

Hydrous transfer within fibrous-cement slates

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ABSTRACT

In-situ distortion of fibrous-cement slates is brought about by exposure to different temperature and hygrometric gradients across the material. The aim of the research was to understand in detail the effect of each compositional change on the physical properties, and to use this knowledge for the manufacture of slates which would exhibit better mechanical properties. The present research has concentrated on measuring the physical parameters controlling moisture transfer. A second paper (in preparation) shall use these experimental results in a numerical model to simulate moisture movements. The key physical parameters measured include: porosity, saturated permeability, vapour diffusivity and the suction-moisture content relationship. The experimental results show clearly the effects of various treatments (painting the slate, adding additional layers, replacing the fibres, slate compaction) on the physical properties.

RÉSUMÉ

Les déformations locales dans les ardoises de fibro-ciment peuvent être engendrées par l'existence de gradients de température et d'hygrométrie au sein du matériau. Le but de ce travail est de comprendre en détail l'effet des modifications de composition des ardoises sur leurs propriétés physiques et d'utiliser ces connaissances dans la fabrication d'ardoises présentant de meilleures propriétés mécaniques. Le travail présenté dans cet article est axé sur l'étude et la mesure des paramètres physiques influant sur les transferts d'humidité. Dans un second article (en préparation) nous présenterons l'élaboration et l'exploitation d'un modèle numérique apte à décrire les phénomènes étudiés. Les paramètres pertinents sont : porosité, perméabilité saturée, succion et perméabilité en fonction de l'humidité relative. Les résultats expérimentaux montrent clairement l'effet des traitements subis par les ardoises (peinture de la surface, ajout d'une couche minérale supplémentaire, remplacement de l'amiante par d'autres fibres, compactage des ardoises) sur leurs propriétés physiques.

1. INTRODUCTION

There are two major problems encountered by fibrous-cement slates in use: distortion and frost damage. Both of these defects are closely related to moisture movements occurring within the slate. The existence of moisture gradients within the slate thickness can produce a distortion due to a moisture shrinkage gap between the two faces. This gap is caused by exposure to thermal and hygrometric gradients across the slate, which creates a moisture gradient due to unsymmetrical hydrous movements. Distortion may produce cracks, which impair the slates' protective capacity. Due to the

presence of fibre pockets close to the surface, frost damage can appear after exposure to freeze-thaw cycles. The fissures produced are due to the presence of large pores surrounding the fibre pockets. The main aim of this study was an attempt to reduce slate distortion; therefore, frost damage was neglected.

The aim of the research was to understand in detail the effect of each compositional change on the physical properties, and to use this knowledge for the manufacture of slates which would exhibit better mechanical and physical properties. Another goal was a better understanding of hydrous movements occurring within the different slates. The numerical finite element model,

Editorial note

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NSAT (Non-saturated), would lead to a greater knowledge of hydrous transfer under different conditions, which could then be used to improve the slates' properties. Once a theoretical model was obtained, and had been validated by experimental results, it could be used to predict a material expected to have improved properties, in particular, to display less distortion. Work began with an experimental determination of the physical parameters which control hydrous transfer. These parameters are the porosity, the saturated permeability, the suction and the permeability relationships. A second article, in preparation, will detail the results from the numerical simulations, using the experimental values obtained in this paper.

2. MATERIALS SELECTED FOR THE INVESTIGATION

For the experimental study six materials were chosen, which involved variation of the composition, the fibres and the finishing processes. The composition of each slate is shown in Fig. 1. The composition of the ordinary slates was: 78% Ordinary Portland cement (OPC - type 55R), 10% asbestos fibres (of diameter 0.1 to 2 μm and length of 50 to 1000 μm), 2% colourant (pigments) and 10% filler (crushed limestone - fineness of 3000 $\text{cm}^2 \text{g}^{-1}$). The water:cement ratio was 0.25. At the end of the fabrication process, the slates were compressed with a pressure of 280 bars. The raw sheets of fibrous slates were piled up (separated by steel bars) and oven-cured at 55% relative humidity and at a temperature of 70 $^{\circ}\text{C}$ for six hours. After this period, the slates left the oven and were left at room temperature for a further eight hours before they were separated. The slates were further conserved at the factory for a period of two weeks before they were painted. An acrylic resin-based emulsion paint was used. A layer of approximately 30 μm was painted onto each side of the slate.

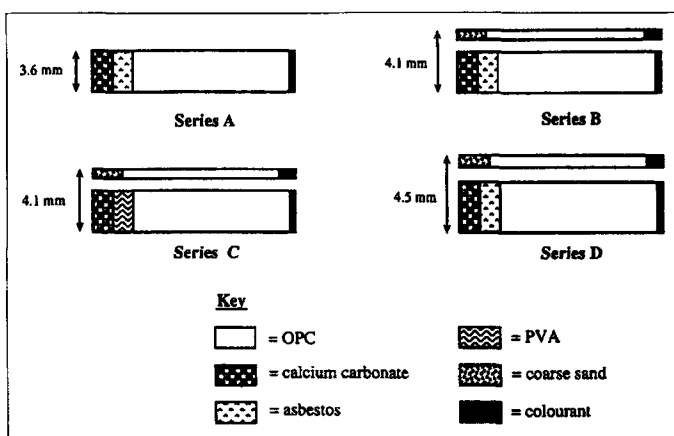


Fig. 1 – Composition of the slates chosen for the present investigation.

The ordinary slates (composition detailed above) were labelled series A. The series B slates had the same composition as series A, except that an additional layer

(top layer) was added in which the asbestos fibres were omitted. The top layer (thickness of 0.4 mm) contained 15% coarse sand (granulometry of 0 - 400 μm), 80% of OPC, and 5% colourant. In both these series, painted (P) and unpainted (UP) slates were analysed to evaluate the effect of the painting process on the material's behaviour. Series C [C-PVA] (painted), similar to series B, was chosen to enable an assessment of changes produced by the replacement of asbestos fibres by a mixture of cellulose and PVA (polyvinylalcohol) fibres. The cellulose fibres were bleached kraft softwood pulp and had a length of 1 - 3 mm and a width of 20 - 30 μm . The synthetic PVA fibres had a diameter of 50 μm and a length of several millimetres ($\approx 6 \text{ mm}$). Series D [D-UC] were the same as the B-series slates; however, they were neither painted nor compressed. The D-series was selected to evaluate the changes caused by compression at the end of the manufacturing process.

3. DETERMINATION OF THE PHYSICAL PARAMETERS CONTROLLING HYDROUS TRANSFER

There are four essential parameters (porosity, saturated permeability, suction and permeability relationships) required for the use of NSAT, as mentioned above. A detailed explanation of this work may be found in the thesis by Raouf [1].

3.1 Determination of the structural parameters

Three structural parameters are often used to characterise porous materials: porosity, pore size distribution, and specific surface [2]. The porous nature of a material influences such properties as strength, permeability, durability, fracture toughness, and elastic modulus [3]. The mechanisms of moisture movement are dominated by the structure of the porous system and consequently, it is important to determine accurately these three structural parameters. Several techniques were used to measure these three structural properties.

3.1.1 Mercury intrusion porosimetry

Mercury intrusion porosimetry is a well-established technique [4, 5], which gives access to several parameters, including the total porosity and the pore size distribution of materials. A Micromeritics Autopore II 9215 analyser, which can detect pore diameters in the range of 1000 μm to 0.002 μm (20 \AA), was used.

Results for all the samples may be seen in Figs. 2 and 3, which illustrate the log differential intrusion and pore diameter relationship. The relevant measured values are given in Table 1. The results show that the A and B series slates have a quite similar pore size distribution. The main difference for the unpainted A (A-UP) and B (B-UP) series slates is the presence of a region of capillaries (pore width $> 2 \mu\text{m}$) around 200 μm , created by

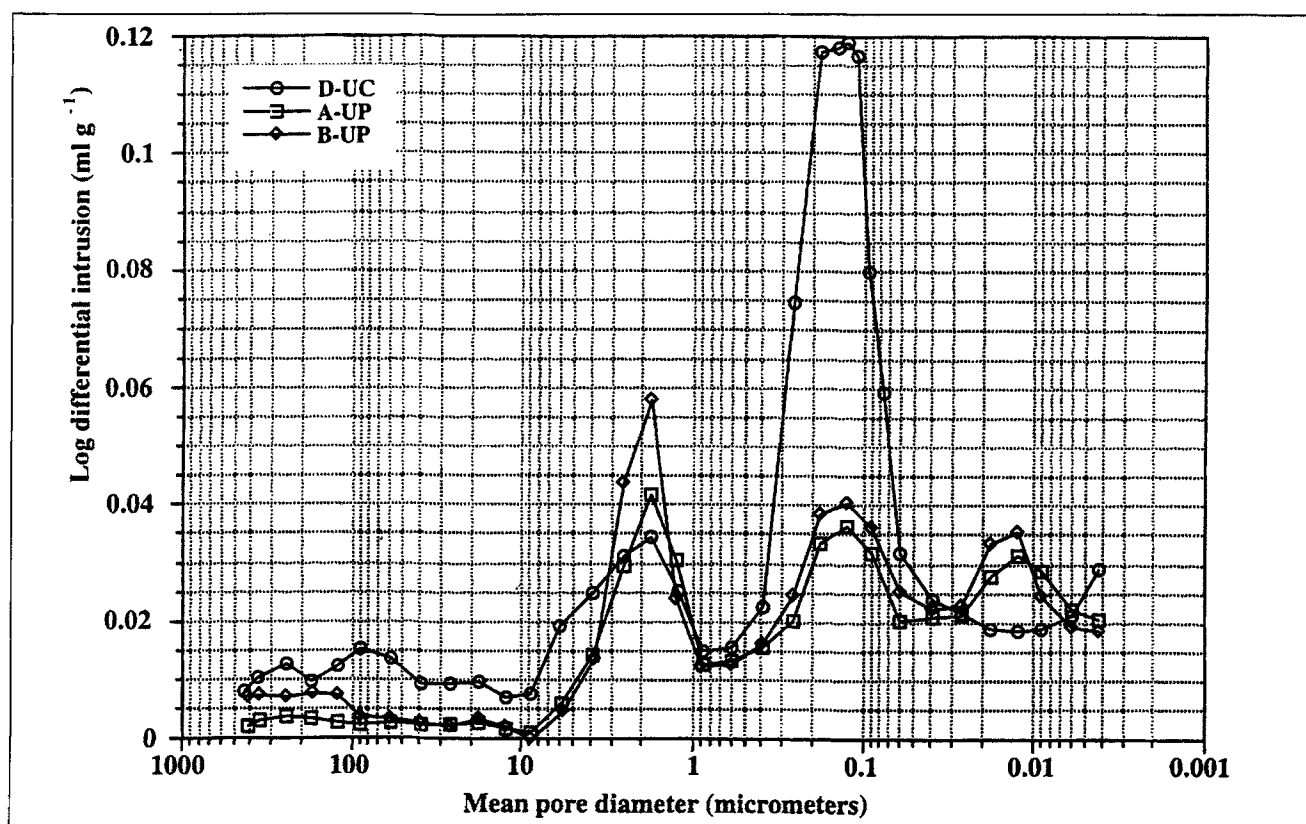


Fig. 2 – Mercury porosimetry results for the unpainted slates.

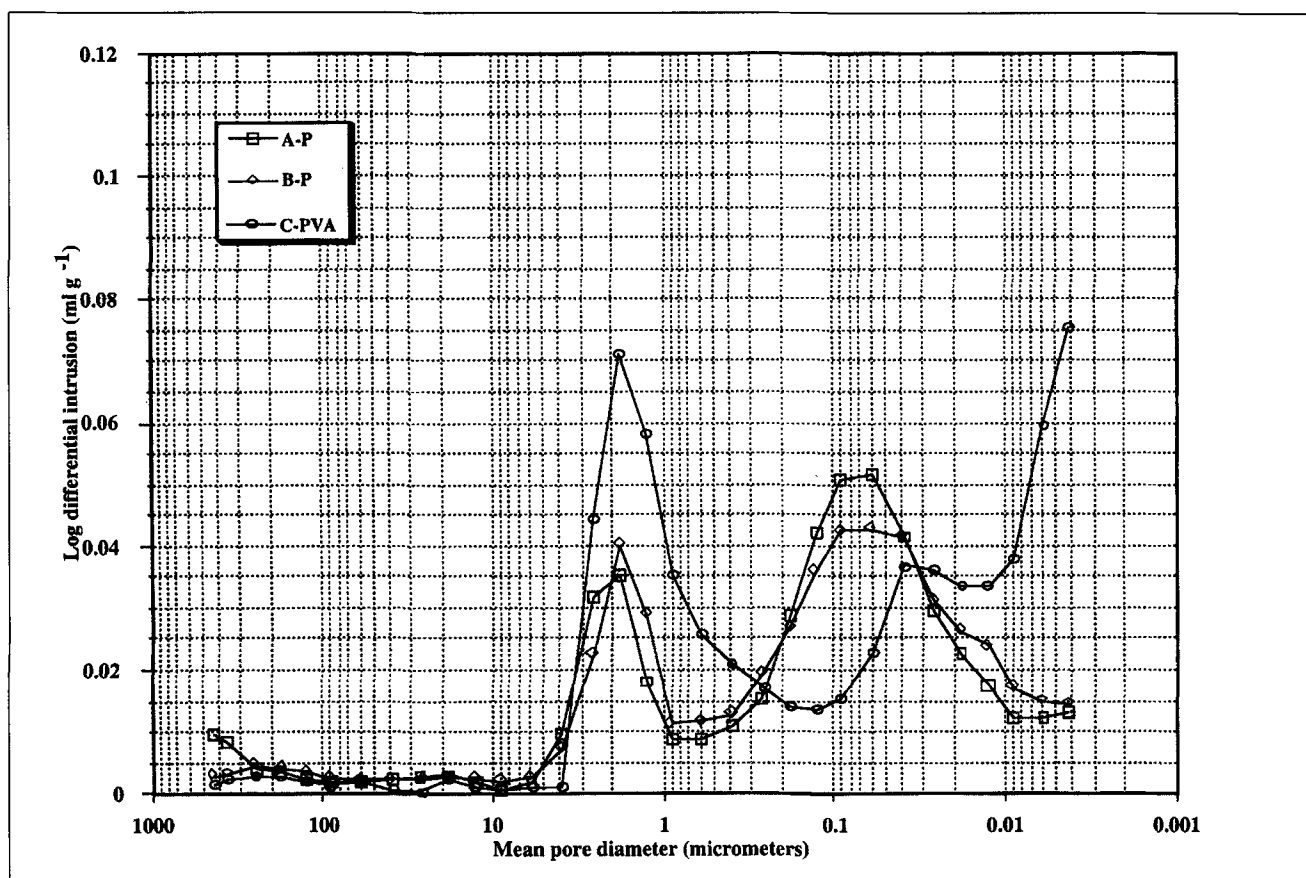


Fig. 3 – Mercury porosimetry results for the painted slates.

the use of coarse sand in the top layer, which is absent in the A series. No general trend in the porosity was discovered on the addition of the top layer. The failure to

observe a general trend may be attributed to the heterogeneous nature of the fibrous slates. In general, it was assumed that a rise in porosity would occur for the

Table 1 – Summary of the main results from the mercury porosimetry experiments

Sample	A-P	A-UP	B-P	B-UP	C-PVA	D-UC
Total intrusion volume (ml g^{-1})	0.08	0.08	0.08	0.09	0.10	0.14
Total pore area ($\text{m}^2 \text{g}^{-1}$)	10.2	12.3	10.0	11.5	20.3	11.8
Bulk density (g ml^{-1})	2.03	2.04	2.03	2.04	1.84	1.77
Total open porosity (%)	16.76	17.13	16.35	18.28	18.69	24.54
Median pore diameter (μm)	0.09	0.12	0.09	0.13	0.17	0.14

B-series slates due to the presence of coarse sand and the absence of asbestos fibres in the top layer.

The replacement of asbestos fibres by a mixture of cellulose and PVA fibres has an important effect on the porosity and the pore size distribution (Fig. 3). There is a 2% increase in porosity for the PVA slates (C-PVA). The increased porosity is due to the very different geometries of the two fibres used. Asbestos fibres have a diameter of 0.1 – 2 μm and a length of several micrometers, whereas the synthetic PVA fibres have a diameter of 15 μm and a length of several millimetres ($\approx 6 \text{ mm}$). The uniformity and much greater size of the PVA and cellulose fibres will make it harder for these fibres to ‘fit into’ the cement matrix as well as the asbestos fibres. The PVA slates have a much greater quantity of capillaries and mesopores (2 $\text{nm} < \text{pore width} < 50 \text{ nm}$). Once again these differences can be ascribed to the different dimensions of the two fibres.

The unpainted slates have three dominant pore sizes: capillaries, macropores (50 $\text{nm} < \text{pore width} < 200 \text{ nm}$), and mesopores. Each type of pore appears to be present in approximately equal quantities within the unpainted slates. However, the painted slates show only a bimodal pore size distribution: capillaries and macropores. Macropores are present in greater quantities within the painted slates. As well as a change in the pore size distribution, a reduction in the porosity was also observed for the painted slates. The painting process reduced the porosity of the A and B-series slates by 2% and 0.4%, respectively. It is possible that the paint (applied to both sides of the slate) is capable of blocking the mesopores and micropores within the slates, especially at the surfaces. Hence, the paint reduces the porosity of the slate.

The uncompressed slate (D-UC) had the largest porosity of 24.5%. It displayed a much broader distribution of pore sizes, and a far greater quantity of capillaries. The compaction of the slate reduces the volume of the slate, hence reducing the porosity; at the same time, the sizes of the pores present are also reduced. The largest quantity of pores appears in the 0.02 μm region, indicating an abundance of macropores. The compaction of a slate (reduction in the thickness of the slate from 5 mm to 4 mm) compresses the cement matrix and reduces the quantity of macropores by more than half.

3.1.2 Measurement and analysis of sorption isotherms

The suction-water content relationship (sorption isotherm) enables the total porosity and the pore size distribution to be determined [6]. The principle of the method used involves allowing a sample to reach moisture equilibrium at a known relative humidity, which is created by using saturated saline solutions in a desiccator [7, 8]. The samples were maintained at a temperature of $23 \pm 0.1^\circ\text{C}$. The saturated saline solutions used produced the following relative humidities: 97, 93, 81, 66, 55, 44, 33, 22, and 12%. These relative humidities may be converted into capillary pressures (suction) by the use of the equation of vapour pressure equilibrium.

The results are presented in Fig. 4. The water content of the slates has been converted into relative saturation values to enable a better comparison of the drying behaviour. It was found that, in general, 40 days were necessary for the system to reach equilibrium at the higher relative humidities and approximately 70 days at

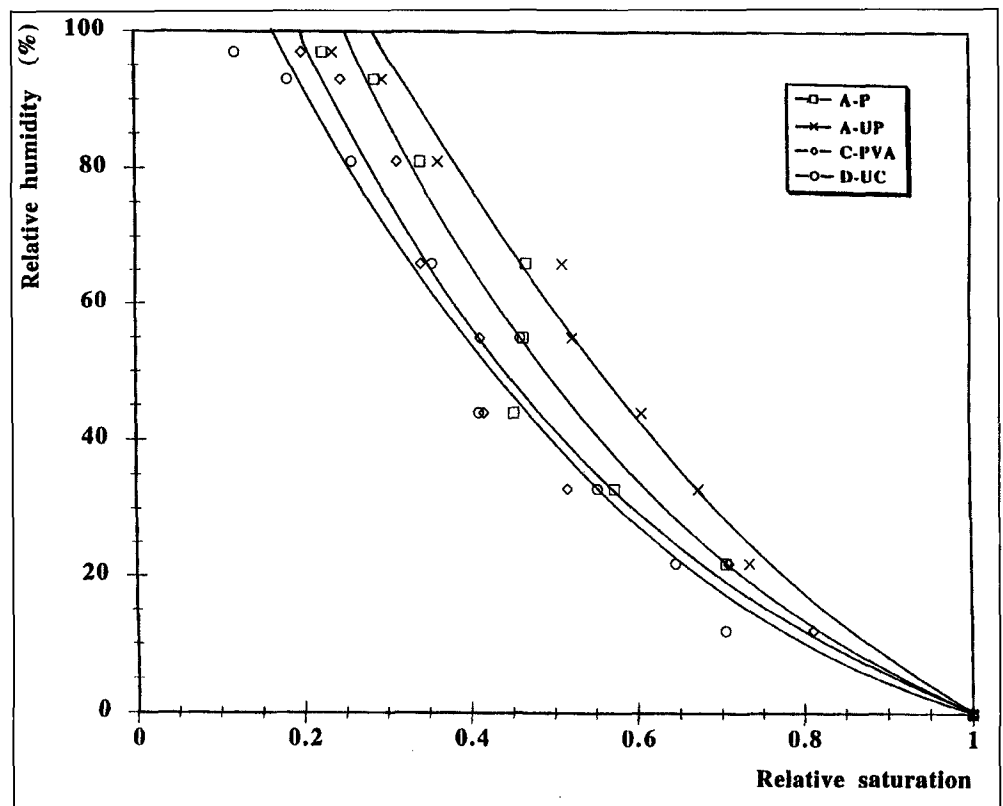


Fig. 4 – Suction graph for four slate products.

the lower relative humidities. The bulk of the moisture loss, however, occurred during the first seven days for all the samples. All the graphs have a quite similar shape. The water contents at saturation vary between 20.7% for the uncompressed slate and 17.7% for the unpainted A-series slate.

A drying order can be established from the results in Fig. 4. The drying order is as follows:

$$\frac{D-UC > A-UP > A-P > C-PVA}{\text{Decrease in drying capacity} \Rightarrow}$$

The establishment of the drying order is an important result, because it can be used to predict the likely behaviour of the slates under similar environmental conditions.

The water contents from suction porosimetry were converted to open porosities. The porosities calculated from suction porosimetry are twice as large as those obtained from mercury intrusion. A comparison between the two techniques is given in Table 2. This suggests that all the samples had a significant quantity of small mesopores and micropores which were undetected by mercury intrusion. Many other authors have also found a discrepancy between the results from mercury intrusion and other techniques [9, 10].

The results from suction porosimetry reinforce the

Table 2 – Comparison of porosities obtained from mercury and suction porosimetry

Sample	Porosity from suction data (%)	Porosity from mercury porosimetry (%)
A-P	38.1	16.8
A-UP	35.4	17.1
C-PVA	42.1	18.7
D-UC	51.4	24.5

earlier findings from mercury porosimetry: that replacing asbestos fibres by PVA fibres increases the porosity; that uncompressed slates have a much higher porosity than compressed slates; and that painted slates have a smaller porosity than unpainted slates.

3.2 Hydrous transport properties

An important hydrous property required for the numerical model is the permeability-water content relationship. This relationship was determined in two stages. Initially, the saturated permeability was measured and then, the vapour diffusivity-water content relationship was determined, which allowed the calculation of the permeability-water content relationship.

The saturated permeability was calculated by the use of Darcy's law [11]. The sample to be analysed was vacuum saturated for five days. Once saturated, it was placed in a cell and a column of water (height = 1.3 m) was applied to the sample. The experiment involved

inflow/outflow measurements. The samples were maintained at a temperature of 23 ± 0.1 °C. The saturated permeability of two samples (A-UP and B-UP) was measured. The average value of 35 measurements, obtained for B-UP and A-UP slates, was 3.3×10^{-13} and $2.9 \times 10^{-13} \text{ ms}^{-1}$ respectively. For both samples, the measured saturated permeability fluctuated between 10^{-12} and 10^{-13} ms^{-1} . The present results agree well with the results of previous studies of HCP (hardened cement paste). Powers, Copeland et al. [12] measured the permeability coefficients of fresh HCP pastes, w:c = 0.5, to range from 5×10^{-7} to $8 \times 10^{-7} \text{ ms}^{-1}$. As the pastes hardened and matured, the permeability decreased and varied from 1.0×10^{-15} to $1.2 \times 10^{-15} \text{ ms}^{-1}$. Similar results were also found by Nyame and Illston [13] for HCP. These authors investigated the influence of the water: cement ratio and the time of hydration on the saturated permeability. For a HCP with a w:c ratio of 0.23, the saturated permeability was measured at 14 days to be $7.0 \times 10^{-13} \text{ ms}^{-1}$; after 28 days the value had decreased to $5.0 \times 10^{-13} \text{ ms}^{-1}$. Over the following months the value stabilised to a value of $2.0 \times 10^{-14} \text{ ms}^{-1}$ (after 20 months).

The present findings correlate broadly with the results of previous studies. Any differences with earlier results for the saturated permeability are likely to arise from the fact that this study analysed fibrous-cement materials and not HCP. It is well known that the presence of fibres modifies the porosity and pore size distribution of a material. This modification of the microstructure is also likely to affect the saturated permeability.

The vapour diffusivity measurements were conducted using the classic 'cup-cup' method, where a different hygrometric environment is imposed on each side of the slate sample. The samples were maintained at a temperature of 23 ± 0.1 °C. The lower compartment had a higher relative humidity than the upper one; thus a vapour pressure difference was created, which led to moisture transfer. The weight of the top cup was measured regularly, so that the moisture movements could be followed accurately. The above procedure was repeated several times with different hygrometric gradients, allowing the construction of the complete vapour diffusivity graph.

The permeability-relative humidity relationship is presented in Fig. 5. In the measured relative humidity region, the permeability of all the samples increased by a factor of ten between values of 10^{-16} and 10^{-17} ms^{-1} . The measured permeabilities are lower than the saturated permeabilities by a factor of 1000 at the higher humidities.

The saturated permeability was determined experimentally to have an average value of 10^{-13} ms^{-1} . The present results indicate that, at a relative humidity of 85%, the permeability declined to a value of 10^{-16} ms^{-1} . As the humidity continued to decline to 20%, the permeability decreased by a factor of ten to 10^{-17} ms^{-1} . The greatest change in the permeability occurs, therefore, when the relative humidity decreases from 100% to 85%. The large permeability variation indicates that a small amount of drying is sufficient to dry the surface layers, which limits the evaporation process. The initial drying may

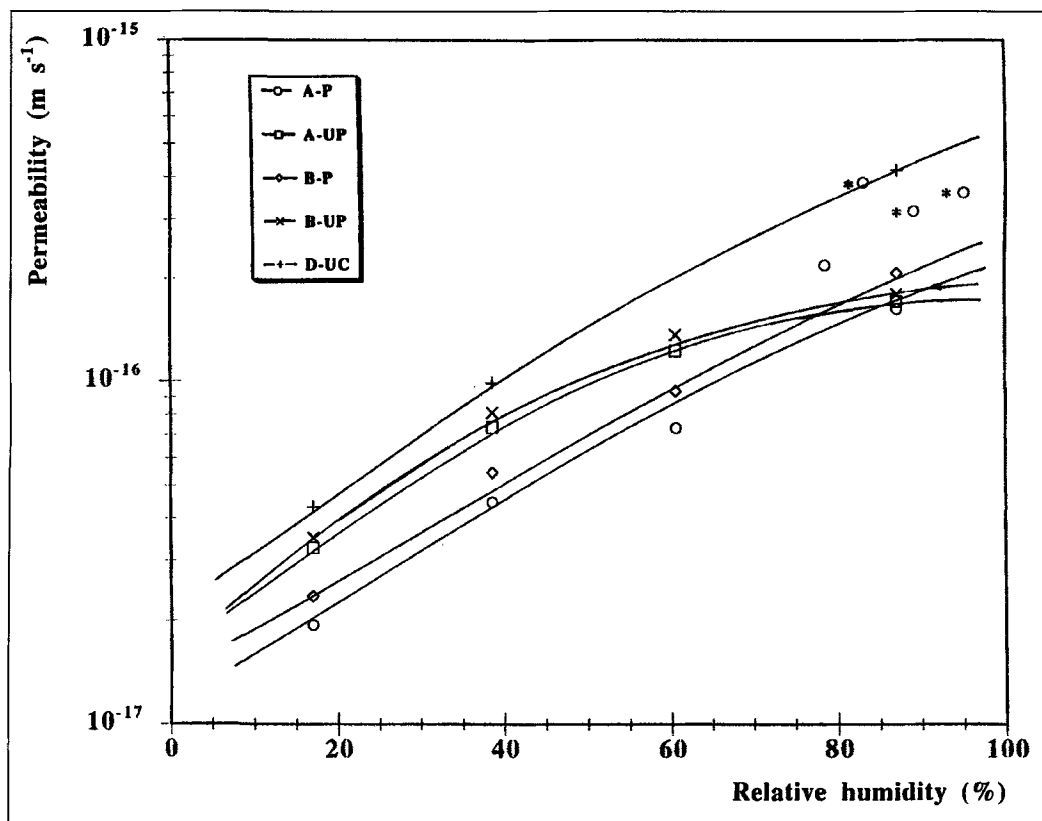


Fig. 5 – Evolution of the permeability as a function of the relative humidity.

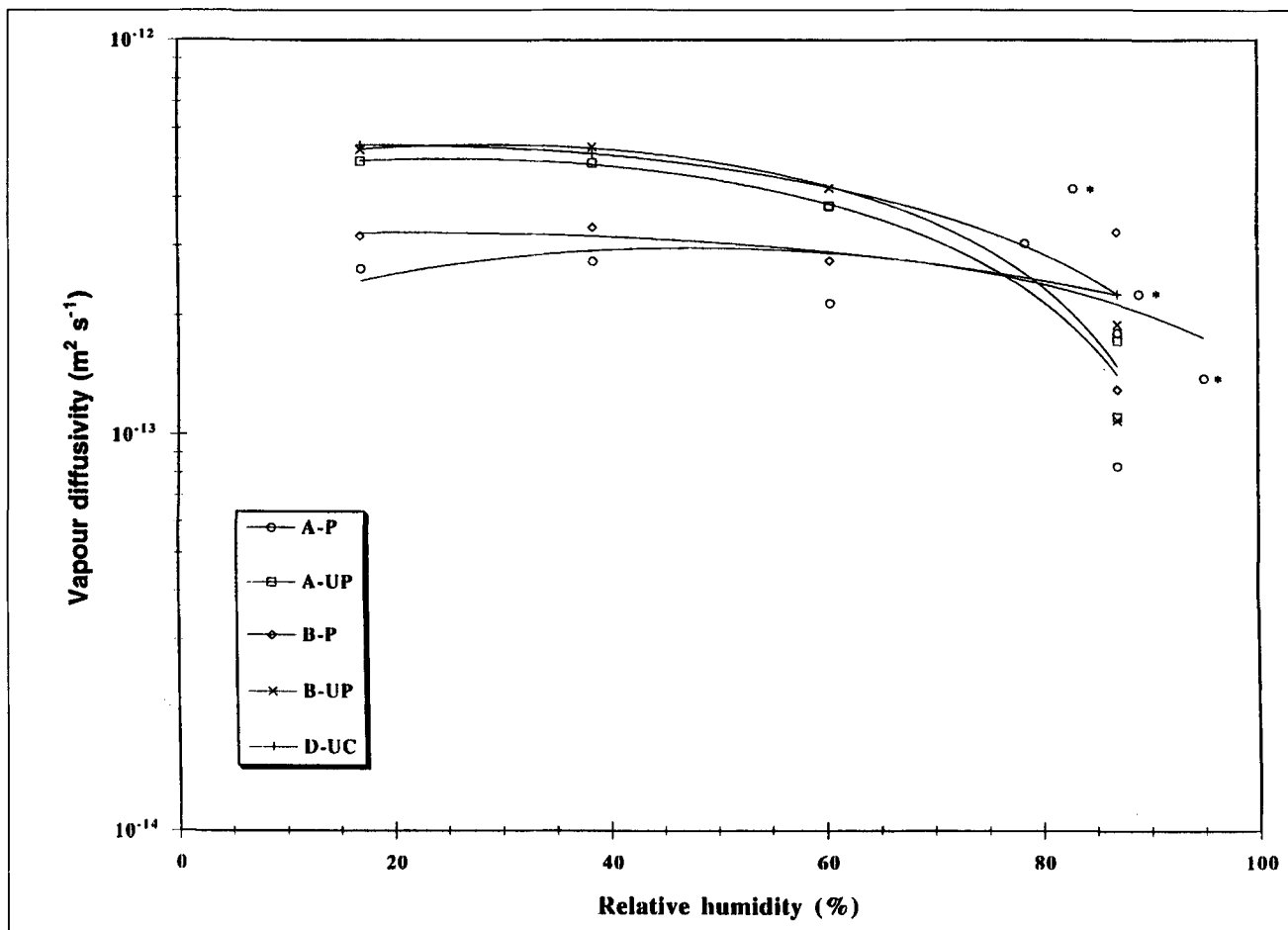


Fig. 6 – Evolution of the vapour diffusivity as a function of the relative humidity.

also create numerous discontinuities in the pore water network, thus greatly reducing the diffusion process.

The graph shows that the permeability increases as the relative humidity increases. For the unpainted slates from the A and B-series, the permeability tails off above a relative humidity of 60%. The uncompressed slate had the largest permeability, due to its higher porosity and a greater proportion of larger pores. The painted slates had a lower permeability than their unpainted counterparts, indicating the possible blocking effect of the painting process.

The experimental points for the A-UP slate (in Figs. 5 and 6) which are asterisked, are from measurements conducted with initially saturated slates. The development of a hysteresis of the diffusivity coefficient is seen between the drying/saturation conditions. Therefore, it is necessary to state the exact sample moisture history when values of the hydrous transport properties are cited.

Fig. 6 shows the diffusivity coefficient and relative humidity relationship. The graph shows that the diffusivity coefficient decreases by almost a factor of ten in the measured range, between 10^{-12} and $10^{-13} \text{ m}^2 \text{ s}^{-1}$. Nilsson [11] measured the diffusivity of three cement mortars with different w:c ratios (0.4, 0.6, and 0.8). The diffusivities measured had values close to $10^{-11} \text{ m}^2 \text{ s}^{-1}$, which are similar to the present values. The results indicate that as the relative humidity increases the vapour diffusivity decreases. The fall in the vapour diffusivity becomes more accentuated as the relative humidity increases above 60%. These results are in agreement with previous observations. Rose [12] measured the vapour diffusivities of several porous materials (soils, sandstones). He found that as the moisture content of the materials increased the vapour diffusivity decreased, almost linearly, up to a relative humidity of 60%. Above a relative humidity of 60%, there was simultaneously a greater decline in the vapour diffusivity and a considerable increase in the liquid diffusivity. It was suggested that at 60% relative humidity, liquid condensation begins, which leads to greater liquid diffusion and a restriction of vapour diffusion.

4. CONCLUSION

The physical parameters necessary to enable the use of NSAT have been measured for several different slates. The experimental results lead to a more comprehensive understanding of the effect of modifying the composition of the slates and that of changing the production process on the physical properties of the slates.

The addition of a top layer, as performed in the B-series slates, leads to an increase in the proportion of capillaries in the sample. As a consequence, the saturated permeability of the B-series was found to be marginally higher. No fixed trend in the porosity was established on the addition of the top layer.

The replacement of asbestos fibres by a mixture of PVA and cellulose fibres has a significant effect on the microstructure of the slate. The porosity of the PVA slate is increased and the pore size distribution is altered

(a general shift of pore sizes to smaller diameters). These changes arise from the difference in the physical properties and dimensions of the two fibres. The changes created by the PVA fibres lead to a reduction in the rate of hydrous transfer and hence, should limit the amount of distortion caused by water movements.

Painting the upper slate surface reduces the porosity and the rate of hydrous transfer. The reduced diffusion rate is beneficial for the slate, as it should result in less distortion during climatic changes.

Compaction of the slate at the end of the fabrication process has a significant effect on its physical properties. There is a major decrease in the total porosity and a narrowing of the pore size distribution once the slate has been compressed. The quantity of capillaries is reduced considerably after compaction. The physical properties of the uncompressed slate would probably lead to greater distortion due to the larger hydrous movements involved.

The present results have improved the understanding of hydrous transport in fibrous-cement slates.

5. REFERENCES

- [1] Raoof, A., 'An experimental and numerical study of hydrous transfer within fibrous-cement slates', MPhil. Thesis, Nottingham Trent University, England, 1994.
- [2] Haynes, J. M., 'Determination of pore properties of constructional and other materials', *Materials and Structures* 6 (33) (1973).
- [3] Feldman, R. F., 'Pore structure, permeability and diffusivity as related to durability', 8th Int. Congr. Chem. of Cem., Bresil, Vol. 1 (1986) 336-356.
- [4] Washburn, E. W., 'Note on a method of determining the distribution of pore sizes in a porous material', *Proc. Nat. Acad. Sci. U.S.A.*, 7 (1921) 115-6.
- [5] Ritter, H. L. and Drake, L. G., *Ind. Eng. Chem. Anal. Ed.*, Vol. 17 (1945) 787.
- [6] De Castro, E., 'Determination of the pore-size distribution in stones by means of the moisture suction', 441, *Doc. RILEM*, Lisbon, 1974.
- [7] Croney, D., Coleman, J. D. and Bridge, P. M., 'The suction of moisture held in soil and other porous materials', D. S. I. R., Road Research Technical Paper No. 24, H. M. S. O., 1952.
- [8] Baroghel-Bouny, V., 'Caractérisation des pâtes de ciment et des bétons', LCPC, Paris, 1994.
- [9] Alford, N. McN. and Rahman, A. A., 'An assessment of porosity and pore sizes in hardened cement pastes', *J. Mats. Sci.* 16 (1981) 3105-3114.
- [10] Winslow, D. N. and Diamond, S., 'A mercury porosimetry study of the evolution of porosity in Portland Cement', *J. of Mat., JMLSA* 5 (3) (1970) 564-585.
- [11] Darcy, H., 'Les fontaines publiques de la ville de Dijon', Dalmont, Paris, 1856.
- [12] Powers, T. C., Mann H. M. and Copeland L. E., 'Flow of water in hardened cement paste', *Highway Res. Ed. Spec. Report No.* 40, July 1959.
- [13] Nyame, B. K. and Illston, J. M., 'Pore distribution and permeability of hardened cement pastes', 7th Int. Symp. on Chem. of Cem., Paris, 1980 Vol. III, pp VII-I - VII-5.
- [14] Nilsson, L. O., 'Hygroscopic moisture in concrete - Drying, measurements and related material properties', Ph. D. Thesis, Lund Institute of Technology, Division of Building Materials, Lund, 1980.
- [15] Rose, D. A., 'Water movement in porous materials - II: The separation of the components of water movement', *Brit. J. Appl. Phys.* 14 (1963) 491-496.