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FUNDAMENTAL ASPECTS OF FLUID FLOW THROUGH POROUS MATERIALS

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Synopsis An approach to the study of fluid flow through porous materials based on Darcy's law and the continuity equation is introduced as a possible means of studying fluid uptake by paper. The approach leads to a flow equation that may be put in the form of a diffusion equation with a diffusivity dependent on the fluid content. Hence, with the boundary conditions imposed during one-dimensional fluid uptake by a porous material initially at a uniform fluid content, the uptake is proportional to the square root of time, which is the same result as given by the Green & Ampt equation and the Lucas-Washburn equation, both of which use the analogy of fluid flow through capillary tubes.

Fundamentals

THE physics of the flow of fluids through porous materials has been developed by workers in soil physics and oil-reservoir engineering and it is now well understood. This work is applicable to many other subjects, but it appears that those working on fluid uptake by paper have taken little note of this work and instead have continued to use simple models of the paper's porous structure in their studies. This contribution attempts an introduction to this more basic approach to the study of fluid flow through porous materials. For greater detail, reference should be made to review $\operatorname{articles}^{(1-3)}$ and textbooks.⁽⁴⁻⁶⁾

Porous materials consist of assemblies of interconnected pores through which fluids can flow. Fundamental to a physical treatment of the flow of fluids through porous materials is the assumption that the geometry of a porous structure is repeated regularly throughout the porous body—that is, that the latter is made up of unit cells joined together. The size of the unit cell depends on the structure of the porous body. So long as a porous body is made up of several unit cells, the flow of fluid may be described by Darcy's law⁽⁷⁾—

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$$V = -K \operatorname{grad} \varphi \quad . \qquad . \qquad . \qquad . \qquad (1)$$

where V is the flow per unit area and ϕ is the potential, the sum of two components, one due to the fluid pressure p and the other due to the gravitational field. That is—

$$\varphi = p / \circ g + z$$
 (2)

where ρ is the density of the fluid, g the acceleration due to gravity and z the height measured from a given datum level. The proportionality constant K in equation (1) is the permeability of the porous material; in soils, where the wetting fluid is water, it is referred to as the hydraulic conductivity. Darcy's law describes the average flow behaviour of the fluid, not the microscopic flow in individual channels.



Fig. 1—Variation of hydraulic conductivity K with water content c for (a) a slate dust and (b) a beach sand

Flow movement

ALTHOUGH Darcy's law was originally formulated for water movement through saturated sands, unsaturated conditions often occur with air or 454

another fluid entering the porous body. Richards⁽⁸⁾ argued that, as far as the water conduction properties of unsaturated soils were concerned, the air-filled pores behaved as if they were solid, so that Darcy's law would be expected to hold for unsaturated soils as for saturated ones, but of course with a reduced value of conductivity. Experimental confirmation of this was given by Childs & Collis-George⁽⁹⁾ some years later. Measured values of the hydraulic conductivity as a function of water content of a slate dust and of a sand are shown in Fig. 1, where it is seen that the values are greatly reduced as the porous material becomes less saturated.

In addition to the variation of the conductivity as the fluid saturation changes, a further complicating factor occurs in the calculation of fluid flow. This arises because the pore-fluid pressure, one component of the potential, depends on the fluid content. In an inert porous material, the difference in pressure across a meniscus is given by the well-known expression—

$$p = T\left(\frac{1}{R_1} + \frac{1}{R_2}\right)$$
 (3)

where T is the surface tension of the interface, R_1 and R_2 are the maximum and minimum radii of curvature of the meniscus. Thus, the smaller the pores in which the menisci reside, the greater the pressure difference. For a fluid wetting the porous material, as in the general case of water in soils, the fluid pressure in an unsaturated porous material is negative. In addition, the relationship between the fluid content and fluid pressure shows hysteresis. An explanation of this hysteresis was given by Haines,⁽¹⁰⁾ who considered as a model of the pores in a porous body a capillary tube of varying diameter.



Fig. 2—The hysteresis in the relationship between water content *c* and pressure *p* for a model cylindrical pore

In Fig. 2*a*, the model cylindrical pore, when filled with water, drains when the negative pressure exceeds $2T/r_1$, but cannot fill until it is relaxed beyond $2T/r_2$. The hysteresis in the relationship between saturation and pressure of the single model pore is shown in Fig. 2*b*. For a porous material with a complicated interconnecting porous structure, the hysteresis is not so simple. An example is given in Fig. 3.



Fig. 3—The hysteresis in the relationship between water content c and pore-water pressure pfor a slate dust: the relationship during drainage from saturation and the subsequent closed loop for draining and wetting cycles with air entrapment are shown The dashed lines are primary scanning curves

When the flow of fluids in unsaturated porous materials is considered, it is obvious that the porous body may increase or decrease its degree of saturation and a second fluid must be displaced by the fluid or itself displace the fluid. The behaviour of the two fluids must be considered simultaneously in working out the fluid movements. In the case of water movement in soils, the second fluid is air, which has a much smaller viscosity than water has and its effect is negligible on the water flow so long as there is a free passage for air escape. Thus, in soil/water studies, it has become usual to neglect the air phase in calculations of soil/water movement. Then, considering the flow of water in an inert porous material, the continuity equation—

where c is the water content at a given position and time t may be combined with Darcy's law to give—

$$\frac{\partial c}{\partial t} = \operatorname{div} K \operatorname{grad} \varphi = \operatorname{div} K \operatorname{grad} (p/\rho g) + \frac{\partial K}{\partial z} \qquad . \qquad (5)$$

This is the basic flow equation that is used in calculating the changing water content of soils. The same equation may be used generally for fluid movement in porous materials when air is being displaced.

For one-dimensional horizontal flow, equation (5) becomes-

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x} \left[K \frac{\partial (p/\rho g)}{\partial x} \right] \qquad (6)$$

Writing $D = K \partial(p/\rho g)/\partial c$, this may be rewritten—

which is a diffusion equation in which the 'diffusivity' D is a function of the water content c. Equation (7) may be written so long as D is a monotonic function of c—that is, if only drying or wetting is occurring in the profile. If both wetting and drying are occurring, then hysteresis complications exist with discontinuities in D at the reversal positions. When water enters a dry porous material, D increases rapidly with water content, as shown in Fig. 4 for the example of a slate dust. If the porous material is initially moist, however, it is found that D is almost constant over a fairly large range of water contents, as shown in the case of the sand shown in Fig. 4.

The difference in the soil/water diffusivities in dry and moist porous materials is reflected in the water content profiles obtained during water uptake. When the diffusivity increases rapidly with water content, as when water enters a dry material, the wetting front is very steep (shown in Fig. 5a for water uptake by a dry slate dust). For the water uptake by an initially moist material, however, the wetting front is not so well pronounced (shown by the profile for the sand in Fig. 5b). These profiles are for the case when the



Fig. 4—Variation of wetting soil/water diffusivity D with water content c—(a) for water entering an air-dry slate dust and (b) for water entering a beach sand, initially brought to a water content $c_n = 0.086$ cm³ cm⁻³ by drainage

porous material is at a uniform moisture content initially and at t = 0; water is supplied at x = 0 so as to maintain a uniform water content there. These conditions allow the use of the Boltzmann transformation⁽¹¹⁾ $\lambda = xt^{-\frac{1}{2}}$, when the diffusion equation takes the form—

and the saturation profiles are given by unique plots of c against λ . If saturation profiles are determined experimentally, then the diffusivity may be obtained by evaluating at a number of water contents—

where c_o and c_n are the saturation and initial water contents, respectively. This method was first used by Matano⁽¹²⁾ for the diffusion of metals and has been used extensively in soil studies following initial work^(13, 14) some decades ago.



Fig. 5—Profiles of water content during water uptake (a) by an initially air-dry slate dust and (b) by a beach sand initially brought to a moisture content $c_n = 0.086$ cm³ cm⁻³ by drainage, when the surface at x = 0 is maintained at a constant water content of c_0 ; the profiles are plotted as functions of water content of the variable $\lambda = xt^{-\frac{1}{2}}$

Since the moisture profiles may be expressed as single plots of water content against the variable $\lambda = xt^{-\frac{1}{2}}$, the cumulative uptake *i* is given by—

$$i = \int_{c_n}^{c_o} x dc = t^{\frac{1}{2}} \int_{c_n}^{c_o} \lambda dc = At^{\frac{1}{2}} \quad . \qquad . \qquad . \qquad (10)$$

This means that the cumulative uptake is proportional to $t^{\frac{1}{2}}$. A similar relationship was deduced by Green & Ampt⁽¹⁵⁾ by considering that the

porous column was equivalent to a bundle of capillary tubes, as in the derivation of the Lucas-Washburn equation used in the uptake of fluids by papers. The proportionality constant A in equation (10) has been shown⁽¹⁶⁾ to be given to a good approximation by—

$$A = \sqrt{2(c_o - c_n)(S_o - S_n)}$$
 (11)

where

$$S_{i} = \int_{-\infty}^{p_{j}} K/\rho g dp = \int_{0}^{c_{j}} D dc \qquad . \qquad . \qquad . \qquad (12)$$

Thus, a physical interpretation to the constant in the Lucas-Washburn equation may be given in terms of the basic physical parameters of the porous material.

Conclusion

IN CONCLUSION, certain difficulties in applying this work on the physics of fluid movement through porous materials to paper are apparent. Since the flow equation is based on macroscopic considerations, it is important that any porous material contains a sufficient number of pores in the direction of fluid flow and that the porous body is sufficiently homogeneous (in the macroscopic sense). It could well be that many papers do not conform to these conditions, but the more basic approach to fluid flow in porous materials gives a physical insight into the process of fluid uptake by paper.

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Transcription of Discussion

Discussion

Chairman The coating of paper is an ancient art—the Chinese did it 2 000 years ago (surface pigmenting their paper), also the Turks in the 15th and 16th centuries (treble treatment). Machine coating was introduced in this country for the first time in the sixties of last century. Now the surface treatment of paper and board is a growth industry, with web widths on or off the machine up to 10 m for printing papers on an airknife coater and speeds up to 1 000 m/min on a blade coater. What is our scientific understanding of these paper treatments after doing them for the last 2 000 years?

Prof. B. G. Rånby I have one comment about the possibilities of making paper less permeable. As pointed out by Prof. Stannett, the only reasonable way is to put a coating on as a continuous surface layer. Any attempt to disperse particles or latex in the fibre suspension or impregnate the paper to decrease permeability are much less effective.

There is one more point I wish to make related to the problem of having polymers other than those now commercially used to decrease the permeability by surface coating. The new polymers that have been brought on to the market recently-plastics like Barex and Lopac containing acrylonitrile-are very impermeable, they have a high mechanical modulus and they can also be used within a wide temperature range, which makes them favourable as packaging materials. It has also been stated that these new materials have advantages from an environmental point of view: we have to be careful there. PVC develops hydrogen chloride when burned, which is not really poisonous, although it is an acid. We must look also at the burning of the acrylonitrile polymers: at low temperature with insufficient oxygen, they give hydrogen cyanide, which is poisonous. If you burn at high temperatures with an excess of oxygen present, these plastics develop nitrous oxides, which are rather detrimental to the environment. The same is true for other polymers containing nitrogen—polyamides, polyurethanes and native products like leather and wool. We have to define the burning conditions carefully for temperature and oxygen flow to prevent formation of poisonous and otherwise undesirable gases.

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Fluid flow through porous materials

Mr J. R. Parker I would like first of all to thank Dr Youngs for so kindly talking to papermakers about this rather complicated subject. I think his approach is of great value and this will become more apparent in the further papers to be presented.

Here, we have in fact a macroscopic measure of the behaviour of fluids in unsaturated porous materials. This has great advantages over the approach for which a model is needed. This is not to say that models are not of great interest and importance, but here is a means of describing the movement of liquids without making any assumptions about the nature of the material.

Dr Youngs' approach also gives the answer to a question posed by Dr Tollenaar many years ago, 'How can we explain the distribution of oil up a strip of paper during a capillary rise experiment?' He had to do this in terms of a rather complicated model based on interconnected capillaries. This experimental result, as we see now, can be subjected to quite a different sort of treatment and useful information can be obtained for application to other problems. This approach is relevant to the setting of inks on printing papers.

May I say a word of appreciation about Dr Oliver's contribution. His work reveals phenomena that are of great importance in the printability field.

Dr J. D. Peel Mr Gate, referring to your graphs from the mercury porosimeter apparatus, did your analysis of the results take account of the effect of the pressure of the mercury on the pore size distribution in the clay? The pressures were fairly high and would probably alter the pore size distribution as the experiment progressed.

Mr L. F. Gate This is quite a valid point. My answer to it is that we only get to about 20.7 MPa (3 000 lbf/in²), which may sound a lot, but the local distribution of pressure inside a coating will in fact be much less than that.

We have also looked at the forces necessary to disrupt a coating by using the Talystep. By increasing the loading on the stylus, a point is reached when you begin to disrupt the coating and a track is left behind as the stylus passes over it. From this, you can, from the loading on the stylus, the area of contact and the time of contact, get an estimate of the forces necessary to start disrupting the coating—and these in fact work out to be something of the order of 69 MPa (10 000 lbf/in²). By the fact that you can get pool values that agree with other methods, using materials you might expect to be crushed, you could come to the conclusion that it is not particularly important, as the local variation pressures within the coating under the intrusion pressures are not great.

Discussion

 $Dr \ E. \ L. \ Back$ With respect to both papers by Dr Youngs and Mr Gate, it might be appropriate to say a few words about the directional anisotropy of pores in paper. It means comparing the porosity and flow restriction in the Z-direction with that in the sheet direction. One way to quantify this comparison is by the effective pore radius for capillary flow by the Lucas-Washburn equation in these two directions. Fig. J shows the ratios of these two effective pore radii, that in the Z-direction divided by that in the machine-direction, for boards over a range of densities. These are rather thick boards made on Fourdrinier machines and the layered structure of paper increases this anisotropy. The ratio for the papers we tested is in the range of 0.01–0.04.

In these measurements, the same liquid is used in both directions, so the wettability does not enter this ratio. We found this to be true when we compared oil and water, so long as the papers were unsized or free from inherent resin.



The ratios in this case were the same, within ± 10 per cent. For sized papers and papers with a large amount of mechanical pulp with inherent resin(such as newsprint), even if a surface agent was added, the ratios fell another one or two decades with water compared with oil. This might indicate that there is an additional anisotropy superimposed when resin or rosin is added to the paper. The distribution of this oleophobic compound will thus change the pore anisotropy.

Pore anistropy is important when evaluating the paper by impregnation, as well as for all printing.