

## CAPILLARY RISE IN POROUS MEDIA

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**Synopsis** Capillary rise is one of the most complicated transport phenomena in porous media. So far there has been no theory which can claim to describe it adequately. As for fine powders, adequate experimental data in cases of imbibition do not even exist.

Both measuring and evaluation methods have, therefore, been developed by which the most essential parameters can be obtained. Tests on fine powders show that the capillary rise is decisively influenced by dynamic effects. Static capillary pressure—so far regarded as the driving force—can only be decisive at very low transport velocities. The penetration rate of liquid into fine powders is characterised by a dynamic capillary pressure, at least in the initial stages of high velocities.

The newly developed theory was tested with fine powders of different substances. The results confirmed the original assumptions. Whether, or to which extent, this theory can also be applied to fibrous materials, remains to be seen from tests still to be carried out.

### 1—*Introductory remarks*

THERE exist many publications on the capillary rise in porous systems, compiled and to some extent classified by van Brakel.<sup>(1,2)</sup> As a result of his experiments, van Brakel succeeded in determining the quality of many of the results which had, so far, met with some undefined opposition. The porous systems used by van Brakel were packings of particle size  $x = (100-1\ 000) \mu\text{m}$ . For the results reported in this paper, packings consisting of considerably smaller particles of the size range  $x = (10-100) \mu\text{m}$  were used. Only Newtonian fluids and porous systems which remained unchanged during experiments were used. Based on findings in the literature, a new kind of formulation of the capillary rise can be given for the materials used. It also becomes evident, however, that imbibition is strongly influenced by material properties, as well as by the shape and size distributions of the pores. It cannot, therefore, be taken for granted that the capillary rise in fine powders can be treated in

*Under the chairmanship of Prof. M. Judt*

the same way as liquid transport in fibrous materials to be dealt with at this symposium, the more so as it is by no means certain that paper remains unchanged when penetrated by liquids. Considering, however, that basically identical processes are involved here, it should be possible to derive from this contribution some suggestions for the capillary liquid transport in fibrous materials.

## **2—Classification of systems with capillary liquid transport**

CAPILLARY liquid transport in static porous systems can be distinguished by the characteristic features shown in Fig. 1.  $\dot{V}_1$  represents the volume flow rate of the wetting phase. A phase is described as wetting, if the effective contact angle  $\delta < \pi/2$ . In this case, pressure  $p_1$  measured in the immediate neighbourhood of the interface is, in the wetting phase, smaller than pressure  $p_2$  in the non-wetting phase.  $p_c = p_2 - p_1$  is defined as capillary pressure.  $\dot{V}_1$  is, according to Fig. 1, invariably a liquid volume flow rate.  $\dot{V}_2$  is the volume flow rate of the non-wetting phase, in this case the volume flow rate of air. We distinguish between drainage and imbibition of porous media. Mechanical drainage is made possible by an external field—for instance, centrifugal field—or by external pressure. In this case the liquid is displaced by the air. For an effective pressure drainage, a diaphragm should be used that is permeable for the liquid only.

From the distribution of liquid—shown schematically in Fig. 1—the essential differences of the two processes can be seen. In the case of pressure drainage, a practically uniform distribution of the liquid is observed above the height of the porous system, while with drainage caused by an external field, the liquid distribution is always irregular.

During the drying process, the liquid is first drained from the upper surface of the packing, while reverse air currents flow into the drained pores. Only in cases where rapid package driers are used, liquid is removed from all areas of the porous system by the drying processes. In this connection, the counter-flow principle is a characteristic feature of volume flows  $\dot{V}_1$  and  $\dot{V}_2$ , though it remains confined to smaller pore areas.

The opposite occurs in imbibition where liquid penetrates into the porous system by capillary action while air escapes from the pores in parallel flow, with air bubbles left in the porous system. The liquid may also be pressed into the pores by an external field or by pressure.

As can be seen schematically from Fig. 1 the distributions of the liquid in the various cases differs in a characteristic manner.

## **3—Models describing capillary liquid transport**

SINCE a description of capillary liquid transport is rather complicated,

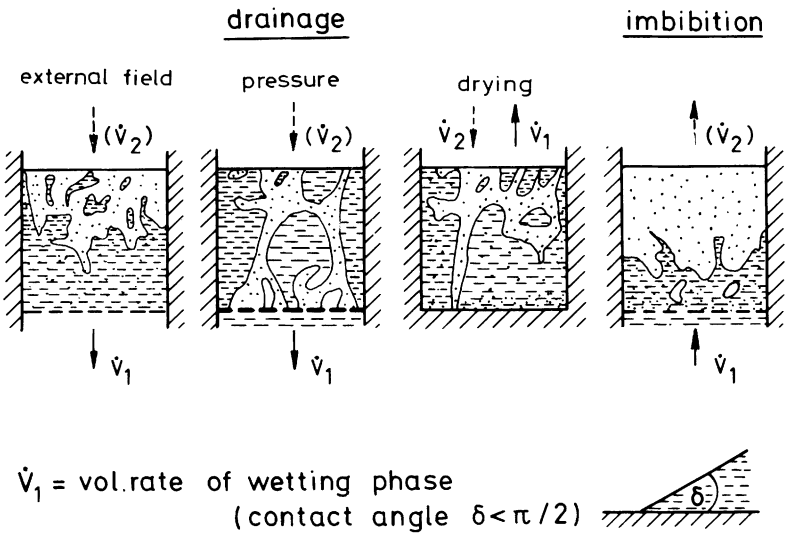


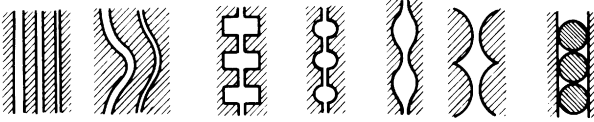
Fig. 1—Diagram of transport of capillary flow in porous media

attempts have been made to describe the pore space with the aid of models. Fig. 2 shows some of the proposed models which can be classified as one-dimensional, two-dimensional and three-dimensional. Cylindrical capillary tubes of different radii, in parallel or tortuous, are most frequently used. Each packing consists, however, of pores connected to one another by pore necks. The other models shown provide, therefore, a much better reproduction of the actual porespace. Very comprehensive computations of capillary action and of permeability were sometimes required in such cases.<sup>(3)</sup>

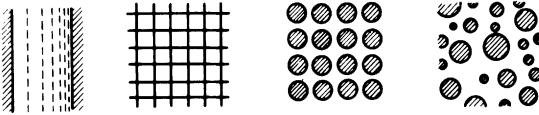
Krischer<sup>(4)</sup> proposed as a model cylindrical capillaries interconnected without mutual resistance. Since cross transport is allowed for here, this model can, in a wider sense, be described as two-dimensional. In Germany, this model is frequently used for the description of fluid motions during drying processes; Sommer<sup>(5)</sup> also used it for imbibition while taking into account the existence of a resistance between the single pore cavities. The main disadvantage of this model is that a succession of cavities connected by windows is not taken into account. The other models, including the two-dimensional network, allow for various cross-sections along continuous pore cavities.

porespace models

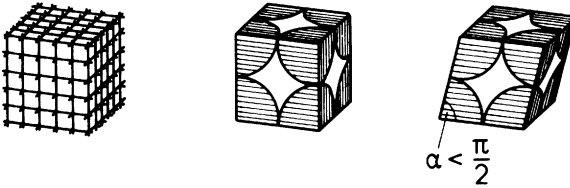
1) one-dimensional



2) two-dimensional



3) three-dimensional



**Fig. 2**—Proposed models for the description of the porespace of packings (van Brakel, 1975)

The three-dimensional models are of a much more complicated, though more realistic, structure. Particularly the models of Freval<sup>(6)</sup> and those of Mayer and Stowe<sup>(7)</sup> have gained recognition for the analysis of pore structures.

In both cases the models are based on regular sphere packings. As a case in point, Fig. 2 (in the centre of the three-dimensional models) shows a section of a cubic packing. Eight spheres form the cavity connected by 6 windows. Since the cubes can be inclined towards each other, oblique-angled structures are formed, with the result that packings with different porosities can be simulated. Experiments have shown, however, that even these models are not suitable for the computation of the capillary pressure in porous media.<sup>(8)</sup>

As a general rule it may be stated that all models are rather problematical for the formulation of quantitative statements. Models that need no fitting

parameters and can be used for computations are an exception. The great advantage provided by models of the kind described is the fact that they make it possible to describe the phenomenology of complicated processes in a satisfactory manner. If models are not suitable for the reproduction, in a fundamentally correct way, of the phenomenological behaviour, they are of questionable value, particularly because of the risk of misinterpretations. Normally, simply and clearly structured models should be used. On the other hand, models of extremely simple construction are liable to describe important processes wrongly or unsatisfactorily. This applies to the model of cylindrical capillaries as it is still used at the present time for characterising the pore-space in packings for all capillary liquid transports. As recently demonstrated,<sup>(9)</sup> this model is not suitable for the description of capillary liquid transport phenomena. Capillary rise is the most complicated one. Van Brakel<sup>(1)</sup> has illustrated that none of the porespace models proposed in the literature is capable to explain, not even qualitatively, the phenomena observed with capillary rise.

**4—Liquid transport in relation to the capillary pressure curve and the relative permeability**

As a result of capillary pressure, liquid penetrates into a porous system. The resistance of the porous flow counteracts capillary pressure. By equating the two forces, we obtain the law of the description of capillary rise. Forces of inertia will, in general, be negligible in capillary rise.<sup>(10)</sup> In very fast wetting processes, however—as they occur in the printing of paper—forces of inertia should be taken into account.

Capillary pressure within porous media can be described by the capillary pressure curve which is the capillary pressure  $p_c$  as a function of the degree of liquid saturation  $S$  ( $S = \text{liquid volume/pore volume}$ ). Fig. 3 shows a typical capillary pressure curve. All physical conditions are found between drainage and imbibition. The hysteresis is governed by the existence of pores of irregular shape consisting of cavities and windows, and by the contact angle hysteresis. The capillary pressure curve indicates a state of equilibrium, i.e. the static capillary pressure. The capillary pressure curve  $p_c(S)$  can be measured with the aid of suitable equipment (cf., for example<sup>(11)</sup>).

The reciprocal resistance of the porous flow—the so-called permeability  $B$ —can also be measured.  $B$  is strongly influenced by the degree of liquid saturation  $S$ . The simple product formulation

$$B(S) = B_r(S) \cdot B \quad \dots \quad (1)$$

is in regular use.

$B_r(S)$  is the relative permeability. For completely filled pores, i.e.  $S = 1$ ,  $B_r(S = 1)$  becomes 1; for sufficiently low liquid saturation,  $B_r$  tends to zero.

For isotropic, homogeneous porous media, the law of resistance can be formulated by equation (1) if forces of inertia are negligible and Newtonian fluids are considered. We have to take into consideration the liquid transport only, the effect of gases being negligible. According to Darcy's law, the following equation holds for the velocity  $v$  of the free cross section of a unidimensionally flowing liquid in the gravitational field of the earth

$$v = \frac{-B}{\eta} B_r(S) \left[ \frac{dp}{dy} + g\rho \cos \alpha \right], \quad (2)$$

where  $\eta$  = dynamic viscosity of liquid

$p$  = pressure

$y$  = coordinate in Fig. 4

$g$  = acceleration due to gravity

$\rho$  = liquid density

$\alpha$  = angle of direction of flow against acceleration due to gravity (see Fig. 4).

Taking into account the law of conservation of mass

$$\frac{\partial(\varepsilon\rho S)}{\partial t} = -\frac{\partial(\rho v)}{\partial y} \quad (3)$$

where  $\varepsilon$  is the porosity and  $t$  the time, and further introducing the diffusivity,

$$D(S) = -B_r(S) \frac{B}{\eta} \cdot \frac{dp_c}{dS}, \quad (4)$$

we obtain for  $\rho = \text{const}$  the partial differential equation

$$\varepsilon \frac{\partial S}{\partial t} = \frac{\partial}{\partial y} \left[ D(S) \frac{\partial S}{\partial y} \right] + \frac{Bg\rho \cos \alpha}{\eta} \cdot \frac{\partial B_r(S)}{\partial y}. \quad (5)$$

This equation, ascribed to Richards,<sup>(12)</sup> is normally used for dealing with problems in petroleum production and soil science. Detailed descriptions are contained in the papers of Philip,<sup>(13)</sup> Swartzendruber,<sup>(14)</sup> Morel-Seytoux,<sup>(15)</sup> Wooding and Morel-Seytoux.<sup>(16)</sup>

Equation (5) describes the local and time-dependent distribution of liquid during capillary permeation. Apart from the aforementioned premises, equation (5) is limited to conditions where there is a definite, differentiable relationship between the capillary pressure and the degree of liquid saturation.

Because of the capillary pressure hysteresis, this condition is not always met.<sup>(17)</sup>

Methods for solving the partial differential equation (5) have been given by Philip.<sup>(13)</sup> The results are contained in Chapter 5.2 of this paper. If the

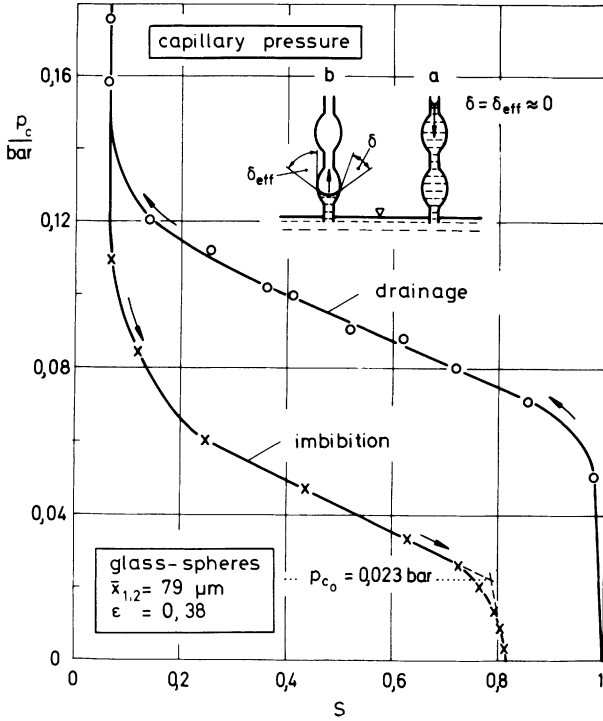


Fig. 3—Capillary pressure curve of a packing of glass spheres of mean diameter  $\bar{x}_{1,2} = 59 \mu\text{m}$  and porosity  $\epsilon = 0.38$

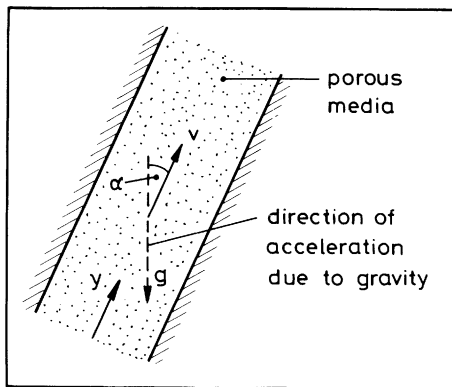
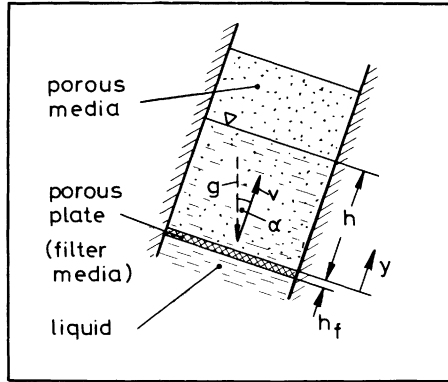


Fig. 4—One-dimensional liquid transport in porous media



**Fig. 5**—One-dimensional capillary rise in porous media

capillary pressure curve  $p_c(S)$  and the relative permeability  $B_r(S)$  are available as experimental values for the whole region, they can be used to describe the capillary rise in porous media.

On the other hand, equation (5) has one serious drawback of which no notice has been taken in the literature. The static capillary pressure gradient  $dp_c/dy$  is considered to be the driving force. As will be demonstrated, however, it is not the static but the dynamic capillary pressure that is of decisive influence in sufficiently fast transport processes. In imbibition, the dynamic capillary pressure is invariably smaller than the static pressure. The main reason for capillary pressure dynamics is the dynamic contact angle which, with an advancing three-phase line depending on the velocity, reaches higher values than the static contact angle.

Analogous to equation (5) we could formulate a partial differential equation with regard to the dynamic capillary pressure. We will not, however, quote the equation here, because we know of no simple solutions; nor do we see a possibility to measure the data which would be required. Instead, we will give a simplified description of capillary rise.

## 5—Simplified description of capillary rise

### 5.1—Capillary rise at constant capillary pressure

FOR one-dimensional imbibition as shown in Fig. 5, with regard to acceleration due to gravity  $g$ , the following simplifications are used:

- Movement of a sharp liquid front under constant capillary pressure  $p_c$ .
- The liquid completely fills the pores, i.e.  $B_r = 1$  for  $y \leq h$  ( $B_r = 0$  for  $y > h$ ).
- Darcy's equation (2) is applied to the liquid, with the influence of gas being negligible.



- (d) The filter medium is filled with liquid.
- (e) The pressure is the same at  $y = h$  and  $y = -h_f$ .
- (f)  $h_f \ll h$ , i.e. the gravimetric effect in the filter medium is negligible.

As experiments for the determination of the capillary rise in powders cannot be done without a solid porous plate (filter medium), the filter resistance

$$R_f = h_f/B_f \quad (6)$$

must be taken into account, where  $h_f$  is the filter thickness (Fig. 5) and  $B_f$  is the permeability of the filter medium. Since  $v = \epsilon dy/dt$  and  $dp/dy = -p_c/y$ , the following equation is obtained from equation (2):

$$\frac{dy}{dt} = \frac{B}{\epsilon\eta} \cdot \frac{p_c - g\rho y \cos \alpha}{y + R_f B} \quad (7)$$

The integration of this equation with the initial condition  $y = 0$ , at time  $t = 0$ , yields, because of  $p_c = \text{const.}$ ,

$$\left(\frac{p_c}{g\rho \cos \alpha} + R_f B\right) \ln \left(1 - \frac{g\rho h \cos \alpha}{p_c}\right) + h = \frac{-Bg\rho t \cos \alpha}{\epsilon\eta} \quad (8)$$

Equation (8) describes in explicit form the dependence of the penetration height  $h$  of the liquid in a packing on the penetration time  $t$ . For  $\alpha = \pi/2$ , or for negligible gravimetric effect, equation (8) can be simplified to

$$h = \sqrt{\frac{2Bp_c t}{\epsilon\eta} + B^2 R_f^2 - BR_f} \quad (9)$$

For  $R_f = 0$  we obtain the well-known relation of Washburn<sup>(18)</sup>

$$h = \sqrt{\frac{2Bp_c t}{\epsilon\eta}} = \text{const} \cdot \sqrt{t} \quad (9a)$$

In many experiments it is not the penetration height  $h$  that is determined, but the total liquid by volume

$$V = \epsilon h A \quad (10)$$

penetrated up to time  $t$ , where  $A$  is the cross sectional area of the porous medium located perpendicular to the direction of flow. Equation (9a) will then read

$$V = A\sqrt{2\epsilon Bp_c t/\eta} = \text{const} \cdot \sqrt{t} \quad (9b)$$

### 5.2—Quasistatic capillary rise

Sufficiently slow transport processes can be detected by equation (3). In such a case the static capillary pressure is the determining factor, hence the process is described as quasistatic capillary rise. With the gravimetric effect being negligible, the solution of equation (5)<sup>(13)</sup> is

$$V = \epsilon A Q_1(S_1, S_0)\sqrt{t} = \text{const} \cdot \sqrt{t} \quad (11)$$

The value of  $Q_1$  is computed as a function of the initial degree of saturation  $S_0$  and the saturation degree  $S_1$  at  $y = 0$ , after a computing process found by Philip. A comparison of equations (9b) and (11) shows the same relation  $V = \text{const} \sqrt{t}$ , with only the constant differing.

If the gravimetric effect must be taken into consideration, we can use the series expansion<sup>(13)</sup>

$$V = \varepsilon A(Q_1 t^{1/2} + Q_2 t + Q_3 t^{3/2} + Q_4 t^2 + \dots), \quad (12)$$

The values  $Q_i$  can be computed by approximation.

### 5.3—Summary of experimental results

On the strength of the tests carried out by van Brakel<sup>(1)</sup> and our own experiments the following two critical cases can be identified between which any transition is possible.

- (a) For a contact angle  $\delta > \delta_{\text{crit}}$  or for sufficiently fast penetration velocities, a sharp liquid front will occur; the porous system can be completely filled with liquid ( $S \approx 1$ ).
- (b) For a contact angle  $\delta \approx 0$  and a sufficiently slow wetting process, or for pre-wetted porous systems, where the liquid in the pores is hydraulically interconnected, a distribution of liquid is formed in the packing, with isolated gas bubbles left in it, i.e. the porous system cannot be completely filled with liquid ( $S < 1$ , often  $S = 0.8-0.9$ ).

Case (b) can often be satisfactorily dealt with as quasistatic wetting, according to the conditions stated in chapter 5.2. A typical application is infiltration or underground irrigation of soils. In many cases, the conditions of  $\delta \approx 0$  or a rather slow wetting process are fulfilled, and often pre-wetted soils are available. The wetting velocities of  $v < (10^{-2}-10^{-4}) \text{ cm/s}$ <sup>(13, 14)</sup> can be regarded as sufficiently low. Often a satisfactory consistency is obtained between theory and practical experiment. Variations stated at times are attributed to the effect of air bubbles in the pores.<sup>(16)</sup> It cannot be excluded, however, that dynamic effects must be taken into consideration. Dry layers of soil can be saturated by a sharp liquid front ( $\delta > \delta_{\text{crit}}$ ). A compilation of experimental results obtained by several authors shows that with the water-and-sand system, all kinds of imbibition are feasible.<sup>(1)</sup>

For all problems relating to processing technology which are of interest here, case (a) is more significant than case (b). In most cases, imbibition processes are involved which must be completed within a minimum of time, as the imbibition of agglomerates of so-called 'instant' products, coloured pigments, etc., where a quick dispersion or solution of particles

is needed.<sup>(19, 20)</sup> Such ‘quick’ imbibition processes may occur even at wetting speeds of  $v = (0.1-1)$  mm/s. As a rule, the porous systems contain no hydraulic moisture before wetting. For describing these processes, Washburn’s equation (9a) has, so far, been exclusively used. From experimental results so far published and his own measurements, Schicketanz<sup>(21, 22)</sup> detected, however, a considerable discrepancy between equation (9a) and the test results. Schicketanz proposes to use the formulation

$$h \propto t^u \dots \dots \dots (13)$$

instead of Washburn’s equation, where  $u$  may assume values ranging from 0.5 to 1, provided the gravimetric effect is negligible. For short penetration times  $t \leq 10$  sec,  $u \rightarrow 1$  is observed, and only after longer penetration times, the exponent  $u$  in equation (13) will assume the value  $u \approx 0.5$ .

It can be demonstrated (Schubert, in preparation), that the explanations so far given in the literature about the discrepancy between the statement of equation (9a) and experiments cannot be right. This makes it necessary to find a new formulation for capillary liquid transport in the imbibition of dry porous systems.

5.4—Capillary rise considering dynamic capillary pressure

On the assumption that a liquid transport takes place with a sharply defined wetting front, and with  $S = 1$  in the wet area, equation (7) can be applied. Equation (7) cannot be easily integrated if it is assumed that it is not the static, but the dynamic capillary pressure which is of decisive importance. The dynamic capillary pressure is a function of the velocity distribution, viewed microscopically, at the wetting front. In a first approximation, the mean pore velocity

$$v_p = \frac{dy}{dt} = \frac{v}{\varepsilon} = \frac{\dot{V}}{\varepsilon A} \dots \dots \dots (14)$$

can be regarded as the determining velocity. The dynamic capillary pressure can then be formulated by the statement  $p_c = p_c(v_p)$ . A typical curve  $p_c(v_p)$  is shown in Fig. 6 for the case of a packing of glass spheres of a mean diameter  $\bar{x}_{1, 2} = 79 \mu\text{m}$ . Over a wide range the curve can be approximated by the relation

$$p_c = p_{c0} \left( 1 - \frac{v_p}{v_0} \right) \dots \dots \dots (15)$$

Only in the region of very low velocities of  $v_p < 5 \cdot 10^{-2}$  cm/s the capillary pressure will rise to greater values. In Fig. 6 the static capillary pressure belonging to a wet saturation degree  $S = 0.2$  is shown. The important effect

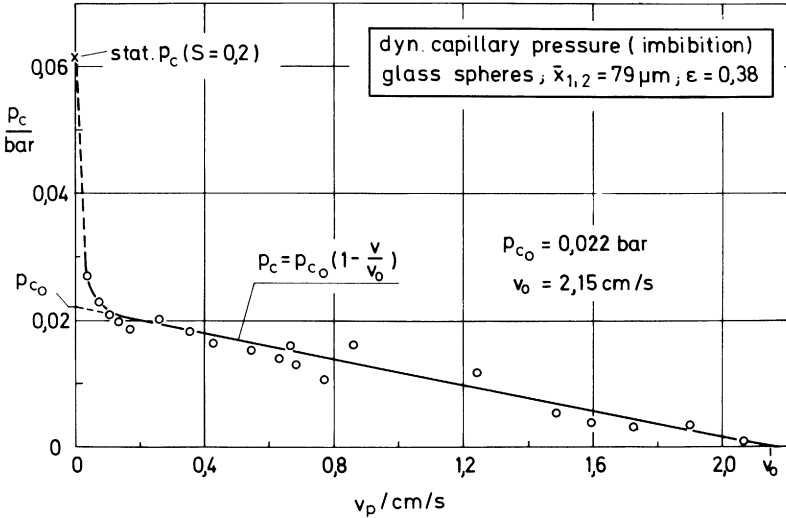


Fig. 6—Dynamic capillary pressure  $p_c$  as a function of the mean liquid velocity  $v_p$  in the pores of a packing of glass spheres

of capillary pressure dynamics on imbibition is made clear by the difference between static and dynamic capillary pressure.

Values  $p_{c0}$  and  $v_0$  are fitting parameters to be determined by an experiment. They characterise the dynamic capillary pressure and are functions of the combination of substances used. For  $v_0 \rightarrow \infty$  the dynamic capillary pressure is equal to the static capillary pressure. On the basis of the present state of knowledge a generally valid statement on dynamic capillary pressure cannot be formulated. The experiments known to us, and theoretical considerations, indicate that the dynamic capillary pressure depends to a lesser extent on the Capillary Number  $Ca = \eta v_p / \gamma$  ( $\gamma =$  surface tension of the liquid), than on the absolute velocity  $v_p$ . As shown below, values of  $p_{c0}$  and  $v_0$  can easily be determined by a simple experiment. With the aid of (15), equation (7) can be integrated. For  $p_{c0} \gg g\rho h \cos \alpha$  we obtain after some calculation

$$\frac{t}{V} = mV + b, \quad \dots \dots \dots (16)$$

with

$$b = \frac{\eta R_f}{A p_{c0}} + \frac{1}{A \epsilon v_0} \quad \dots \dots \dots (17)$$

and

$$m = \frac{\eta}{2 B p_{c0} \epsilon A^2} \left(1 + \frac{B b A g \rho \cos \alpha}{\eta}\right), \quad \dots \dots \dots (18)$$

It therefore suffices to determine the entire liquid volume  $V$  penetrated into the porous system as a function of the penetration time  $t$ , and to plot  $t/V$  over  $V$  in a linear coordinate system. If a straight line is obtained, statement (15) is confirmed. From the slope  $m$  of the straight line and from the ordinate intercept  $b$ , we obtain the values for

$$p_{c0} = \frac{\eta}{2mB\epsilon A^2} \left( 1 + \frac{BbAg\rho \cos \alpha}{\eta} \right) \quad (19)$$

and

$$v_0 = \frac{1 + BbAg\rho \cos \alpha/\eta}{\epsilon A [b(1 + BbAg\rho \cos \alpha/\eta) - 2mB\epsilon R_f A]}, \quad (20)$$

which characterise the dynamic capillary pressure.

**6—Experiments**

FOR the experiments a simple piece of equipment was used, similar to that described several times in the literature.<sup>(21)</sup> Fig. 7 shows the set-up as a skeleton sketch. The powder to be tested is filled into a cylindrical vessel of 30 mm diameter, closed by a thin, porous plate, or filter medium. The vessel is suspended in a load cell and can be lowered until its lower bottom touches the surface of the liquid. From that moment on, the capillary pressure sucks the liquid into the powder. The weight of the test equipment is recorded throughout the test. At the moment  $t = 0$ , the meniscus of the liquid jumps against a sharp-edged ring and so reduces the weight of the equipment. The weight then rises according to the sucked-in liquid volume  $B$  which can, by equation (10), be converted to the penetration height  $h$  if a sharp

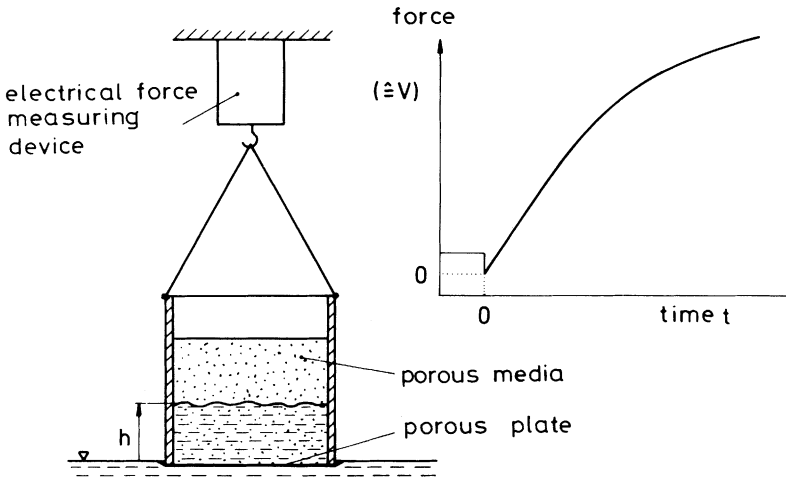
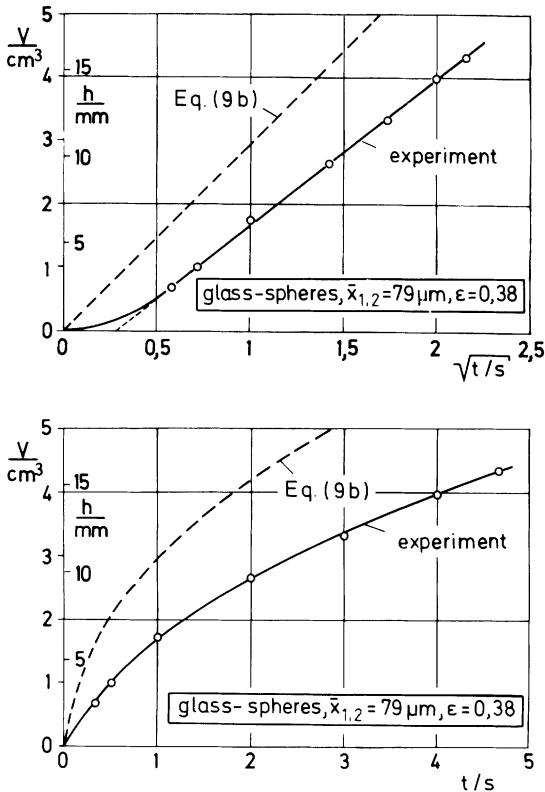


Fig. 7—Schematic of the equipment for measuring the capillary rise

liquid front develops. As can be seen, the equipment is extremely simple and, consequently, also suitable for use in production plants. For the experiments described here a digitalising and storage unit was attached to the equipment to permit the results to be fed into a computer for further evaluation.

The most important element of the apparatus is the porous plate which must be carefully chosen. It must be so designed that a satisfactory hydraulic connection between plate and powder is guaranteed. The hydraulic connection was made by a thin intermediate layer of fine powder between the filter medium and the powder to be tested. Before the powder was filled in, the porous plate and the intermediate layer were saturated with liquid. Both porous materials must be so fine that the powder cannot suck any liquid from the test material before the test is started.

Without a satisfactory hydraulic connection, the results are unreliable. During the preset time, deviations of the penetrated volume by more than a factor of 10



**Fig. 8**—Penetrated liquid volume  $V$  as a function of the penetration time  $t$  or  $\sqrt{t}$  for a packing of glass spheres

were observed. Previous publications have paid no attention to this phenomenon. From a comparison with our own results we have to fear that many results obtained with powders and published in the literature cannot be used. Only the tests of consolidated packings needing no porous plate have shown correct values.

As an example, Fig. 8 shows the liquid volume  $V$  against the penetration time  $t$  and the root of  $t$ . We selected a simple system, a packing of glass spheres of a mean diameter  $\bar{x}_{1,2} = 79 \mu\text{m}$ , and a porosity of  $\epsilon = 0.38$ . In this case it was possible to work without a porous plate, as the packing was slightly sintered.

For comparison, the curves computed according to equation (9b) are drawn into Fig. 8 as broken lines. The computation is based on the permeability  $B = 5.8 \cdot 10^{-8} \text{ cm}^2$  and the mean static capillary pressure  $p_c = 0.04 \text{ bar}$  with  $S = 0.5$  (cf. Fig. 3). The considerable discrepancy between measured values and the 'theory' makes it clear that it is not the static capillary pressure shown in Fig. 3, but the dynamic capillary pressure that matters.

Fig. 9 shows a plot of similar results according to equation (16). Here measurements were made both with and without a porous plate. The evaluation of this test is shown in Fig. 9 for a negligible gravimetric effect (or  $\alpha = 90^\circ$ ). The values for  $p_{c0}$  and  $v_0$  agree well with the values measured independently, as shown in Fig. 6. The plot in Fig. 9, therefore, characterises in a convincing manner the capillary rise.

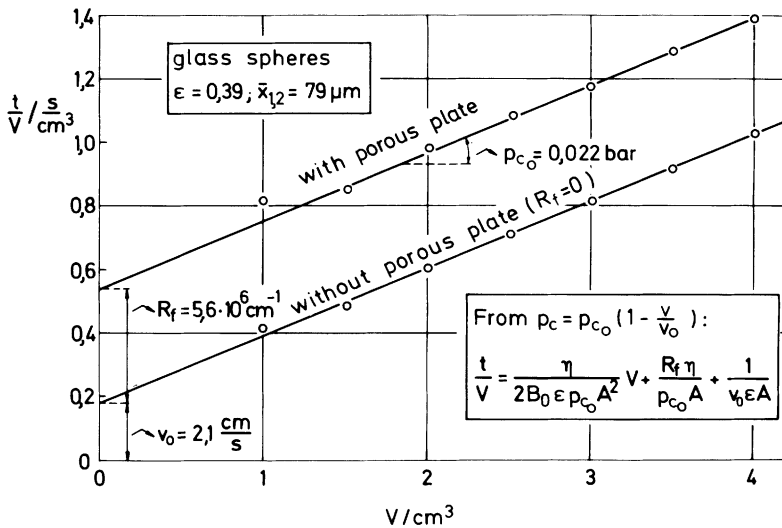


Fig. 9—Plot of the test results from Fig. 8 according to equation (16)

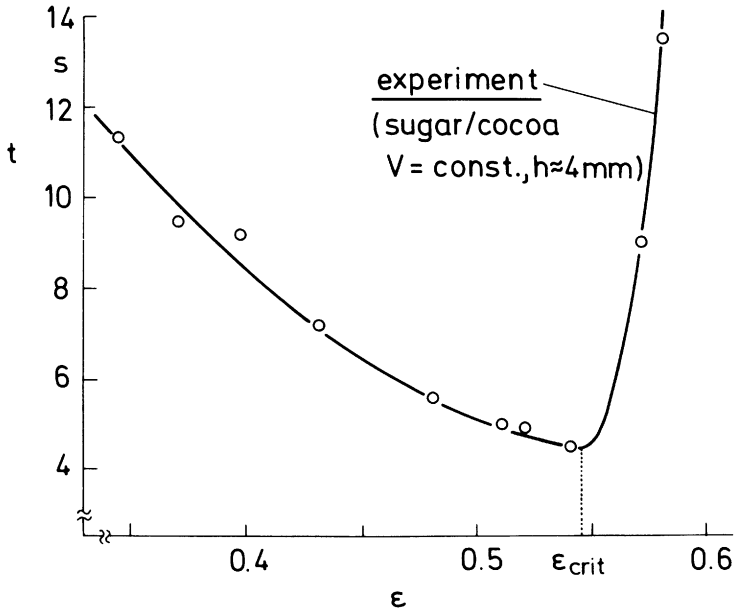


Fig. 10—Determination of the critical porosity  $\epsilon_{crit}$  (explained in the paper)

A great number of different powders were tested in the way described here. All the tests yielded a straight line, if  $t/V$  is plotted over  $V$  as illustrated in Fig. 9. Thus the simple formulation (15) was invariably valid for the samples used. In our presentation we are going to report fully on the experimental results. We are also going to deal with some peculiar features. One of these features is the so-called critical porosity as explained by Fig. 10. It is related to the capillary rise of water inside a packing consisting of sugar and cocoa particles. We plotted the time  $t$  required for the penetration of a constant liquid volume  $V$ , against the porosity  $\epsilon$ . The time of penetration decreases until the critical porosity  $\epsilon_{crit}$  is reached, according to the theory. For  $\epsilon > \epsilon_{crit}$ , the penetration time rises steeply. Packings having the value  $\epsilon > \epsilon_{crit}$  can only be wetted with difficulty. This behaviour can be explained by the arrangement of the packing.<sup>(20)</sup>

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# Transcription of Discussion

## *Discussion*

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*Dr M. B. Lyne* Could I ask Prof. Schubert how he measured the capillary pressure, without implying a model?

*Schubert* We had a tube with a diaphragm at one end. This diaphragm was permeable to the liquid but not the gas. Above the diaphragm the packing was established and we measured the difference in pressure between points above the top level of the packing and below the diaphragm. This method is well-known but our apparatus works automatically so that we can reach equilibrium quickly. We can reduce the time to produce a curve from one week to one day, but of course this depends on the pore size or particle size distribution of the material used.

*Prof. J. T. G. Overbeek* Prof. Schubert, you mentioned that dynamic capillary pressure was due to a contact angle that was not the equilibrium contact angle. Could you give us an idea of the change in contact angle that you require to explain the dynamic capillary pressure?

*Schubert* That is a most difficult question to answer. Nobody has so far succeeded in measuring the contact angle in such a porous system. We know the dependence of the capillary pressure on the dynamic contact angle in simple sphere packings only. In this case the contact angle can be determined by comparing it with that against a plane of the same surface properties. The relationship between the capillary pressure and the contact angle is considerably more complicated than the well-known cos-function suggests. For contact angles between 0 and approximately 50–60° the capillary pressure decreases until it reaches zero.

*Mr J. R. Parker* At the 1973 Fundamental Research Symposium I made reference to a paper by Inverarity who measured dynamic contact angles in

*Under the chairmanship of Prof. M. Judt*

### *Discussion*

relation to glass fibres by drawing them continuously downward into a pool of liquid. I wonder if you have objections to the technique he used ?

*Schubert* There are two methods of measuring the dynamic contact angle for systems with very well defined geometry. In the first method you move a rod immersed in the fluid, this is the best method provided you have a well defined cross-section of the tube. The second method is that in which you apply a drop of a fluid to a plate and this drop is measured for its contact angle as a function of the time. Several people have tried to correlate these two methods but not very successfully. An equivalent to the first method is the determination of the dynamic contact angle by moving a liquid meniscus in a cylindrical tube.