Nordic Hydrology, 19, 1988, 281-292

On the Physico-Chemical Basis for the Capillary Barrier Effect

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The capillary barrier concept, using fine and coarse soil layers to reduce water infiltration into piles of hazardous wastes, is investigated theoretically. A detailed account of the hydrological and physico-chemical basis for the phenomenon is given. It is established that the capillary barrier will, in practice, only function if the fine layer remains somewhat unsaturated, *i.e.* the upper menisci exist and no ponding over the fine layer occurs. Accordingly, water reaching the fine layer must be transported laterally within this layer. The pressure conditions are dependent on the length of the interface, the slope of the interface, the thickness of the fine layer, the type of soil in the fine layer, the water influx at the surface, and the total volume of water infiltrated during an infiltration event. A simple estimate shows that the capillary barrier concept is feasible only for small heaps with steep interfacial slopes.

Introduction

The possibility of using capillary barriers to reduce water infiltration into dumps of hazardous wastes has been explored for some time. The capillary barrier concept involves the principles of unsaturated flow between soils of different texture. A capillary barrier system is constructed such that a fine-textured soil overlies a coarse-textured soil. Fine soils would tend to have a distribution of small diameter pores while coarse soils would tend to have a distribution of larger diameter pores. Since the capillary forces are higher in a fine-textured soil than in a coarse-textured soil, at a particular value of pressure head, more water would be retained in the former.

At an infiltration event, the smallest pores in the fine soil begin to fill. Capillary flow can, in this concept, only occur to the coarse layer when the pore-size just filled in the fine material matches the smallest pore sizes in the coarse material. If this system was designed in such a manner that the fine-textured material could drain, while maintaining negative pressures at the interface with the coarse material, then the coarse-textured soil would remain essentially dry. This may be the case if the slope of the interface is sufficient to establish a lateral hydraulic gradient.

The principles of unsaturated flow between soils of contrasting textures were, probably, first applied to the problem of waste containment by Corey and Horton (1969), who referred to the phenomenon as the "wick effect". Their laboratory work showed that, provided the fine textured material remained unsaturated, water would not flow into the underlying gravel. They also showed by laboratory experiments that water could be diverted around a gravel lens. Rançon (1972, 1979) also investigated the wick effect and obtained positive results in laboratory and small-scale field experiments.

Frind *et al.* (1976) investigated the capillary barrier concept using numerical simulation procedures. The practical applicability of the concept was tested using a two-dimensional finite element model for transient flow in a saturated-unsaturated porous medium. The simulations showed the effect on the system's performance of the length of the interface, the slope of the interface, the thickness of the fine layer, the type of soil in the fine layer, the water influx at the surface, and the total volume of water infiltrated during a storm event. The pressure head distribution within the wick layer was predicted for various rain events using various combinations of the above variables. Lindsay *et al.* (1979) extended the model to include hysteresis effects in the hydraulic properties of the media. Furthermore, an attempt was made to validate the numerical model against a physical system. At least a qualitative agreement was obtained.

Madsen (1983) experimentally studied capillary barriers as a method for reducing infiltration into piles of flying ash. Critical infiltration rates versus interfacial slope were obtained in meter-scale laboratory experiments.

None of these papers contain a description of the phenomenon at the pore level. The purpose of the present paper is to give a detailed physico-chemical evaluation of the capillary barrier effect.

Theory

Physical Principles According to the Unsaturated Flow Theory

We will start this section by giving an account of the macroscopic description, based on the unsaturated flow theory (Childs 1967; Swartzendruber 1969), of the capillary barrier effect.

The capillary barrier concept involves the principles of unsaturated flow between soils of different texture. If a fine-textured soil overlies a coarse-textured soil and both materials are initially dry, water infiltrating from the surface will not move into the coarse-textured soil until the pressure head in the fine-textured soil approaches zero (*i.e.* the fine-textured soil is at or near saturation). The hydraulic properties which control this phenomenon are the relationships between water content, θ, and pressure head, $\psi(\theta)$, and water content and hydraulic conductivity, $K(\theta)$.

A convenient (but simplified) model of the pore space in a porous medium is an assemblage of capillaries of different diameters. Fine-textured soils would, by definition, tend to have a distribution of small diameter capillaries while coarse-textured soils would tend to have a distribution of larger diameter capillaries. Applying this model, water in an unsaturated soil is retained by capillary forces. For cylindrical pores, assuming zero contact angle, we have the simple "capillary rise" equation

$$\psi = \frac{2\gamma}{\rho g R} \qquad (1)$$

where R is the pore radius corresponding to a certain pressure ψ. Inserting the values of ϱ, g and γ, at 20 °C, one obtains

$$\psi \cong \frac{1.5 \times 10^{-5}}{R} \qquad (2)$$

Since the capillary forces are higher in a fine-textured soil than in a coarse-textured soil, at a particular value of pressure head, more water would be retained in the former.

Fig. 1 shows hypothetical wetting curves for both fine- and coarse-textured soils. In wetting an initially dry soil, the smallest pores will take up water first and successively larger pores will fill until the capillary pressure exerted by the pores is equal to or greater than the pressure in the applied water. At a given pressure head, for example ψ_x in Fig. 1, the water content of the fine-textured material will be much greater than that of the coarse-textured material. Now, at the interface between two soils the pressure head is continuous. Owing to this boundary condition and to the $\psi(\theta)$ relationships for a coarse and a fine soil one may conclude that the water content in the fine soil will be larger than in the coarse one (*i.e.* the water content is discontinuous at the interface).

Fig. 2 shows hypothetical water content versus hydraulic conductivity relationships for the soils of Fig. 1. Although, at saturation, θ_s, the hydraulic conductivity of the coarse-textured soil would be greater than that of the fine-textured material, at pressure head ψ_x, the water content of the coarse material is much less than that of the fine material and consequently the hydraulic conductivity of the fine-textured soil is greater. For a medium of theoretical infinite permeability (*i.e.* infinite pore radius) the capillary pressure curve coincides with the horizontal axis. In such

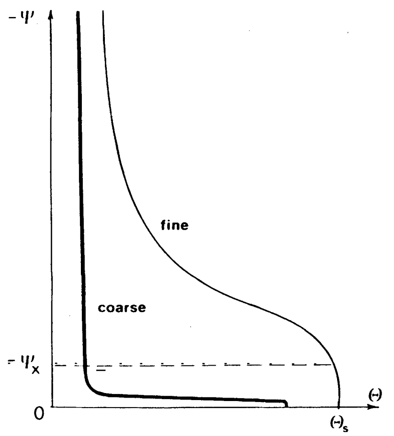

Fig. 1. Hypothetical wetting curves for a fine sand and a coarse gravel.

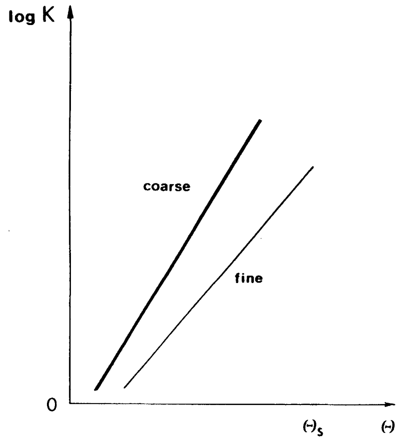

Fig. 2. Hypothetical $K(\theta)$ curves for a fine sand and a coarse gravel.

a case the only intersection point of the two curves is at saturation, which means that water cannot flow from a low permeability soil into a high permeability one until saturation in the low permeability soil is reached.

If a system is constructed such that the fine soil overlies the coarse soil and water is applied at the surface, then the hydraulic conductivity of the fine material (*e.g.* clay) would increase according to Figs. 1-2. The coarse-textured material (*e.g.* gravel), however, would remain relatively dry and with a low hydraulic conductivity until the pressure head at the interface between the clay and gravel approaches zero. As can be seen in Fig. 1, as the pressure head increases towards zero, the fine-textured material becomes saturated and the water content of the coarse-textured soil increases rapidly and, consequently, water would move readily into the coarse-textured material. If this system was designed in such a manner that the fine-textured material could drain, while maintaining negative pressures at the interface with the coarse material, then the coarse-textured soil would remain dry. This may be the case if the slope of the interface is sufficient to establish a lateral hydraulic gradient.

Hydrostatics in a Single Cylindrical Vertical Capillary

In this section we simplify the problem in studying a single cylindrical, vertical capillary with perfect wetting properties (contact angle = 0 degrees). The following discussion is based on the fundamental Young-Laplace equation (see *e.g.* Adamson 1976), which relates the bulk phase pressure difference, Δp, the mean curvature at a certain point on the interface, and the interfacial tension, γ

$$\Delta p = \gamma\left(\frac{1}{r_1} + \frac{1}{r_2}\right) \qquad (3)$$

where r_1 and r_2 are the principal radii of curvature. The phase on the concave side of the surface must have a pressure which is greater than the pressure on the convex side. In a cylindrical tube with comparatively small radius the meniscus becomes spherical and Eq. (3) simplifies to

$$\Delta p \equiv \frac{2\gamma}{r} \tag{4}$$

If the lower tip of the initially air-filled capillary, is placed into contact with water a concave meniscus will form in the tube provided its inner surface is wetted by water. According to Eq. (4) a pressure difference is obtained over the interface where the pressure is lower in the water than in the air, the difference being $2\gamma/R$ (where $R=r$ is the radius of the capillary). Water will now rise in the tube until the weight of the water column balances Δp. Neglecting the weight of air we obtain

$$\Delta p = \rho g h_c \rightarrow \qquad h_c = \frac{2\gamma}{\rho g R} \tag{5}$$

where h_c is the height of capillary rise.

As long as there is an upper meniscus this water column can be mechanically balanced.

Assume now that a vertical capillary is subjected to a certain overpressure. The lower opening is in contact with air while the upper is in contact with a "water reservoir" eliminating the upper meniscus. To force water from the lower tip, a drop of water must first form. The drop development passes through a stage where the radius is approximately equal to the tube radius. According to Eq. (4) a pressure difference of $2\gamma/R$ is generated across the drop surface with the higher pressure in the water phase. Taking into account the "half" drop weight a force balance yields

$$\left(\frac{2\gamma}{R}\right)(\pi R^2) = \pi R^2 h\rho g + \left(\frac{2\pi R^3}{3}\right)\rho g \rightarrow \qquad h = \frac{2\gamma}{\rho g R} - \frac{2R}{3} \equiv h_c - \frac{2R}{3} \tag{6}$$

In fine capillaries ($R < 1$ mm) the term $2R/3$ is negligible and the capillary can, even under saturated conditions at the top, balance a pressure nearly corresponding to the capillary rise. Finally, one may conclude that if the capillary is surrounded by air on both sides it can, in principle, balance a water column corresponding to $2h_c$ (because of the concave upper meniscus and the curvature of the half-drop).

From this section one might be encouraged to conclude that a capillary barrier can function even if the capillary pressure $\psi(\theta) \sim 0$ (*i.e.* saturation) in the fine material at the interface with the underlying coarse material. However, there are at least two effects which make this quite improbable for real porous materials.

They are:

i) Overlapping of adjacent pore openings in the fine material making the development of isolated drops improbable even in the absence of the coarse layer. A water film having negligible curvature and thereby water-holding properties is more likely to develop.
ii) Wetting properties of the underlying coarse material causing water to flow at the contact points with the fine material.

These effects will be the topic of the final subsections in this section.

Single Porous Layer

As a model of the layer of fine-grained material discussed above we shall adopt here a porous disc made out of glass. Such a disc will not normally have a very well-defined porosity unless the manufacturing process is based upon sintering of glass-spheres with a narrow enough size distribution. But even in this case, the geometry of the pores will be rather complex and characterized by narrow slits close to contact points as well as rather wide empty spaces in between the spheres. In recent years, however, studies of glass adsorbents have been reported (Pierotti *et al.* 1986) where the porosity can be modelled by means of a narrow distribution of very thin (< 10 nm) cylindrical capillaries. Although a glass disc with cylindrical capillaries may seem only remotely related to the fine-grained soil material we are primarily considering here, we still believe that the essential features in terms of capillary behaviour of a fine-grained material will be reproduced by a porous glass disc of this particular kind.

Suppose first that the porous glass-disc with chromic-acid-cleaned, perfectly wetting pores is dipped into water followed by removing excess fluid by gentle suction using *e.g.* a filter paper. A water-saturated disc state will then result characterized by concave meniscii on both sides, provided, of course, that the influence of gravity can be neglected, or, that the thickness, d, of the disc is not too large (Fig. 3).

Upon increasing the thickness of the disc while keeping it in a horizontal position there will eventually be a drop-wise flow of water through each pore separately, assuming for the time being that any lateral flow along the lower disc surface is inhibited. This flow would ultimately cease when the upper meniscus in each cylindrical pore is located $\sim 2h_c$ above the lower disc surface.

Thus for discs with thickness $d > 2h_c$ the water content would be less than what corresponds to saturation whereas discs with $d < 2h_c$ would always remain saturated with water (Fig. 4). This reasoning is obviously in line with our discussion in the previous subsection about the hydrostatics in a single cylindrical capillary.

When the top surface of the disc is flooded with water, there will also be a drop-wise flow insofar as the total water level above the lower disc surface exceeds, approximately, the height h_c. We emphasize here that h_c can readily amount to a few dm for capillaries with small enough diameters.

Let us now switch over to a slightly different, but at the same time more realistic, case where we assume that there is no hindrance, whatsoever, for the contact lines

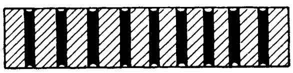

Fig. 3. Porous disc in a water-saturated state.

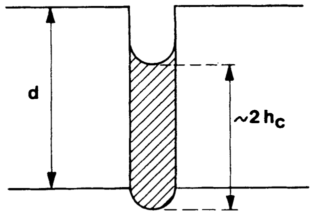

Fig. 4. Cylindrical pore with length $d > 2h_c$ partly filled with water.

at the meniscii on the lower side of the disc to move freely around the pore openings and along the flat portions of the surface between the pore openings. What might happen then is that a somewhat gravity-deformed, rather thick water film will tend to form hanging in the entire lower disc surface from which droplets are generated until the water level $\sim h_c$ is reached. If the upper menisci disappear, however, immediate breakthrough of water occurs.

However, we shall have to assume also that the porous disc is fitted into a glass tube with a wide bore having a perfectly wetting inner surface. Drainage will then preferentially take place along this inner surface of the wide tube. In fact, at the perpendicular contact between the porous disc and the glass tube a meniscus will form which exerts a »capillary pressure« acting to drain the disc-supported water film. It will act similarly as so-called Plateau borders in foams because of its comparatively high curvature (Isenberg 1979).

We now ask ourselves at what pressure level the flow of water through the porous disc will stop in this case, *i.e.* what the mechanical equilibrium state actually is like. The answer is that this equilibrium state may correspond to any degree of water saturation of the porous disc depending upon the water level in the wide tube below the disc (Fig. 5). The radius of curvature of the quasi-cylindrical meniscus at the disc/tube contact will adjust so as to balance the action of gravity on the water film adhering to the tube wall. The same underpressure will also act immediately inside the lower meniscus, hence determining its radius of curvature.

Depending upon the exact location of the lower water level any radius of curva-

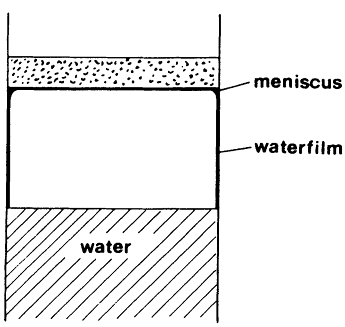

Fig. 5. Porous disc fitted into a glass tube of wider bore. The meniscus curvature is adjusted to match the height over the water level.

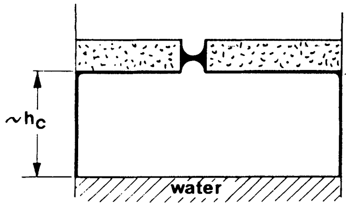

Fig. 6. Porous disc located above a water level corresponding to an almost empty cylindrical pore.

ture of the lower meniscus can hence be realized. When this radius approaches the pore radius the water content in the porous disc tends to zero (Fig. 6). At this state the water level in the wide pore is evidently h_c. Thus, at equilibrium, the degree of saturation in the porous disc will strongly depend on this lower water level. If it is below $\sim h_c$ the water content in the disc will be $\sim$ zero.

In summary we may claim that a porous disc of thickness d containing cylindrical pores with perfectly wetting walls but with a hydrophobized lower disc surface (which is unlikely in reality) can support an overpressure of $\sim h_c$ when $d < h_c$ (*i.e.* there are no upper meniscii). For $d > 2h_c$ a water column of height $\sim 2h_c$ will, at mechanical equilibrium, be present in the porous disc.

In contrast, when the entire disc, inclusive of its lower horizontal surface, is hydrophilic and exhibits perfect wetting by water, the situation is more complex but is similar to the case of two coupled capillaries (Fig. 7) where the lower capillary has a variable inner diameter. Complete drainage of the upper capillary may then occur as soon as the diameter of the lower capillary becomes smaller than the diameter of the upper capillary. However, in practice the transport capacity of water flow in the coarse layer will be very small under conditions of a Plateau border meniscus. The water film on the particles in the coarse layer is thin and the hydraulic conductivity is therefore low (*cf.* Fig. 2). An increased infiltration rate will destroy the Plateau border meniscus and the negative pressure on top of the coarse layer disappears.

The various situations above are summarized in Table 1.

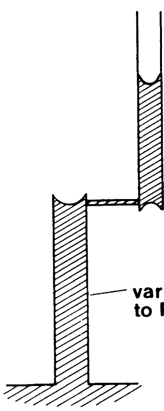

Fig. 7. Two coupled capillaries modelling the equilibrium state of a porous disc fitted to a wide bore glass tube.

Table 1 – Maximum possible water column sustained in three different pore configurations. h_c is height of capillary rise and H is height above the water table of the lower disc interface.

	hydrophilic capillaries no lateral wetting	hydrophilic capillaries lateral wetting	Plateau border
with upper meniscus	$2h_c$	h_c	h_c-H
without upper meniscus	h_c	0	0

Fine Porous Material Adjacent to Coarse Porous Material

Let us now turn to the even more realistic case when the porous glass disc (or a layer of fine-grained material) rests upon a support of coarse-grained material. Provided that saturation humidity is retained so that there is no drying out of the water films adhering to the disc and particle surfaces we should anticipate a behaviour that is quite similar to what we have discussed in the previous subsection.

Meniscii of variable radii of curvature may form at the contacts between the coarse particles and between the coarse particles and the disc. At mechanical equilibrium the geometries of all these meniscii will be determined by the general water level in the volume containing the coarse particles. Accordingly, this water level has to be maintained sufficiently high, otherwise complete drainage of the porous disc may occur.

The degree of water saturation will then in practice be determined by the flow rates which control the water level in the volume with the coarse particles. If the water drainage of this volume is chosen to be comparatively efficient, one will easily run the risk to dry out completely the fine-grained material. For a very fine-grained barrier layer the water level can be kept comparatively low. At the same time, however, the flow rate through the fine-grained layer diminishes substantially.

Discussion and Conclusions

It has been shown that capillary barriers may act to reduce infiltration as long as $\psi(\theta) < 0$ at the bottom of the fine layer. Following the discussion in the previous section, it is deemed highly improbable that the capillary barrier would function also at saturation when the upper menisci disappear and ponding conditions prevail. As shown earlier the only case where this may (in theory) occur is when the fine material is equivalent to a collection of *non-interacting* capillaries. In addition, the underlying coarse layer must be either hydrophobic or the height above the water table small. Since most soils are hydrophilic in character and the choice of the fine layer material is based on this property, the condition of no lateral interaction along the lower interface of the fine layer is most certainly not fulfilled. A thin layer of hydrophobic material between the fine and lower coarse layers might open some interesting possibilities. However, the hydrophobic material would probably be of organic origin and thereby subjected to long-term degradation reactions. The hydrophobic surfaces would then gradually become hydrophilic.

If the capillary barrier then should at all function, the pressure at the interface must everywhere be negative (as well as no draining by the underlying coarse layer). The pressure conditions are dependent on the length of the interface, the slope of the interface, the thickness of the fine layer, the type of soil in the fine layer, the water influx at the surface, and the total volume of water infiltrated during a storm event. These conditions were in detail investigated numerically by Frind *et al.* (1976) for two different soils in the fine layer.

Detailed calculations are not performed here. However, we will give a simple, conservative, estimate to determine if the lateral flow capacity is large enough to avoid positive pressures at the bottom of the fine layer.

Assume that the angle between the horizontal plane and the sloping interface is α giving a hydraulic gradient in the lateral direction of $i = \sin\alpha$ m/m. Accordingly the maximal water flow (*i.e.* at saturation) laterally would be $Q_L = K_s \sin\alpha\, B\, 1$ m^3/s where K_s is the saturated hydraulic conductivity and B the thickness of the fine layer. If the infiltration rate is N m/s the flow rate infiltrating at the top surface is $Q_T = N \times 1 \times L \cos\alpha$ m^3/s. Accordingly for no vertical flow through the fine layer $Q_L \geqslant Q_T$ or

$$N < \frac{K\,B}{L} \tan\alpha \qquad (7)$$

Consider the following specific example. In Sweden the possibility of using capillary barriers to reduce water infiltration into mine tailings, containing pyrite, has been explored. Typically, in these waste rock dumps water must be transported laterally over distances of at least 75 m (Qvarfort 1983) in order not to percolate downwards. Interface slopes larger than 5 % are difficult to produce in practice. Assuming a thickness of $B = 0.3$ m we obtain

$$N < 2\times10^{-4}\, K_s$$

Accordingly, the saturated hydraulic conductivity must be larger than 1×10^{-4} m/s in order to balance an infiltration rate of 0.1 mm/h. A hydraulic conductivity of this magnitude is found only in coarse materials with low capillarity. This is in conflict also with the second purpose of the fine layer: to reduce inward gaseous diffusional transport of oxygen. For this a high moisture content and a high capillarity is needed. In practice it will be difficult to avoid ponding above the fine layer and thereby eliminating the capillary barrier effect. The percolation downwards will then essentially be governed by the hydraulic conductivity of the fine layer. However, the low hydraulic conductivity of the fine layer together with higher hydraulic conductivities in the layers above it, could diminish infiltration to the waste rock significantly (Collin and Rasmuson 1986).

On the other hand, for specific hazardous wastes in small dumps the technique may be quite feasible. Not only are the distances smaller but it should also be possible to create steeper slopes. Fρr example, if $L = 5$ m, $\tan\alpha = 0.10$, and $B = 0.3$ m, K_s must be larger than 4×10^{-6} m/s to balance an infiltration rate of ~ 0.1 mm/h.

Finally, it is noted that the capillary breaking layer may be turned up-side down preventing upward capillary water transport in response to a drying event. This would slow down the drying of a high-moisture content gaseous diffusion resistance layer since water vapour diffusion is a slow process.

Acknowledgements

Several stimulating discussions with Professor Carol Braester at the Dept. of Civil Engineering, Technion, Haifa, are gratefully acknowledged. The study was sponsored by the research program "Environmental hazards from the storage of tailings from the sulphide ore processing" administered by the National Swedish Environmental Protection Board.

Notation

B	– thickness of fine layer	m
d	– thickness of porous disc	m
g	– gravitational acceleration	m^2/s
H	– height above water table	m
h	– water column height	m
h_c	– height of capillary rise	m
K	– hydraulic conductivity	m/s
L	– length of interface	m
N	– infiltration rate	$m^3/m^2/s$
Δp	– pressure difference across curved interface	N/m^2
R	– radius of capillary	m
r	– radius of curvature	m

Greek letters

α	– inclination of sloping interface	
γ	– surface tension of air-water interface	N/m
θ	– water content, vol H_2O/vol tot	m^3/m^3
ϱ	– density of water	kg/m^3
ψ	– pressure head	m of H_2O

Subscript s – value at saturation

References

Adamson, A.W. (1976) *Physical Chemistry of Surfaces,* 3rd ed. Wiley.

Childs, E.C. (1967) Soil moisture theory, in *Advances in Hydroscience* (V.T. Chow ed.), Academic Press, New York 4, 73.

Collin, M., and Rasmuson, A. (1986) Efficiency and design of layered soil covers for mine tailing dumps containing pyrite, Report to SNV, April.

Corey, J.C., and Horton, J.H. (1969) Influence of gravel layers on soil moisture content and flow, E.I. DuPont de Neumors and Company, Savannah River Laboratory, OP-1160.

Frind, E.O., Gillham, R.W., and Pickens, J.F. (1976) Application of unsaturated flow properties in the design of geologic environments for radioactive waste storage facilities, Proceedings of the First International Conference on Finite Elements in Water Resources, Princeton University.

Isenberg, C. (1978) The Science of Soap Films and Soap Bubbles, Tieto Ltd, Clevedon.

Lindsay, L.E., Gillham, R.W., and Frind, E.O. (1979) Studies of engineered geologic environments for radioactive waste storage, Final report Department of Earth Sciences, University of Waterloo, Ontario.

Madsen, B. (1983) Den Kapillære Barriere. En alternativ overdækningsmetode ved deponering af flyveaske, Danmarks Geologiska Undersøgelse (in Danish).

Pierotti, R.A., Blankenship, D.W., Tallent, M.A., and DeVita, F.A. (1986) Adsorption-desorption hysteresis scanning isotherms for Kr and Xe on controlled pore glass. Paper presented at the 60th Colloid and Surface Science Symposium in Atlanta, June 15-18.

Qvarfort, U. (1983) En kartläggning av sandmagasin från sulfidmalmsbrytning; The National Swedish Environment Protection Board, Report SNV PM 1699 (in Swedish).

Rançon, D. (1972) Structures Sèches et barrières capillaires en milieux poreux - Application au stockage dans le sol. Département de Sûreté Nucleaire, France, Rapport CEA-R-4310.

Rançon, D. (1979) Application de la technique des barrières capillaires au stockages en tranchées, Proceedings of IAEA Symposium: Underground Disposal of Radioactive Wastes, Otaniemi, Finland.

Swartzendruber, D. (1969) The flow of water in unsaturated soils, in *Flow Through Porous Media* (R.J.M. de Wiest ed.), Academic Press, New York 215.

Received: 11 April, 1988

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