relative humidity of 40 % then yields:

$$\delta_p = \frac{\int_{x_0}^{\infty} x \frac{d\phi}{dx} du}{2t_0 \left(\frac{d\phi}{dx} \right)_{x_0}} = \frac{\int_{0.03}^{0.065} x \cdot 11 du}{2 \cdot 86400 \cdot 11} = \frac{[x^2/2 \cdot 11]_{0.030}^{0.065}}{2 \cdot 86400 \cdot 11} = 9.6 \cdot 10^{-9} \text{ m}^2/\text{s} \quad (4)$$

which confirms the order of magnitude of the results shown in Figure 2. Alternatively, assuming this vapour permeability δ_p of $9.6 \cdot 10^{-9} \text{ m}^2/\text{s}$ as a representative constant allows solving equation (1) analytically (Crank, 1975). For simplicity, the surface transfer resistance is ignored, bringing about this analytical solution:

$$\rho_v = \rho_{v,0} + (\rho_{v,1} - \rho_{v,0}) \operatorname{erfc} \left(\frac{x}{2\sqrt{\delta_p t}} \right) \quad (5)$$

with $\rho_{v,0}$ and $\rho_{v,1}$ [kg/m^3] the initial and final vapour density. When calculating the vapour density profile after 1 day, the location of the 40 %RH point on that profile is 0.028 m, closely matching the 0.03 m observed in Figure 1, again reconfirming the order of magnitude of the results shown in Figure 2. On the contrary, with δ_p $1 \cdot 10^{-6} \text{ m}^2/\text{s}$, representative for the (order of magnitude of) the Pavlik et al. (2011) results, this location would become 0.28 m, far higher than what is noted in Figure 1.

Regrettably, the author of this critique cannot establish how the outcomes of Pavlik et al. (2011) have been obtained, and can therefore not determine the processing errors that must have been made. Based on the verifications above however, it must be concluded that the results here are correct, implying that the findings of Pavlik et al. (2011) are flawed. Unfortunately, it is not solely the specific result that is erroneous, also the general method is invalid. This is demonstrated in the section below, and that also explains the large deviations between the results here and the reference dry-cup findings, which differ with a factor of about 100 to 200.

4. INVALID APPROACH IN PAVLIK ET AL. (2011)

The general equation for one-dimensional isothermal moisture transfer in building materials can be formulated as:

$$\frac{dw}{dt} = \frac{d}{dx} \left(D_w \frac{dw}{dx} \right) \quad (6)$$

with t [s] time, x [m] coordinate, w [kg/m^3] moisture content, and D_w [m^2/s] moisture diffusivity (for moisture content as potential). In an alternative version based on the vapour density, this becomes:

$$\frac{dw}{dt} = \frac{dw}{d\rho_v} \frac{d\rho_v}{dt} = \xi_p \frac{d\rho_v}{dt} = \frac{d}{dx} \left(\delta_p \frac{d\rho_v}{dx} \right) \quad (7)$$

with ξ_p [-] the vapour storage capacity (for vapour density as potential). Equation (7) differs from equation (1), adopted by Pavlik et al. (2011), on one essential feature: the moisture storage description in the left-hand side of the equations. By simply presuming the vapour density as descriptor of the mass storage in the porous material, Pavlik et al. (2011) completely disregard the material's hygroscopicity. And particularly for aerated concrete, but generally for porous materials, hygroscopicity has a critical impact on the moisture storage. At 40 % relative humidity (and 298.15 K temperature), the vapour density – and therefore Pavlik et al.'s (2011) assumed moisture content – is a mere 9.3 g/ m^3 . The actual moisture content for the P2-350 aerated concrete at 40 %RH is 0.02 kg/kg (Jerman et al., 2013) or 7.3 kg/ m^3 though, roughly a factor 800 higher. Alternatively, while the vapour density rises with 9.3 g/ m^3 when going from 20 %RH to 60 %RH, the actual moisture content rises with 3.6 kg/ m^3 (Jerman et al., 2013), about a factor 400 larger. These significant differences in the moisture storage also explain why the results obtained here for the vapour transport properties are much below the reference dry-cup values, with factor 100 to 200, see the earlier section. When accepting constant vapour permeability and capacity values, equation (7) translates to:

$$\frac{d\rho_v}{dt} = \frac{\delta_p}{\xi_p} \frac{d^2 \rho_v}{dx^2} = D_p \frac{d^2 \rho_v}{dx^2} \quad (8)$$

with D_p [m^2/s] the vapour diffusivity (for vapour density as potential). It is this diffusivity that controls the progressive penetration of the vapour density in the porous material, wherein both vapour storage and vapour transport play a role (Crank, 1975): higher vapour capacity or lower va-

Postprint: Janssen H. 2021. A critique on ‘A Boltzmann transformation method for investigation of water vapour transport in building materials’, *Journal of Building Physics*, 45:402-409.

doi: 10.1177/1744259120980041

pour permeability yields a slower penetration. In equation (1) however, the effect of the vapour capacity is strongly underrated (as ξ_p mostly is far larger than 1 in hygroscopic materials), implying that equation (2) translates the slowly penetrating vapour, see Figure 2, to highly underestimated vapour permeabilities. It must therefore be concluded that the approach proposed by Pavlik et al. (2011) is invalid.

This does not mean though that their suggestion is to be entirely annulled. Via equation (6), one could determine the moisture diffusivity D_w from the measured relative humidity profiles, if transformed into moisture content profiles through the adsorption isotherm, as:

$$D_w(w) = \frac{\int_{x_0}^{\infty} x \frac{dw}{dx} du}{2t_0 \left(\frac{dw}{dx} \right)_{x_0}} \quad (9)$$

which can then be translated to the vapour permeability δ_p with:

$$D_w = \delta_p \frac{dp_v}{dw} \quad (10)$$

This approach however necessitates the separate determination of the adsorption isotherm, and it additionally remains sensitive to the potential impact of the surface vapour transfer resistance. The latter issue has not been addressed explicitly here, but the Boltzmann-Matano transformation requires a fixed boundary value, which cannot be guaranteed in an hygroscopic adsorption test with a non-zero surface vapour transfer resistance. Given these concerns, this correction of the Pavlik et al. (2011) approach cannot be encouraged, and instead inverse hygric property determination (Vereecken et al., 2018) should (perhaps) be preferred. In this method, the vapour storage and vapour transport properties are determined simultaneously from short-term dynamic experiments, and their processing moreover allows explicitly considering the impacts of the surface vapour transfer resistance.

5. CONCLUSIONS

This critique has clearly shown that the work published in (Pavlik et al., 2011) is flawed, since it proposes an invalid approach and presents erroneous results. The analysis above first showed that the presented vapour permeability values could not be reproduced, and instead established vapour transport properties that are far lower. These corrected findings are however also much below the dry-cup reference values and it was demonstrated that these deviations stem from the invalidity of the proposed approach, due to the disregard of the material's hygroscopicity. Given Pavlik et al.'s (2011) erroneous results and invalid approach, the article no longer has value for the scientific community, and it is therefore recommended to have the paper officially retracted by the journal, to avoid inappropriate applications like in (Yi et al., 2016; Tian et al., 2019).

CONFLICTS OF INTEREST

The authors declare that they have no conflict of interest.

ACKNOWLEDGMENTS

This research was in part supported by the FWO Odysseus grant (G.0C55.13 N), which is gratefully acknowledged.

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