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Film flow in the water vapour permeability cup test

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Abstract

The Letter sets out the basis for including a contribution from liquid-water film flow to the so-called water vapour permeability [WVP] of porous inorganic materials. In this, the film thickness is derived from the water vapour sorption isotherm. A regression model is obtained for the dependence of the WVP resistance factor on the humidity. The model is tested against two published datasets. It is noted that available data are scarce, and that new precise measurements of the relation between the WVP and humidity are now required.

Keywords: Water vapour permeability, Film flow, Sorption isotherm.

1 Preamble

In our recent review [1] of the water vapour permeability [WVP] of inorganic materials, my co-authors and I noted the strong evidence that the water vapour resistance factor μ of many materials decreases as the relative humidity [RH] increases. The effect is observed at RH > 30 percent and upwards. We attributed this decrease to a contribution to water transport from flow in liquid films within the internal pore system of materials. This contribution was then included in a new regression equation for the dependence of μ on RH. In [1] the theoretical basis of this equation was sketched briefly in an appendix. It may be useful to set out the analysis more fully, and that is the purpose of this short paper.

Film flow has been discussed for some years as a component of unsaturated water transport in soils. Of course, liquid water is an essential component of the unsaturated state in soils (as it is in building materials). Going back even to Edgar Buckingham[1], and certainly to John Philip[2], water in the liquid state was viewed as occupying portions of the pore space. This water was described as capillary water and its behaviour was described by the physics of capillarity. Its thermodynamic potential was (largely or primarily) controlled by meniscus curvature and the Young-Laplace equation. It obeyed the physics of Darcy flow, and therefore was considered to be continuously connected throughout the region of interest.

Philip recognised that adsorption also occurred [4], but (I think) considered that sorbed water was immobile and did not contribute to unsaturated flow. The role of adsorbed water in water transport ("film flow") in soils was first recognised in [5]. It was considered to make the dominant contribution to the unsaturated (liquid) conductivity in relatively dry soils, broadly in the range of capillary pressure (suction) $|p_c|$ > 0.01 MPa. Other analyses of film flow in soils followed [6–10]. Only much later was film flow brought into building physics [11]. In all these treatments, the thickness of the adsorbed film was estimated by rather elaborate methods. It was our main innovation in [1] to suggest that the film thickness can be obtained directly, and more simply, from the water vapour sorption isotherm.

2 Mechanics of free-surface film flow

The film flow that we are considering is the motion of a thin adsorbed layer parallel to the surface on which it is adsorbed. The motion is assumed to be driven by the gradient of humidity across the test sample. We assume that there is a no-slip condition at the interface between the liquid film and the solid surface; and that the film is sufficiently thin that we

[1]Buckingham wrote in [2] "When a moist soil dries by exposure to the air over its surface the greater part of the evaporation into the air takes place at or very near the surface of the soil, the water moving up from below toward the surface by capillary action."

[2]Philip later wrote in [3] "Darcy's law may hold for the flow of liquid water in unsaturated media in a modified form in which K [the hydraulic conductivity] is a function of the volumetric moisture content, θ."

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can apply the lubrication approximation [12] to treat the flow as one dimensional.

We can adapt here the standard analysis of a falling liquid film, for example from [13]. For a thin liquid film (density ρ, viscosity η and thickness δ), moving in laminar flow down a vertical surface under gravity, we have the mean flux in the film $u_f = \delta^2\rho g/(3\eta)$. Here subscript f denotes the film. In what follows, we assume that the liquid is water.

For Darcy flow in porous media, we replace the gravitational potential gradient ρg by a capillary pressure [3] gradient $\mathrm{d}p_c/\mathrm{d}x$ to obtain for the mass flux

$$j_{\mathrm{mf}} = \rho u_{\mathrm{f}} = \frac{\rho\delta^2}{3\eta}\,\frac{dp_{\mathrm{c}}}{dx} \qquad (1)$$

If we compare Eqn. 1 with the defining equation for the mass-flow permeability [14] $j_{\mathrm{m}} = -k_{\mathrm{m}}\nabla p$ and identify p with $|p_{\mathrm{c}}|$, we see that the quantity $\rho\delta^2/(3\eta)$ is the film mass-flow permeability, dimension [T] as required (see also Appendix). We call this quantity k_{mf}.[4]

3 Obtaining the film thickness δ

The film permeability $k_{\mathrm{mf}} \sim \delta^2$, so a good estimate of the film thickness δ is required, ideally as a function of RH. For this we can use the experimental water vapour sorption isotherm, which gives data on the relation between the water content and the relative humidity (RH), generally over a wide range of RH. Here we use the symbol H for the fractional humidity RH/100. We note that $H = p_{\mathrm{w}}/p_{\mathrm{w0}}$ where p_{w} is the water vapour pressure and p_{w0} the saturated water vapour pressure, both at the temperature of interest. If sorption data are analysed using an isotherm model that includes the monolayer capacity as a parameter, then the thickness of the adsorbed layer is obtained directly. A suitable equation for that purpose is the Guggenheim–Anderson–de Boer [GAB] equation [1].[5] The procedure for extracting δ from isotherm data fitted to the GAB model is described in [1], and repeated verbatim below.

"The film thickness δ and its variation with fractional humidity H can be obtained from the water vapour sorption isotherm $F(H)$, where F is the mass fraction water content at H. . . . The GAB model . . . generally represents well the experimental $F(H)$ behaviour of inorganic construction materials up to high RH [14]. The GAB equation is

$$F = \frac{m_{\mathrm{G}}c_{\mathrm{G}}k_{\mathrm{G}}H}{(1-k_{\mathrm{G}}H)[1+k_{\mathrm{G}}H(c_{\mathrm{G}}-1)]} \qquad (2)$$

where m_{G}, c_{G} and k_{G} are dimensionless parameters. The quantity m_{G} is the mass fraction water content at which a complete monolayer exists. The corresponding fractional humidity at monolayer coverage is $H_{\mathrm{m}} = 1/[k_{\mathrm{G}}(1 + c_{\mathrm{G}}^{1/2})]$, which for inorganic construction materials lies typically in the range 0.15–0.35. The thickness of the monolayer water film is taken as δ_{m} = 0.27 nm from molecular dimensions. At higher humidities $H > H_{\mathrm{m}}$, the thickness of the water film $\delta = \delta_{\mathrm{m}}F(H)/m_{\mathrm{G}}$. If the specific surface area a_s is known, then $\delta(H) = F(H)/(a_s\rho)$. For the purpose of calculating the film thickness, the simpler formula $F(H) = m_{\mathrm{G}}/(1-k_{\mathrm{G}}H)$ is an excellent approximation." The simpler $F(H)$ function is compared with the full GAB function, Eqn. 2, in Figure 13 of [1].

4 Relating the hydraulic potential to the humidity H

Our aim is to add a contribution from film flow to the so-called [6] vapour permeability as measured on building materials. The mass flow water vapour permeability, generally denoted D_{v} in the building physics literature, is measured under conditions of controlled humidity gradient. Therefore we wish to express the gradient of capillary pressure p_{c} in Eqn. 1 in terms of humidity H. We do that using the standard relationship (in which M_{w} is the molar mass of water and T the absolute temperature):

$$p_{\mathrm{c}} = \frac{\rho RT}{M_{\mathrm{w}}}\ln H \qquad (3)$$

Hence, we have

$$\frac{dp_{\mathrm{c}}}{dx} = \frac{\rho RT}{M_{\mathrm{w}}H}\frac{dH}{dx} = \frac{\rho RT}{M_{\mathrm{w}}p_{\mathrm{w}}}\frac{dp_{\mathrm{w}}}{dx} \qquad (4)$$

We now have from Eqns. 1 and 4

$$j_{\mathrm{mf}} = \frac{\rho_{\mathrm{w}}^2\delta^2 RT}{3\eta M_{\mathrm{w}}\rho_{\mathrm{w0}}H}\frac{dp_{\mathrm{w}}}{dx} \qquad (5)$$

5 Film flow contribution to D_{v}

It remains only to include two geometrical approximations. First, to estimate the film-flow contribution to the overall mass flux in the cup test, k_{mf} is scaled by the film cross-section area per unit cross-section area of the bulk solid, $F_{\mathrm{v}} = F\rho_{\mathrm{b}}/\rho$ where ρ_{b} is the dry bulk density of the test material and ρ is the density (as elsewhere) of liquid water. Second, we apply also a geometrical tortuosity factor G by replacing x by x/G.[7] We can then finally write the film flow contribution to the water vapour permeability as

$$D_{\mathrm{vf}} = Gk_{\mathrm{mf}}F_{\mathrm{v}} = \frac{G\rho_{\mathrm{b}}\rho RT\delta_{\mathrm{m}}^2}{3\eta M_{\mathrm{w}}\rho_{\mathrm{w0}}m_{\mathrm{G}}^2}\frac{F^3}{H} = \beta\frac{F^3}{H} \qquad (6)$$

[3] For clarity, we note that the capillary pressure $p_c = \Psi_c\rho_w g$ where Ψ_c is the capillary (or hydraulic or matric) potential, ρ_w the density of liquid water and g the standard acceleration of gravity [14].

[4] In [1], the related Eqn. 12 should read $k_{\mathrm{mf}} = -j_{\mathrm{mf}}/(\mathrm{d}p_c/\mathrm{d}x) = \rho_w\delta^2/(3\eta)$.

[5] See also C Hall, Isothermal history, *Hall's Notes and Queries*, (2025), no 12, 12pp

[6] As noted in [14], in a WVP measurement "It is not safe to assume that water is transferred exclusively by molecular vapour diffusion . . . ". Some liquid-phase transport may contribute to the total mass transfer, as we envisage here that film flow does.

[7] The tortuosity of pore systems is discussed in some detail in [1]. Here $G \geqslant 1/\tau$, where τ is the geometric tortuosity as commonly defined [15]. $1/\tau$ is elsewhere [16] called the *connectivity* of the pore network. The factor *G* includes both a tortuosity component, since film flow must follow the meandering path of pore surfaces, together with a connectivity component to reflect discontinuities in the film.

The quantity β is independent of H, has dimension [T] as required, and depends on physical constants and on the physical properties of water and of the test material only.

The quantity $\alpha = 3\beta/(G\rho_b m_G^2) = \rho RT\delta_m^2/(\eta M_w p_{w0})$ depends only on the properties of water and physical constants. At 25 °C, $\alpha = 3.545 \times 10^{-12}$ m³ s/kg $= 1.913 \times 10^{-2} D_{v0}$ m³/kg, where D_{v0} is the Hellmann water vapour diffusivity in still air [1].

An important constraint on Eqn. 6 is that D_{vf} must be zero at and below some humidity $H = H_c$ close to but greater than H_m. This is to reflect the fact that the first molecular layer (the monolayer) is largely immobile [17]. We then have for $H \leqslant Hc$, $D_{vf} = 0$. In the GAB model, $F = m_G$ when $H = H_m$, so that we expect $H_c \approx H_m$.

6 Regression model for $D_v(H)$

To include the film flow contribution we write $D_v = D_{vv} + D_{vf}$ where D_{vv} is the value of D_v at low H where film flow makes no contribution (pure vapour transport). It is assumed that D_{vv} is independent of H. From Eqn. 6 we see that the D_{vf} depends on the fractional humidity H through the isotherm $F(H)$ by the relation $D_{vf} = \beta F^3/H$.

Therefore the water vapour resistance factor $\mu = D_{v0}/D_v$ including film flow is

$$\mu(H) = \frac{D_{v0}}{D_{vv} + \beta F^3/H} \qquad (7)$$

Then, using the approximate relation $F(H) = m_G/(1 - k_G H)$, we obtain from Eqn. 7 the regression model [1]

$$\mu(H) = \frac{H(1-CH)^3}{AH(1-CH)^3 + B} \qquad (8)$$

where A, B and C are regression parameters. In this model, the quantity C provides an estimate of $1/(4H_c)$; and the quantity $1/[A + (4^4/3^3)BC]$ provides an estimate of μ_{vv}, the vapour-only resistance factor at low RH.

As noted previously, Eqns. 7, 8 can only be applied at $H > H_c$, so the regression model over the full range of H must include piecewise also the relation

$$\mu = \mu_{vv} = \frac{1}{A + (4^3/3^3)BC} \qquad (9)$$

for $H \leqslant H_c$. H_c is found iteratively as $1/4C$. Eqn. 9 is found by setting μ_{vv} equal to the maximum of the function $\mu(H)$, Eqn. 8.

Some first attempts to apply this regression equation to two experimental $\mu(H)$ datasets are to be found in [1]. Here we rework the regression to the same data, but make use of recalculated GAB parameters, which in the case of CS include new datapoints at high RH (author's unpublished measurements). The sorption isotherms are shown in Figure 1 with fits to the GAB isotherm equation.[8] In this first order analysis minor effects of sorption hysteresis are neglected.

In Figure 2 we show the fits of the regression model of Eqns 8 and 9 to the WVP resistance factor μ data on the concrete [20] and calcium silicate sheet [22]. Available data with which to test the regression are extremely scarce. Even these two datasets are sparse, and uncertainties in the best-fit parameters A, B and C are large. The regression model does capture the rapid decrease in μ observed at $H > H_c$, but clearly further measurements of water vapour permeability over a range of imposed RH are now required.

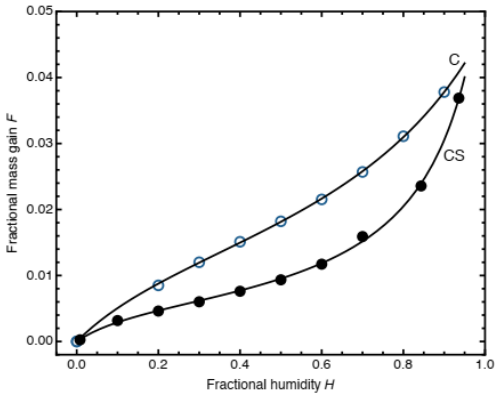

Figure 1: Sorption isotherms. Dense concrete C [18–20], ρ_b (2175 ± 40) kg/m³ [19]; m_G (1.580±0.018)×10^{-2}, c_G 5.718 ± 0.133, k_G 0.691±0.004. Calcium silicate sheet CS (author's unpublished data), 25°C, ρ_b 268±3 kg/m³ [21]; m_G (6.086±0.202)×10^{-3}, c_G 7.814±1.355, k_G 0.896±0.006. Isotherms are means of adsorption and desorption branches.

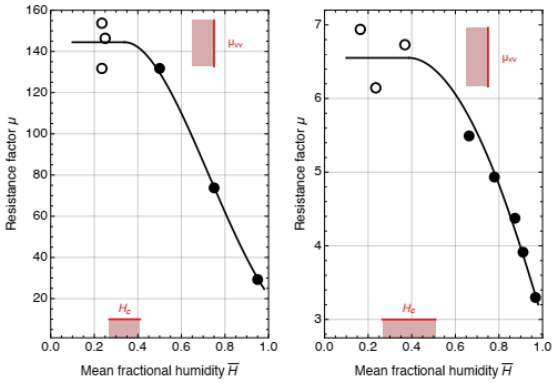

Figure 2: Variation of resistance factor μ with fractional humidity $\bar{H}$ = RH/100. Open symbols: dry state ($\bar{H} < H_c$); filled symbols: wet state ($\bar{H} > H_c$); solid lines: fits to regression model Eqns 8, 9. *Left* Dense concrete [19, 20]: ρ_b 2175 kg/m³ A = 1.10 × 10^{-3}, B = 8.34 ×10^{-4}, C = 0.736; μ_{vv} = 144.5 ± 11.2. *Right* Calcium silicate sheet [22]: A = 8.56 ×10^{-2}, B = 1.09 ×10^{-2}, C = 0.646, μ_{vv} = 6.55 ± 0.40.

The resistance factor attributed to film flow μ_{vf} can be calculated from Eqns. 8, 9 since $1/\mu_{vf} = 1/\mu - 1/\mu_{vv}$. Then

[8]While the overall adsorption capacity of these two materials on a mass basis is similar, the large difference in bulk density means that the dense concrete contains about ten times as much water per unit volume of material as CS at a given RH.

$$\mu_{vf} = \frac{H(1-CH)^3}{B[1-(4^4/3^3)CH(1-CH)^3]} \quad (10)$$

From μ, μ_{vf} and μ_{vv}, we see how film flow and pure vapour diffusion contribute to the overall permeability D_v, since

$$D_v = \frac{D_{v0}}{\mu} = D_{v0}\left[\frac{1}{\mu_{vv}} + \frac{1}{\mu_{vf}}\right] \quad (11)$$

The quantities D_v, D_{vv} and D_{vf} for the two materials are shown in Figure 3. In both cases, the contribution of film flow rises steeply at $H > H_c$ towards high humidities. The large difference in the pure vapour permeability D_{vv} of the dense concrete and the CS is due largely to the difference in porosity [1].

From Eqns. 8, 10 and 11, and for reference, the following equations relate D_v, D_{vv} and D_{vf} to the WVP regression parameters A, B and C

$$D_v = D_{v0}\left[A + \frac{B}{H(1-CH)^3}\right], \quad (12)$$

$$D_{vv} = D_{v0}\left[A + \frac{4^4}{3^3}BC\right], \quad (13)$$

$$D_{vf} = D_{v0}B\left[\frac{1}{H(1-CH)^3} - \frac{4^4}{3^3}C\right] \quad (14)$$

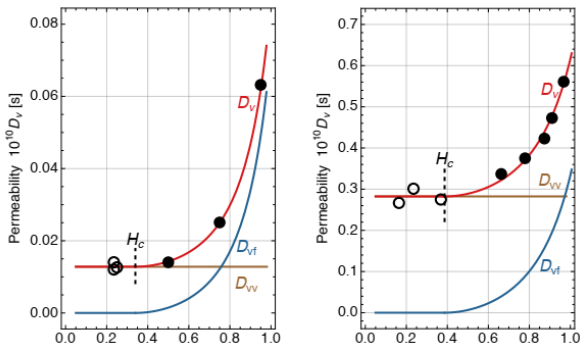

Figure 3: Variation of permeability D_v with fractional humidity H, showing also the components D_{vv} and D_{vf}. Solid lines: calculated from Eqns. 8–11 with values of regression parameters A, B and C as in Figure 2. Open symbols: dry state; filled symbols: wet state. Left Dense concrete [19, 20]: Right Calcium silicate sheet [22].

7 Regression parameters and GAB parameters

While the functional form of the regression model $\mu(H)$ of Eqn. 8 is suggested by the GAB isotherm equation, the quantities A, B, C are free parameters of the nonlinear least-squares fits to the WVP resistance-factor data. We expect nonetheless to find some relationship between these regression parameters and the GAB parameters.

First, it is expected that parameter $C \approx 1/k_G$, and the values of these quantities for both materials are given in Table 1. The uncertainty in C is considerable, given the paucity of data and its low accuracy. From C we also estimate the value of the critical humidity H_c below which film flow does not occur. In Table 1, we give the values of H_c from the WVP regression and the value of H_m from isotherm analysis. We expect to find that H_c and H_m are similar.

Second, and of greater interest, we note that the parameter B provides an estimate of the quantity $\alpha G \rho_b m_G/(3D_{v0})$, from which the tortuosity connectivity parameter G can be calculated. The argument is follows. From Eqn. 6, we have

$$\frac{dD_{vf}}{dz} = \frac{G\rho_b m_{0G}}{3}\alpha \quad (15)$$

where $z = I/[H(1 - k_G H)]$. The film-flow permeability D_{vf} so calculated depends only on the isotherm-derived quantities m_{0G} and k_G. On the other hand, from the WVP regression we have a second estimate of the permeability

$$\frac{dD^*_{vf}}{dz'} = BD_{v0} \quad (16)$$

where $z' = I/[H(1 - CH)]$. Now if we assume that D_{vf} and D^*_{vf} are estimates of the same quantity, we have

$$G = \frac{3BD_{v0}}{\rho_b m_{0G}\alpha}\frac{dz'}{dz} \quad (17)$$

The factor dz'/dz simply corrects for small experimental differences in the quantities k_G and C. The calculated values of the parameter G are given in Table 1. These values of G can only be regarded as semiquantitative, given the large uncertainties in the WVP regression parameters. However they confirm that the simple film-flow theory of Eqn. 6 gives contributions to the overall WVP permeability of the correct order of magnitude.

Table 1: Regression parameters and derived quantities: Comparison of dense concrete and calcium silicate sheet CS.

	Dense concrete [20]	Calcium silicate sheet CS [22]	Note
k_G	0.691 ± 0.004	0.896 ± 0.006	*a*
C	0.74 ± 0.15	0.65 ± 0.20	*b*
H_m	0.427 ± 0.004	0.294 ± 0.019	*c*
H_c	0.34 ± 0.07	0.39 ± 0.12	*d*
G	0.006	0.090	*e*

Notes: *a* GAB model regression parameter, Eqn. 2; *b* WVP regression parameter, Eqn. 7; *c* calculated from GAB relation $H_m = 1/[k_G(1+\sqrt{c_G})]$; *d* calculated from relation $H_c = 1/C$; *e* calculated from Eqn. 17 with the mean value of dz'/dz over the wet-state H range 0.40–0.95. Errors are $k = 1$ uncertainties where the data allow these to be calculated.

8 Caveats and comment

Several simplifications and assumptions should be mentioned.

- It is assumed that the pure vapour transport contribution to the WVP permeability is constant and independent of the humidity H. That is, both D_{vv} and μ_{vv} are constants. This is a reasonable first-order assumption, although there can be no doubt that the connected pore space available for vapour transport must experience some occlusion in the wet state. However, for CS liquid water occupies only about 1 percent of the total pore space at H = 0.95, the highest humidity considered. For this material, the occlusion effect is negligible. For the dense

concrete, the effect is no doubt larger, since liquid water occupies a larger part of the pore system at H = 0.95.

- We have noted that the first adsorbed molecular layer of water (the monolayer) is generally considered to be immobile. The WVP regression model does not attempt incorporate this feature, but identifies a critical humidity H_c below which there is no film flow. H_c is found from the regression parameter C = 1/(4H_c).
- It is long recognised that the WVP cup test is of poor accuracy and reproducibility [1]. A particular limitation of the datasets used here is that the boundary conditions of H on the test samples are often poorly controlled. The humidity gradient across the sample may also be rather large so that it has been necessary here to use a mean humidity $\overline{H}$ in the analysis. Future experimental studies should ideally measure the WVP in test arrangements in which the boundary conditions are accurately established and the H gradient is small.

The simple term *film flow* is now established in the soil physics literature [5, 7, 9], but is used (as it is here) to mean the transport of adsorbed liquid water on pore surfaces, whether this water exists as more or less continuous films or as poorly connected multilayer domains or perhaps even as disconnected clusters. In CS, which has an open fibrous microstructure, it is likely that adsorbed water forms continuous films on the surfaces of individual fibres but that these domains are connected (poorly but continuously) only at points of contact of individual fibres. It is such transport bottlenecks that are reflected in the values of G, described here as the tortuosity-connectivity parameter. The dense concrete, with a much lower porosity than CS, has complex microstructure and transport pathways for adsorbed liquid must also be convoluted. The topic is beyond the scope of this paper, absent further experimental study.

The GAB model has the feature that the film thickness at $H = 1$ is $\delta_1 = \delta_m c_G k_G/[(1-k_G)(1+k_G+c_G k_G)]$. For $c_G k_G \gg 1$, as is usually the case, this reduces to $\delta_1 \approx \delta_m/(1-k_G)$. This is an important feature of the GAB model, as for any $k_G < 1$ the film thickness at H = 1 is capped at some finite value, and does not rise to infinity (as it does for example in the BET isotherm model). This implies that the WVP regression equation may generally be applied to high RH (say $H \leqslant 0.95$), although not in the H range where Kelvin-type capillary condensation may be expected to occur (generally H > 0.99, but may be lower for nanoporous materials).

9 Conclusions

The content of this paper is to some extent speculative. It attempts to reconcile two well established facts. The first is that inorganic construction materials have films of adsorbed water on their interior pore surfaces over much of the humidity range of practical interest. The second is that the WVP resistance factor decreases markedly in wet-state measurements, say at RH > 0.5. The WVP datasets used are not only sparse but also of poor accuracy. The statistical conclusions are subject to considerable uncertainty. Even so, the regression model when applied to two markedly dissimilar materials embodies the main qualitative features of the relation between WVP resistance factor and humidity.

To further develop these ideas, new experimental WVP data are needed, together with supporting data on sorption and porosity. If progress is made in understanding how the WVP resistance factor varies with humidity, a day may come when the cup-test is no longer required for routine estimates of μ, and when the dependence of μ on H can be incorporated into hygrothermal models. To reach this requires also an understanding of the relation between μ_{vv} and porosity [1], the subject of another paper.

Appendix: Note on the mass-flow permeability

The mass-flow permeability k_m is often written (for example in [23]) with unit kg/(m s Pa), but this of course reduces to unit s, and dimension [T]. The question arises: if a material has permeability k_m = N s, to what process does the time N s refer?

Consider steady Darcy flow of fluid of density ρ through a permeable barrier of thickness L, with flux u for pressure drop Δp. Then $k_m = u\rho L/\Delta p = (u^2\rho/\Delta p)t_p = \gamma t_p$, where $t_p = L/u$ is the passage time for an element of fluid to pass through the barrier, and γ is the dimensionless group $u^2\rho/\Delta p$. It then follows that k_m has the value t_p when γ = 1. In other words, the numerical value of k_m = N s is the passage time under steady conditions such that γ = 1.

The group γ can also be written $\gamma = k_m^2 \Delta p/(\rho L^2)$, so that $t_p \sim 1/k_m$, thus explaining why t_p is larger the more permeable the material.

It should be noted also that $\gamma = \tau/\Delta p$ where $\tau = u^2\rho$ is the momentum flux.

Authorship statement (CRediT)

Christopher Hall: Conceptualization; Investigation; Writing – Original Draft; Writing – Review & Editing; Data Curation; Visualization.

Data availability

Data will be made available on reasonable request.

Declarations

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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