Pergamon

Int. J. Heat Mass Transfer. Vol. 39, No. 9, pp. 1925–1934, 1996

0017–9310/96 $15.00+0.00

0017–9310(95)00265–0

Isothermal liquid transport in porous media: capillary hysteresis effects

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(*Received* 24 *March* 1995 *and in final form* 28 *June* 1995)

Abstract—The isothermal transport of liquids in partially saturated packed beds of glass spheres has been examined. Experimental results and numerical calculations show that in the case of hysteresis-affected capillary pressures a flux equation based on the gradient of the local capillary pressure has to be used. Measured capillary pressures for different transport processes, initial states and liquids are presented. The transport rate is controlled by the effective permeability, which depends on the mobility ratio of gas and liquid flow. At high liquid contents the increasing gas flow resistance causes a decrease of the total transport rate.

INTRODUCTION

A multitude of engineering efforts include application and treatment of moist porous media. Drying of wet products, decontamination of polluted soils or irrigation of plants are only a few examples. They include the transport of a liquid in an unsaturated porous material. At low transport rates two different types of flux equations are used in the literature. One describes the transport of the liquid similar to diffusion while the other states a flow process. It has been recognized very early [1], that differences in the local capillary pressure are the physical reason for liquid flow in an unsaturated porous medium. As shown in Fig. 1, the capillary pressure usually depends on the initial state and preceding transport process. This hysteresis and the complications caused by it have been subject of experimental and theoretical studies [2–4]. Nevertheless, in chemical engineering and hydrology it is common to ignore hysteresis and to assume a unique relation between capillary pressure, permeability and liquid content [5–7]. This simplification reduces the mathematical description of the transport process to a nonlinear parabolic differential equation with strongly varying, but continuous kinetic coefficients, while inclusion of hysteresis can originate discontinuous coefficients. A second reason for neglecting hysteresis in most studies on transport processes in unsaturated porous media is that the measurement of hysteresis affected capillary pressures and permeabilities is time consuming and even for customary model systems like packed beds of glass spheres only a few data of both quantities are available (e.g. [8]). To elaborate the need of taking hysteresis into account and to provide a basis for estimation of the controlling factors the transport of a liquid in packed beds of glass spheres has been studied.

MATHEMATICAL FORMULATION

The one-dimensional isothermal displacement of a liquid or gas in an unsaturated, homogeneous, isotropic and rigid porous body is examined. Gas and liquid are assumed to be incompressible and immiscible Newtonian fluids. Neither gas nor liquid dissolve in the solid. Any chemical reaction or osmotic effect is excluded from consideration. Surface transport or diffusion of vapour are ignored and the influence of gravity is neglected.

Using the degree of saturation

$$S_l = \frac{V_l}{\varepsilon V_t} \tag{1}$$

as a measure of liquid content and referring to the superficial velocity

$$u_f = \frac{\dot{V}_f}{A_t} \tag{2}$$

the mass balances for gas and liquid are:

$$-\varepsilon \frac{\partial S_l}{\partial t} = -\frac{\partial u_g}{\partial x} \tag{3}$$

$$\varepsilon \frac{\partial S_l}{\partial t} = -\frac{\partial u_l}{\partial x}. \tag{4}$$

In general, the velocity of each fluid depends on the pressures in all phases. If attention is focused on processes with very low transport rates characterized by a capillary number of

$$Ca = \frac{u_l \eta_l}{\sigma_{gl}} \ll 1 \tag{5}$$

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NOMENCLATURE

a	parameter of the van Genuchten-equation (24)
A	area
b	capacity, defined by equation (18)
C	parameter of the van Genuchten-equation (24)
Ca	Capillary number, equation (5)
$\bar{d}$	mean particle diameter
D	moisture diffusivity, defined by equation (15)
F	force
g	acceleration of gravity
k	permeability of a single fluid
K	permeability
n	exponent
m	mobility ratio
M	mechanical moment
P	pressure
R	mean curvature of gas–liquid interface
S	degree of saturation
t	time
u	superficial velocity
V	volume
x	space coordinate.

Greek symbols

α	parameter of the van Genuchten-equation (24)
ε	porosity
η	viscosity
κ	liquid diffusion coefficient, defined by equation (17)
σ	surface tension.

Subscripts

0	initial
c	capillary
d	drying
eff	effective
f	fluid
g	gas
ir	irreducible
l	liquid
r	relative
s	saturated
t	total
w	wetting.

Abbreviations

PWC	primary wetting curve
MWC	main wetting curve
SWC	secondary wetting curve
PDC	primary drying curve
MDC	main drying curve
SDC	secondary drying curve.

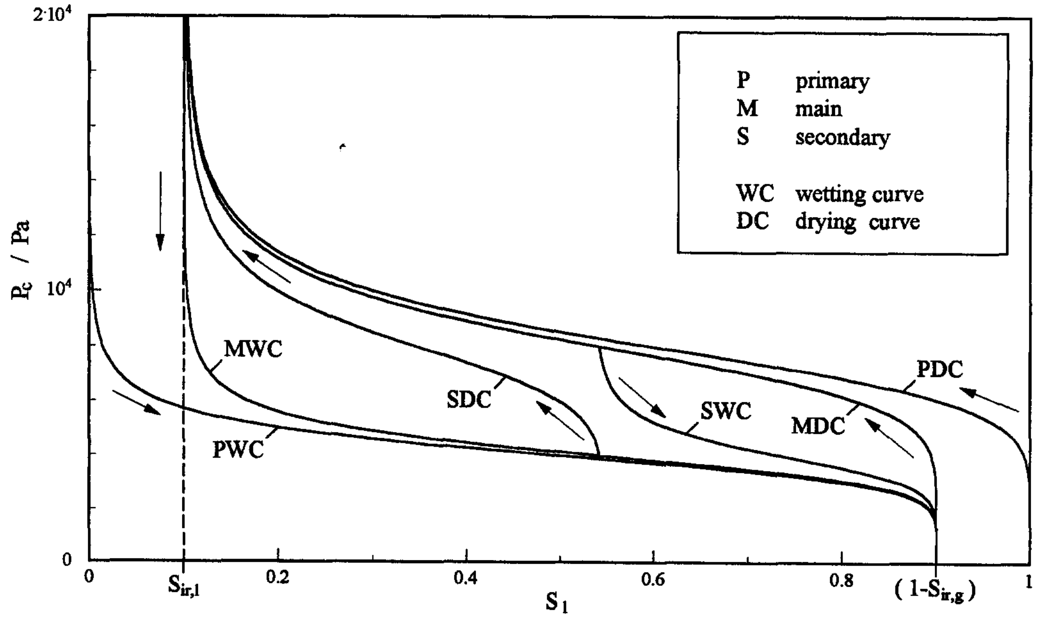

Fig. 1. Typical capillary pressure curves for sand–water.

capillary forces dominate the flow of the liquid. Therefore, the momentum transfer across the gas–liquid interface and also inertial effects can be neglected. In this case it is possible to use linear and uncoupled extensions of Darcy's law for fluid flow through porous media [9, 10]:

$$u_g = -\frac{k_g}{\eta_g}\frac{\partial P_g}{\partial x} \tag{6}$$

$$u_l = -\frac{k_l}{\eta_l}\frac{\partial P_l}{\partial x}. \tag{7}$$

Equations (6) and (7) state, that the superficial velocity u_f of each fluid is simply the product of the pressure gradient in the fluid and the ratio of permeability k_f and dynamic viscosity η_f of the fluid under consideration. Commonly, the permeability k_f is expressed as the product of the permeability k_{sf} of the porous medium saturated with one fluid and the relative permeability k_{rf}, which describes the dependence of the permeability on the degree of saturation:

$$u_g = -\frac{k_{sg}k_{rg}}{\eta_g}\frac{\partial P_g}{\partial x} \tag{8}$$

$$u_l = -\frac{k_{sl}k_{rl}}{\eta_l}\frac{\partial P_l}{\partial x}. \tag{9}$$

The pressures in the fluids are coupled via the capillary pressure. Excluding sorption and restricting on small pressure differences caused by the curvature of the gas–liquid interface [10], the capillary pressure is simply

$$P_c = P_g - P_l. \tag{10}$$

Combination and rearrangement of equations (3), (4) and (8)–(10) using the method of Buckley and Leverett [11] leads to the transport equation

$$\varepsilon\frac{\partial S_l}{\partial t} + u_t\frac{\partial}{\partial x}\left(\frac{1}{1+m}\right) + \frac{\partial}{\partial x}\left(\frac{K_{eff}}{\eta_l}\frac{\partial P_c}{\partial x}\right) = 0. \tag{11}$$

In equation (11) the effective permeability

$$K_{eff} = \frac{m}{1+m}k_{sl}k_{rl} \tag{12}$$

depends on the mobility ratio

$$m = \frac{\eta_l k_{sg} k_{rg}}{\eta_g k_{sl} k_{rl}} \tag{13}$$

and therefore describes the permeability of the porous medium for a imbibition or drainage process including the flow resistance for gas flow. The convective term in equation (11) is a measure of change in saturation caused by the total superficial velocity

$$u_t = u_g + u_l \tag{14}$$

and the local change in the ratio of the permeabilities.

To get a resolvable equation either $\partial S_l/\partial t$ or $\partial P_c/\partial x$ has to be replaced in equation (11). If capillary hysteresis is neglected or one concentrates on processes with the same hydraulic history and local transport direction throughout the porous medium, it is possible to substitute $\partial S/\partial x$ for $\partial P_c/\partial x$ and to use an effective moisture diffusivity

$$D_{eff} = -\frac{K_{eff}}{\eta_l}\frac{\partial P_c}{\partial S} \tag{15}$$

to describe the fluid flow. If also the flow resistance for gas flow is neglected, the convective term disappears and the transport equation (11) is equal to the equation of Richards [12]. In this case, the effective moisture diffusivity defined by equation (15) reduces to the moisture diffusivity

$$D = -\frac{k_{sl}k_{rl}}{\eta_l}\frac{\partial P_c}{\partial S} \tag{16}$$

used in hydrology for description of infiltration processes [13] or the liquid diffusion coefficient

$$\kappa = \frac{D}{\varepsilon} \tag{17}$$

introduced by Krischer [7] in drying technology. Both coefficients can be estimated using a combination of approximation functions for permeability and capillary pressure [14] or by methods based on a geometric interpretation of measured capillary pressures [7].

Hysteresis in the relation of P_c and S_l leads to problems in performing the differentiation of the third term in equation (11). Therefore, the substitution of $\partial P_c/\partial t$ for $\partial S/\partial t$ and the definition

$$b = -\varepsilon\frac{\partial S_l}{\partial P_c} \tag{18}$$

is adopted. Using this capacity term one gets the transport equation:

$$b\frac{\partial P_c}{\partial t} = u_t\frac{\partial}{\partial x}\left(\frac{1}{1+m}\right) + \frac{\partial}{\partial x}\left(\frac{K_{eff}}{\eta_l}\frac{\partial P_c}{\partial x}\right). \tag{19}$$

With the condition

$$u_t|_{\text{boundary}} = 0 \tag{20}$$

at an impermeable boundary and using equations (3), (4) and (14) it follows

$$u_{t(x)} = 0 \tag{21}$$

throughout the porous body. Consequently the convective term in equation (19) disappears. If attention is focused on the effect of hysteresis and the influence of the mobility ratio on the transport of a liquid, it is hence convenient to investigate redistribution processes in closed systems.

EXPERIMENTAL SET-UP

The principle of the apparatus build to observe the transport process is depicted in Fig. 2. During the flow of a liquid in a porous body the centre of gravity moves. This causes a change in the mechanical

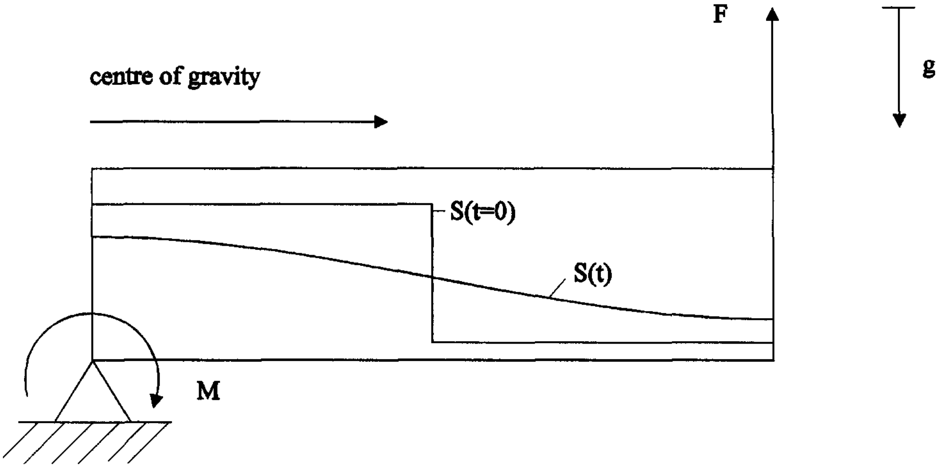

Fig. 2. Principle of the moment method.

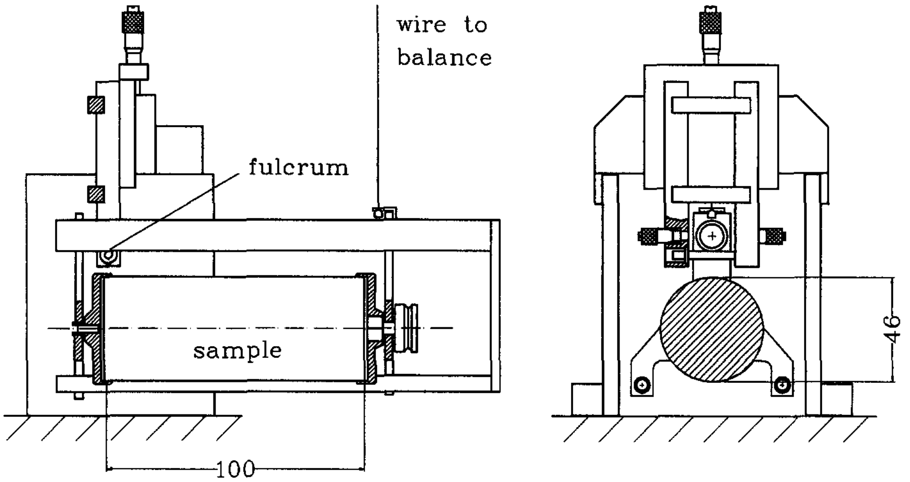

Fig. 3. Apparatus for the moment method.

moment at the fulcrum. It is measured as change of the leverage balancing the moment. Figure 3 shows a simplified scheme of the apparatus actually used. Its main features are displaceable fulcrum, a revolving sample attachment and the use of different precision adjusting-screws to insure an accurate horizontal alignment. The leverage is measured with a balance, enabling a resolution of 10^{-6} N m at the employed sample length of 0.1 m.

Capillary pressures have been measured with the suction apparatus suggested by Haines [15]. The alteration rate of the level in the burette of the device can also be used to determine effective permeabilities. Details of the apparatus and the procedure to evaluate the experimental data are described in [16].

The packed beds were made of glass spheres with normal size distribution. Their main properties are given in Table 1. Distilled water or commercially available liquids of analytical grade have been used.

EXPERIMENTAL RESULTS

The effect of capillary pressure hysteresis on the final liquid distribution is shown in Fig. 4. The redistribution process in a sample with initially step-like loading is monitored by measuring the change of moment. To ensure complete equalization of driving forces, the experiment lasted 60 h although almost the whole transport process is finished during the first 20 h. At the end of the experiment the sample is cut into pieces of 10 mm width and their liquid content is measured. Only a small amount of the liquid in the

Table 1. Properties of the particles and packed beds

$\bar{d}$ [μm]	σ_{STD} [μm]	ε	k_{sl} [m^2]
127	9.2	0.37	9.0×10^{-12}
321	21.7	0.37	1.1×10^{-10}

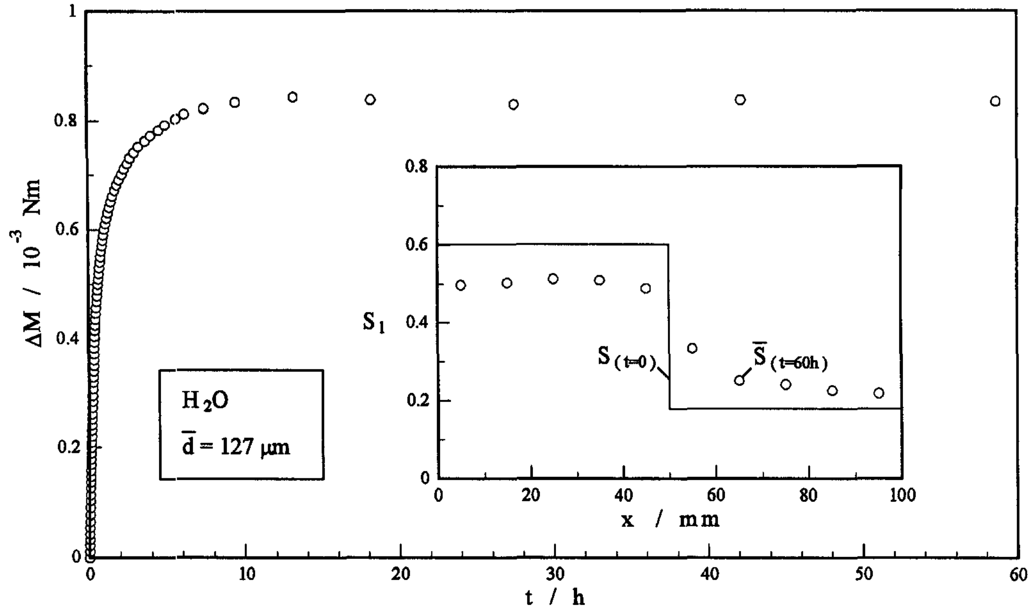

Fig. 4. Redistribution experiment, change of moment and final liquid distribution.

part with initially high degree of saturation moved to the lower loaded part. This result is easy to understand, if one considers a balancing of capillary pressures as driving force for the liquid movement. In Fig. 5, a qualitative explanation is sketched using a detail of the capillary pressure curves shown in Fig. 1. It is assumed that the two parts of the sample have been wetted along the same main wetting curve (MWC) to their different initial degrees of saturation S_{01} and S_{02}. During the redistribution process the part with the lower degree of saturation S_{01} and higher capillary pressure P_{c01} is wetted along MWC while the other part is drained along a secondary drying curve (SDC). At equal capillary pressures the difference in MWC and SDC causes the remaining difference in the final liquid content of the two parts.

To describe this hysteresis effect on the rate of liquid exchange and final liquid distribution quantitatively, effective permeabilities and capillary pressures must be known. A complete set of capillary pressure curves representing a sequence of imbibition and drainage processes of an initially dry and a saturated packed bed is shown in Fig. 6. Supplementary also capillary pressures of a mechanical mixture compressed to the

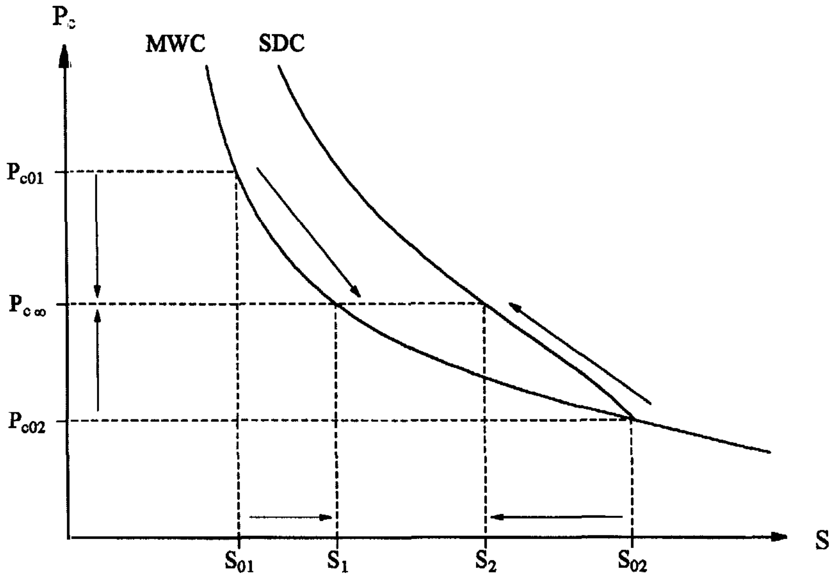

Fig. 5. Effect of capillary pressure hysteresis on the final liquid distribution.

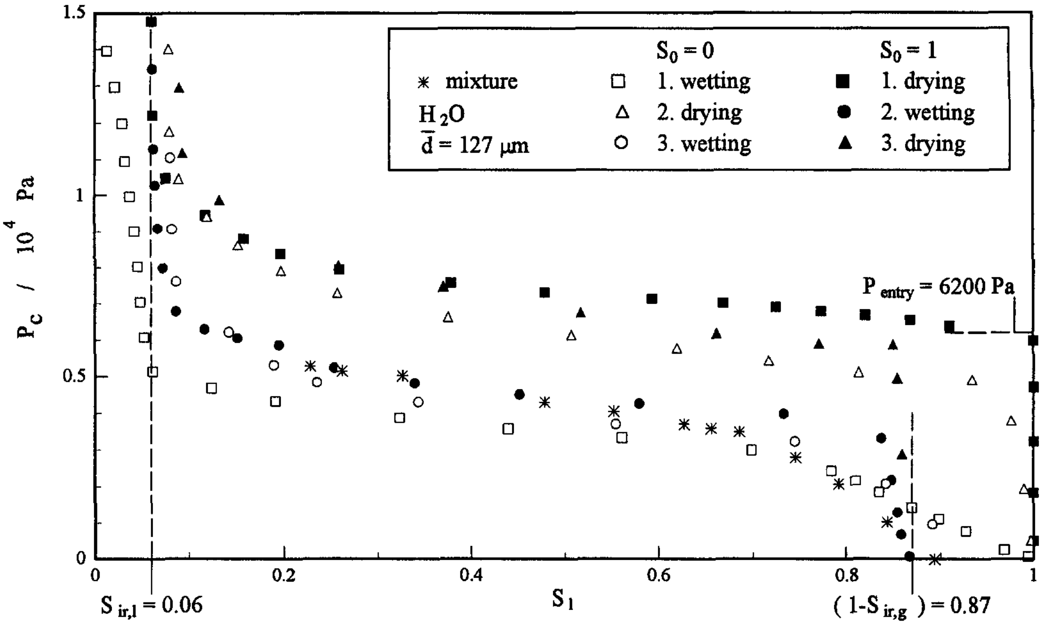

Fig. 6. Capillary pressure curves for a packed bed of glass spheres and water.

same porosity are shown. The values match the imbibition curves probably because the movement of the gas–liquid interface during compression is the same as in a wetting process. In Fig. 7 capillary pressures of the same packed bed but different liquids and of a second packed bed are shown. The results are given in a normalized form using the J-function

$$J = \frac{P_c\sqrt{(k_{sl}/\varepsilon)}}{\sigma_{gl}} \tag{22}$$

suggested by Leverett [17]. Yet unexplained deviations at very low or high degrees of saturation the curves are independent of the viscosity and proportional to the surface tension of the liquid. This result might be expected from the Laplace-equation

$$P_c = \frac{\sigma_{gl}}{R_{(S)}} \tag{23}$$

where $R_{(S)}$ is the mean curvature of the gas–liquid

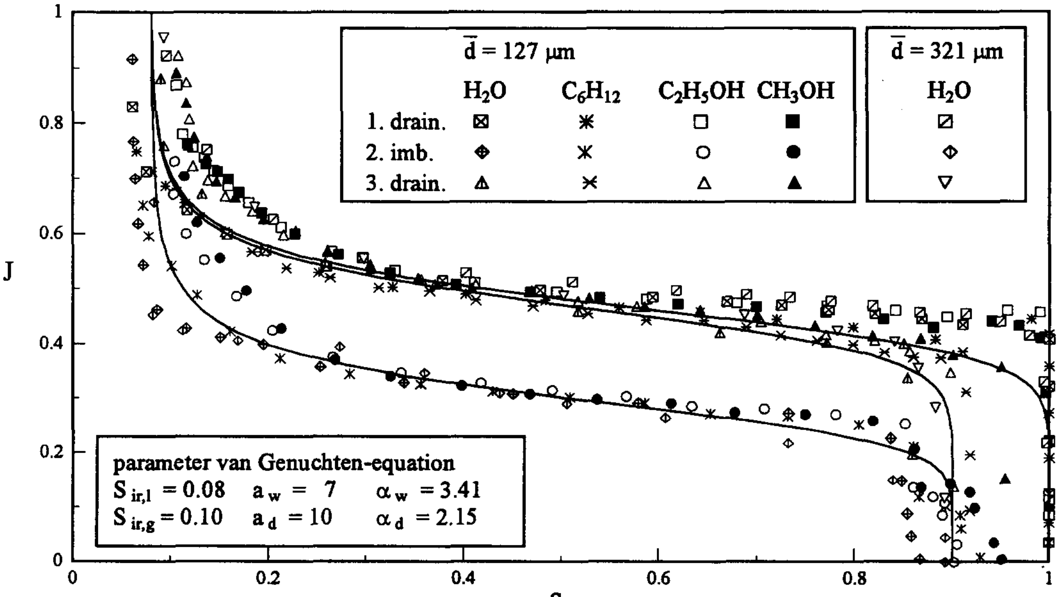

Fig. 7. Normalized capillary pressure curves of different packed beds and liquids.

interface. Nevertheless it is not trivial, because there is evidence that the liquid distribution and thus the capillary pressure depends in general on imbibition or drainage rate and on the mobility ratio [18].

For most applications the experimental results can be represented sufficiently accurate by the empirical equation of van Genuchten [19]

$$\frac{S_l - C_l}{1 - C_g - C_l} = [1 + (\alpha J)^a]^{(1-a)/a} \tag{24}$$

with parameters depending in the initial state and transport direction (see Appendix). It has to be emphasized, that the equation of van Genuchten is only suitable to packed beds of particles with narrow size distribution. Bidispers systems can show capillary pressure curves not describable with the model of van Genuchten [16]. In this case, a more general method, e.g. the domain theory [20], should be applied.

Figure 8 shows some measured effective permeabilities for the same system as in Fig. 6. The most striking result is the existence of a maximum. An explanation is given by equations (12) and (13), considering that at increasing liquid contents the permeability for liquid flow increases but the permeability for gas flow decreases. This results in the observed maximum of the effective permeability at a liquid content determined by the ratio of fluid viscosities and the degrees of irreducible saturations.

Although deviations between the effective permeabilities of the different processes are obvious, no systematic dependence on the transport direction or initial state can be deduced. Therefore, the effective permeability is assumed to be a unique function of the liquid content. In Fig. 8 the dashed lines represent the individual permeabilities for gas and liquid flow according to the customary power law

$$k_{rf} = \left(\frac{S_f - S_{ir,f}}{1 - S_{ir,f}}\right)^n. \tag{25}$$

For convenience the same exponent $n = 4$ and equal permeabilities for gas and liquid flow in a dry and a saturated packed bed have been used. Combination of these curves according to equations (12) and (13) leads to an acceptable description of the measured effective permeability.

If only the permeability of the saturated packed bed is known, different authors have suggested to calculate the relative permeability from the capillary pressure curves. Mualem [21] has reviewed these methods. Following his recommendations, use of the van Genuchten-equation (24) leads to relations compiled in the Appendix and shown in Fig. 8 as dotted lines. They give only coarse estimations of the measured effective permeability. Regarding this results and the scattering of experimental data, the use of more elaborated models seems to be inappropriate.

CALCULATION RESULTS

The transport equation (19) has been solved in its simplified form for closed systems taking advantage of equation (21). Numerical calculations were performed using the finite volume method [22] and a fully implicit scheme. In Fig. 9 the results for a redistribution process in a closed system with a step-like initial loading are shown. Capillary pressures and permeabilities have been calculated by the van Genuchten-equation

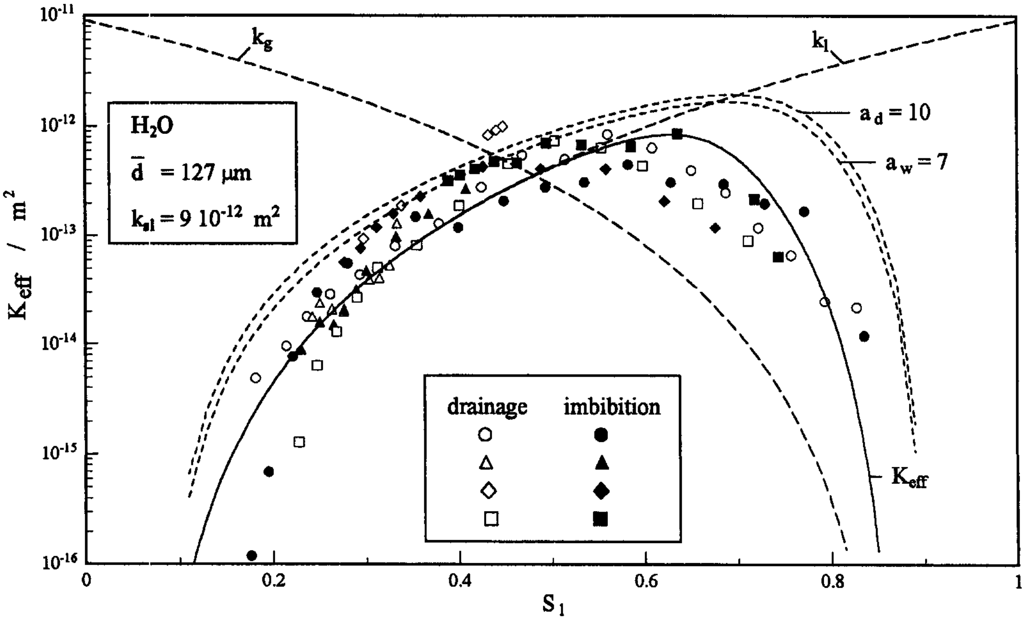

Fig. 8. Effective permeability for a packed bed of glass spheres and water.

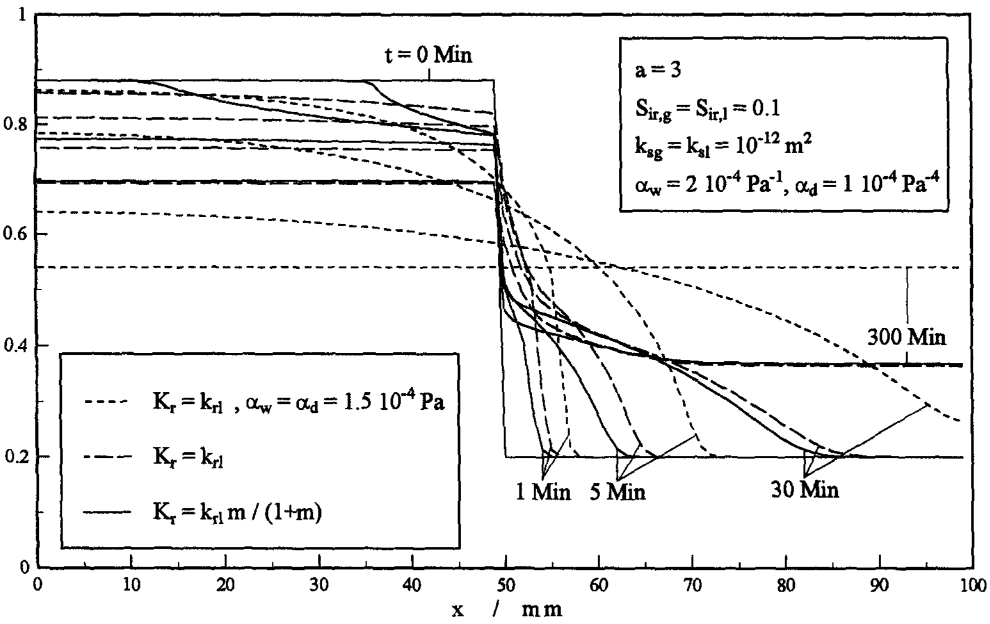

Fig. 9. Calculation results for a redistribution process in a closed system.

(24). The dotted lines represent the results for a system without hysteresis. The dashed lines show results for a calculation taking hysteresis into account but ignoring the gas flow resistance while the full lines represent a calculation including both factors. Figure 9 clearly demonstrates the incomplete exchange of moisture caused by a hysteresis of the capillary pressure. It also indicates the influence of the gas flow resistance resulting in a decrease of the total transport rate in areas of high liquid content.

A comparison between calculation and experiment is shown in Fig. 10. A preloaded sample has been wetted almost instantaneously at one of its surfaces (position $x = 0$) with a limited amount of water. The movement of the centre of gravity during the redistribution process has been monitored by measuring the change of moment and the distribution of the liquid at the end of the experiment has been determined.

The calculation of the capillary pressures is based on the van Genuchten-equation (24) with parameters as listed in Fig. 7 and Table 1. Permeabilities were

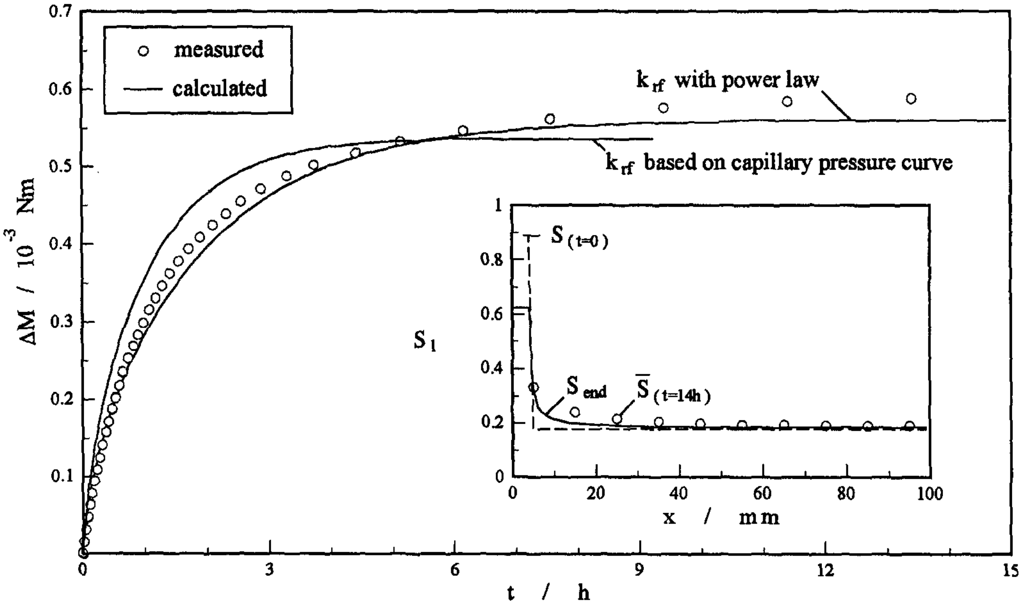

Fig. 10. Comparison between experiment and calculation for a redistribution process.

calculated either by the power law equation (25) or using the estimation based on the capillary pressure curves. Both methods lead to almost the same final distribution of the liquid content with deviations too small to be shown in Fig. 10. As one might expect from Fig. 8, the deviation between experimental values of the change of moment and the calculation based on equation (25) is slightly smaller than the difference between experiment and the calculation based on an estimation of the permeabilities from the capillary pressure curves.

Experiment and calculation show that the infiltration of the liquid is limited by the hysteresis of the capillary pressure. The movement of the liquid is controlled by a drainage capillary pressure at the boundary $x = 0$ while the uptake of the liquid by the inner parts of the sample is controlled by an imbibition capillary pressure. As depicted in Fig. 6, the difference in the relation of P_c and S for different transport directions causes the restricted penetration of the liquid into the packed bed.

CONCLUSIONS

To describe the internal transport of liquids in unsaturated porous media, two different flux equations have been proposed in the literature. One is based on the degree of saturation of the material, the other on the capillary pressure. Experimental results on packed beds of glass spheres and numerical calculations show that in case of hysteresis affected capillary pressures the transport of the liquid has to be described by flux equations based on gradients of the local capillary pressure, which reflect the complete hydraulic history of the system.

The capillary pressures measured for different liquids and packed beds of glass spheres with narrow size distribution can be represented in a normalized form using the J-function of Leverett by the parametric model of van Genuchten.

The rate of liquid transport is controlled by the effective permeability, which depends on the viscosity of the fluids and the permeability for gas and liquid flow. The permeabilities for the individual fluid flow can be represented by the customary power law equations or estimated coarsely using the capillary pressure curves. In areas of high liquid content the imbibition and drainage rate are influenced by the transport resistance for gas flow, resulting in lower total transport rates.

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APPENDIX

Capillary pressures have been calculated by the van Genuchten-equation [19]

$$\frac{S_l - C_l}{1 - C_g - C_l} = [1 + (\alpha J)^a]^{(1-a)/a} \tag{A1}$$

with different parameters α_w during wetting and α_d during drying. The parameters C_g and C_l depend on the transport process and are listed in Table 2. If the capillary pressure changes along a secondary curve, the parameters C_g and C_l depend on the degree of saturation S_0 and capillary pressure P_{c0} at the inversion point of the former transport direction:

Table 2. Parameters of the van Genuchten-equation

	PWC	MWC	SWC	PDC	MDC	SDC
C_l	0	$S_{\mathrm{ir,l}}$	equation (A2)	$S_{\mathrm{ir,l}}$	$S_{\mathrm{ir,l}}$	$S_{\mathrm{ir,l}}$
C_g	$S_{\mathrm{ir,g}}$	$S_{\mathrm{ir,g}}$	$S_{\mathrm{ir,g}}$	0	$S_{\mathrm{ir,g}}$	equation (A3)

$$C_l \frac{1-S_{\mathrm{ir,g}}-S_0[1+(\alpha_{\mathrm{w}}P_{\mathrm{c0}})^a]^{(a-a)/a}}{1-[1+(\alpha_{\mathrm{w}}P_{\mathrm{c0}})^a]^{(a-1)/a}} \qquad \text{(A2)}$$

$$C_g = 1-S_{\mathrm{ir,l}}-(S_0-S_{\mathrm{ir,l}})[1+(\alpha_{\mathrm{d}}P_{\mathrm{c0}})^a]^{(a-a)/a}. \qquad \text{(A3)}$$

Using the van Genuchten-equation (A1) the capacity term is

$$b = -\varepsilon\frac{\partial S}{\partial P_{\mathrm{c}}} = \varepsilon(1-C_g-C_l)\frac{\alpha(a-1)(\alpha P_{\mathrm{c}})^{a-1}}{[1+(\alpha P_{\mathrm{c}})^a]^{(2a-1)/a}}. \qquad \text{(A4)}$$

Permeabilities can be estimated using the effective degree of saturation for each fluid

$$S_{\mathrm{eff}} = \frac{S_{\mathrm{f}}-S_{\mathrm{ir,f}}}{1-S_{\mathrm{ir,f}}} \qquad \text{(A5)}$$

and an equation recommended by Mualem [21]:

$$k_{\mathrm{rf}} = \sqrt{S_{\mathrm{eff}}}\left(\int_0^{S_{\mathrm{eff}}} \frac{\mathrm{d}S_{\mathrm{eff}}}{P_{\mathrm{c}(S_{\mathrm{eff}})}} \bigg/ \int_0^1 \frac{\mathrm{d}S_{\mathrm{eff}}}{P_{\mathrm{c}(S_{\mathrm{eff}})}}\right)^2. \qquad \text{(A6)}$$

With the van Genuchten-equation (A1) it follows (see also [19])

$$k_{\mathrm{rl}} = \sqrt{S_{\mathrm{eff,l}}}\left\{1-\left(1-S_{\mathrm{eff,l}}^{a/(a-1)}\right)^{(a-1)/a}\right\}^2 \qquad \text{(A7)}$$

$$k_{\mathrm{rg}} = \sqrt{S_{\mathrm{eff,g}}}\{[1-(1-S_{\mathrm{eff,g}})^{a/(a-1)}]^{(a-1)/a}\}^2. \qquad \text{(A8)}$$