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Autor: Kamp, Cornelis L. / Roelfstra, Pieter E. / Wittmann, Folker H.
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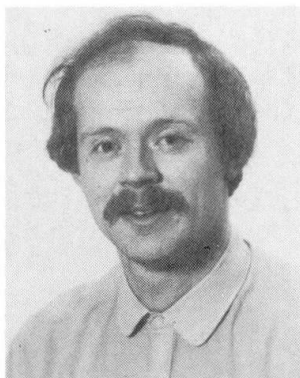
Aspects of Moisture Flow in Hardened Cement Paste and Concrete

Transport de masse dans la pâte de ciment durcie et dans le béton

Feuchtigkeitsdiffusion in erhärtetem Zementstein und in Beton

Cornelis L. KAMP

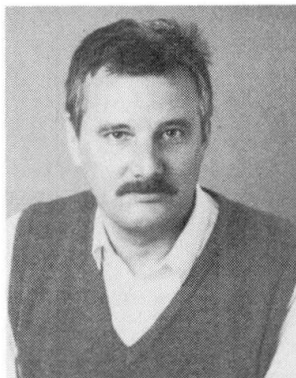
Struct. Engineer
Swiss Fed. Inst. of
Techn.
Lausanne, Switzerland



Cornelis L. Kamp, born 1955, got his civil engineering degree at Delft University of Technology, with honours, in 1981. He worked for several years as a structural engineer at DHV engineering consultants, Amersfoort, and joined the Laboratory for Building Materials at the Swiss Federal Institute of Technology, Lausanne, in 1984.

Pieter E. ROELFSTRA

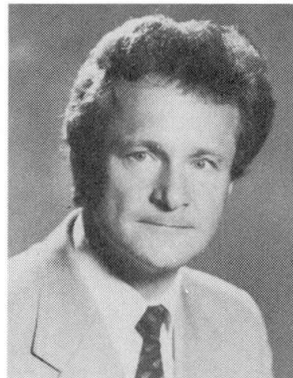
Civil Engineer
Swiss Fed. Inst. of
Techn.
Lausanne, Switzerland



Pieter E. Roelfstra, born 1946, got his civil engineering degree at Delft University of Technology. For 12 years he was involved in structural engineering at the Ministry of Public Works. Actually he is head of the group Modelling and Numerical Analysis of the Laboratory for Building Materials at the Swiss Federal Institute of Technology, Lausanne.

Folker H. WITTMANN

Professor
Swiss Fed. Inst. of
Techn.
Lausanne, Switzerland



Folker H. Wittmann, born 1936, habilitated in Physics of Building Materials at Munich Institute of Technology. For 4 years he was professor for Building Materials at Delft University of Technology, The Netherlands. He is now professor and director of the Laboratory for Building Materials at the Swiss Federal Institute of Technology, Lausanne.

SUMMARY

Different mechanisms of moisture flow in porous media are outlined. Several models are developed and discussed. On the basis of these models it is possible to explain a number of effects commonly observed on drying hardened cement paste and concrete. Numerical methods are applied to test the models quantitatively.

RÉSUMÉ

On discute les différents mécanismes de transport de masse dans les milieux poreux. Des modèles de la structure poreuse de la pâte de ciment durcie sont proposés. Ces modèles permettent d'expliquer les phénomènes habituellement observés lors du séchage de la pâte de ciment durcie et du béton. Des prédictions quantitatives sont obtenues avec des méthodes numériques.

ZUSAMMENFASSUNG

Unterschiedliche Transportmechanismen der Feuchtigkeitsdiffusion in porösen Werkstoffen werden betrachtet. Mehrere Modelle werden entwickelt und diskutiert. Mit Hilfe dieser Modelle ist es möglich, eine Reihe von Erscheinungen, die man gewöhnlich bei trocknendem Zementstein und Beton beobachtet, zu erklären. Mit numerischen Methoden gelingt es über die Modelle zu quantitativen Aussagen zu kommen.



1. INTRODUCTION

HCP (hardened cement paste) has a large internal specific surface and therefore all properties are strongly influenced by moisture content. In addition it is well-known that moisture diffusion is a very slow process. That means that time-dependent moisture gradients exist for a long time in concrete elements. Creep, shrinkage, and crack formation depend essentially on the moisture distribution in a specimen and the stresses created by moisture gradients.

Moisture fields are generally calculated with the diffusion equation :

$$C(H) \frac{\partial H}{\partial t} - \nabla \cdot \lambda(H) \nabla H = Q \quad (1)$$

where : $C(H)$ = moisture capacity, $\lambda(H)$ = permeability coefficient, H = moisture potential, and Q = moisture source.

A convenient choice for the moisture potential is relative humidity; other quantities are sometimes preferred, e.g. suction height. Thermodynamics solely requires that the quantity chosen is uniquely related to the energy of the pore water per unit mass.

The functions $C(H)$ and $\lambda(H)$ are normally determined by experiments. They are defined with the following equations :

$$C(H) = du/dH \quad (2)$$

$$J = - \lambda \nabla H \quad (3)$$

where : J = total water flux, u = water content.

It has to be mentioned, however, that these functions are very difficult to be determined experimentally. These functions depend both on the porous system and on the environmental conditions i.e. the relative humidity and temperature. If one would try to study all those influences in detail it would mean an enormous series of time-consuming experiments. This fact justifies a more thorough analysis of the problem.

2. PHYSICAL ASPECTS OF STORAGE AND FLOW OF MOISTURE

Only a brief review of the main points will be given here. The reader is referred to /7/ for a more thorough discussion.

Porous media can retain liquid water at relative humidities below 100%. Two mechanisms prevent evaporation of the pore water : physical adsorption and capillary condensation.

Physical adsorption is generally viewed as a piling up of molecules on a plane surface, in the presence of attractive forces in the neighborhood of the surface of an adsorbent. Several quantitative treatments have been proposed; the one developed by Brunauer, Emmett and Teller /3/ is the most frequently used.

Direct measurement of physical adsorption is only possible below 40% RH in HCP; above this value capillary condensation falsifies the measurements. An indirect approach is possible, which requires the following steps :

- Measurement of the specific surface area in the low-humidity region with the BET-method.
- Measurement of the physical adsorption on a similar solid in which capillary condensation does not occur.
- Correction of the results obtained under b) for the different surface areas.

Figure 1 shows results obtained by Badmann /1/ for adsorption on unhydrated cement. These results are probably reliable below some 80% RH; above this value, the cement begins to hydrate noticeably.

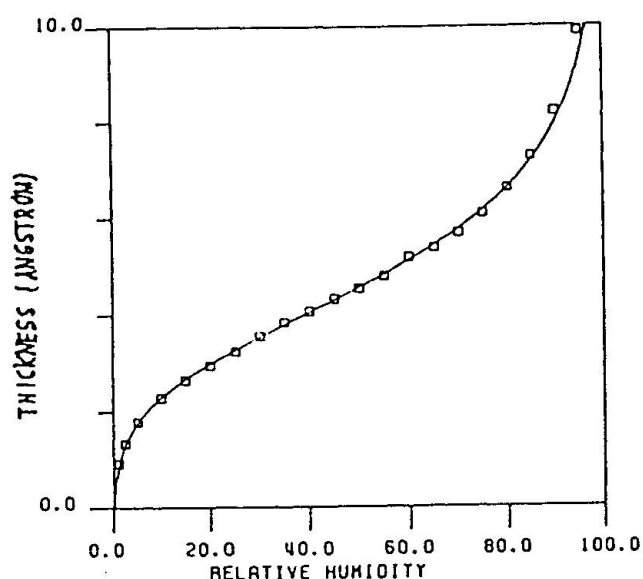


Fig. 1 Physical adsorption on unhydrated cement /1/.

Measured specific surface areas of HCP range from 100 to 250 m²/g; other adsorbates such as N₂ yield only some 20% of this value. This suggests the presence of a "hidden" porosity in the solid material which is accessible to water, but not to nitrogen. The same explanation was proposed by Diamond /4/ for the strikingly different pore-size distributions determined by mercury-porosimetry and vapour adsorption. It might also explain why specific surface areas calculated from pore-size distributions are generally lower than BET-areas determined from the water-vapour sorption isotherm. The second mechanism, capillary condensation was explained by Kelvin, who showed the vapour pressure above a curved liquid surface to be related to surface

curvature and surface energy by the following equation :

$$P \approx P^S \exp\left(-\frac{\sigma VK}{RT}\right) \quad (4)$$

where : P = vapour pressure, P^S = saturated vapour pressure, σ = surface energy, V = molar volume of liquid, K = total surface curvature, R = universal gas constant, and T = absolute temperature.

Application of equation (4) to a porous medium requires the introduction of a



pore model, which permits to relate the curvature K to the pore radius r . The most frequent choice is that of a cylindrical pore with a spherical liquid surface, leading to :

$$K = 2/(r-t) \quad (5)$$

where t is the thickness of the physically adsorbed layer. Note that it is more correct to use only half of the curvature given by equ. (5) in case of adsorption.

Introduction of the corresponding values for water in equations (4) and (5) shows that at 90% RH all pores with radii smaller than 10 nm are filled with liquid water; at 40% RH the critical pore radius has dropped to 1.2 nm.

Several mechanisms contribute to the flow of moisture, owing to the presence of several phases : liquid, vapour, and adsorbate.

Liquid flow is sufficiently slow to be purely laminar. Not even convective effects caused by the continuously varying cross section need to be taken into account. The following equation for the permeability coefficient λ^L can be derived for a porous medium :

$$\lambda^L = \frac{eRT}{36\nu vH} \frac{\langle r^4 \rangle_f}{\langle r^2 \rangle_a} \quad (6)$$

where : e = porosity, ν = kinematic viscosity, v = molar volume of the liquid, $\langle \rangle_f$ = average to be taken over the pores filled with liquid, $\langle \rangle_a$ = average over all pores, and r = pore radius.

Equation (6) is subject to the limitation that the pore-size distribution be uniform in space. In that case, there is a sufficient number of paths around obstacles such as wide pores which contain only vapour. The correction for the somewhat longer flow path in this case is fully accounted for in equ. (6) (see Matheron /8/).

Vapour flow in HCP is not diffusive as is generally assumed. The pore diameters in HCP are small in comparison with the mean free path of water vapour in air, so that molecule-wall interactions dominate molecule-molecule interaction. This type of gas transport is commonly termed "Knudsen diffusion", although the word diffusion is quite misleading. The permeability coefficient for vapour transport λ^V for a porous medium is given by the following equation :

$$\lambda^V = \frac{4}{27} \frac{e p^S \bar{M} \bar{c}}{RT} \frac{\langle (r-t)^3 \rangle_e}{\langle r^2 \rangle_a} \quad (7)$$

where : M = molar mass, \bar{c} = average thermal velocity, and $\langle \rangle_e$ = average to be taken over "empty" pores, i.e. pores not filled by capillary condensate.

The third mechanism flow of the adsorbed layer, will not be considered in this paper. At this time there is not enough reliable information on this phenomenon. There are indications that the mechanism is only of secondary importance in HCP; the heat of adsorption of some 75 kJ/mole corresponds to a surface diffusivity of only $\pm 10^{-8}$ cm²/s. Radjy /9/ showed that flow of the adsorbed layer is negligible under these circumstances. Further support can be found in the generally observed decrease of the permeability between 0 and 40 % RH; if surface flow would have a major contribution an increase in this interval would have to be expected.

3. A MODEL FOR MOISTURE FLOW IN HCP

Figure 2 shows a model which was developed to represent the porous structure of

HCP. The main feature of the model is the assumption that HCP is not a homogeneous porous system, but it is composed of different pore systems. The finest pores are the gel pores within the hydration products. They have been determined to be within a narrow band situated around 4 nm. In the microstructure of HCP these clusters are separated by coarser pores. It is supposed that the second and coarser pore system is centered around an average value of 20 nm. The exact meaning of this bi-porous system will be discussed in comparison with microstructural data elsewhere.

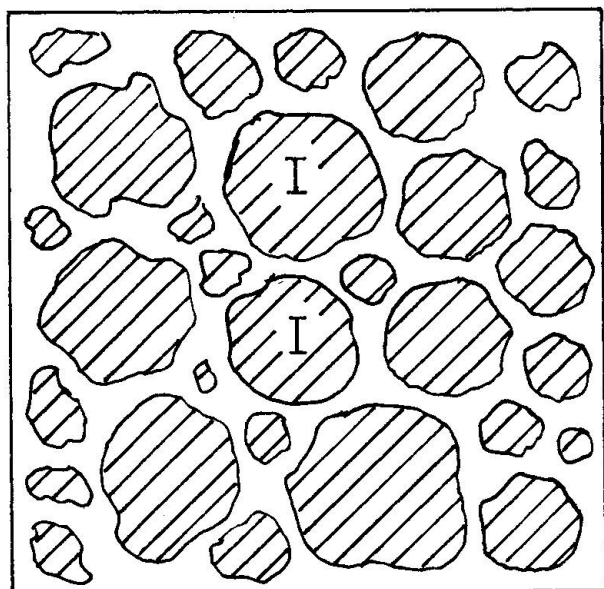


Fig. 2 Model microstructure of HCP.

The permeability coefficient of HCP is much lower than the values predicted by equ. (6) for liquid flow, even when a reasonable increase of viscosity in the narrow pores is allowed for. The model explains this by the interruption of the liquid flow paths. Vapour flow only is possible in the inter-cluster space, unless the porous system is fully saturated.

The model explains the sharp increase of the permeability coefficient at about 80% RH by capillary condensation and ensuing liquid flow inside the clusters (regions I, fig. 2). At low humidities these regions are obstacles; their small pores permit only a small amount of vapour flow. Capillary condensation at high relative humidity, however,

frees the way for low-resistance liquid flow; thus all regions with high resistance are systematically converted into regions with low resistance.

Remark that the model is quite insensitive to the precise flow resistance in regions I; these regions act either as obstacles (low RH) or as short circuits (high RH) for the vapour flow through the porous material. The flow mechanisms in the inter-cluster space are much better known; the water in these large pores behaves "normally", and flow of the adsorbed surface layer is definitely negligible. The global flow is dominated by the transport through the intercluster space and the mechanisms within the clusters only modify this transport phenomenon.

As an illustration of the model we calculated the moisture capacity and permeability coefficient of HCP on the basis of the following data :

- Volume concentrations: finely porous material (clusters) 61%, intercluster space 39%.
- Total porosity: 28%, of which 72% is the finely porous material, and 28% is the coarser one.

The pore size distribution of this model material is shown in Figure 3.

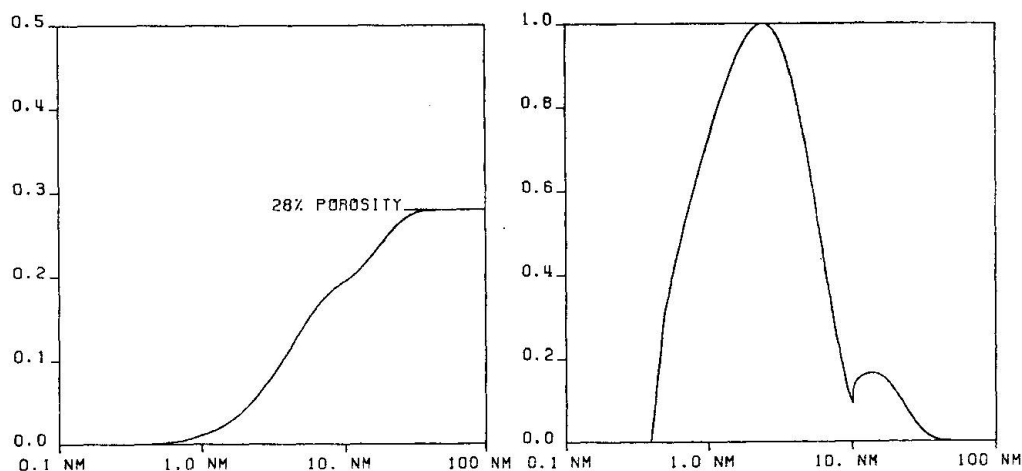


Fig. 3 Cumulative and differential pore size distribution.

Note that this distribution has been generated in a computer by simulating two Weibull distribution functions for the bi-porous model material. By changing the structural parameters, different pore size distributions can be obtained. Permeabilities, capacities, and sorption isotherms were calculated for this model material in a 2D random pore network. For the calculation of the permeability coefficient we generated the random structure shown in Figure 4 (circles represent clusters of hydration products). This structure can be analysed with the software developed for "numerical concrete" /10/ after proper modifications. The calculated permeability coefficient is shown in Figure 5.

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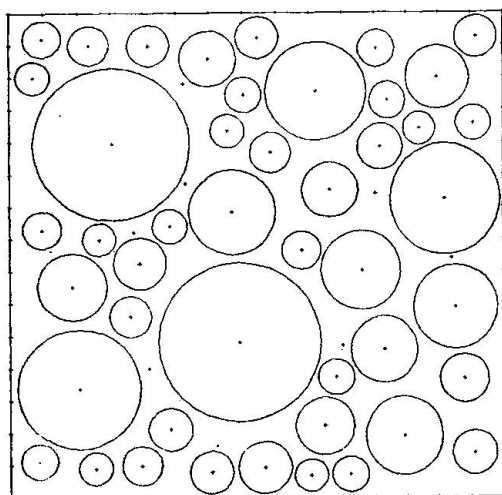


Fig. 4 Numerical model of microstructure.

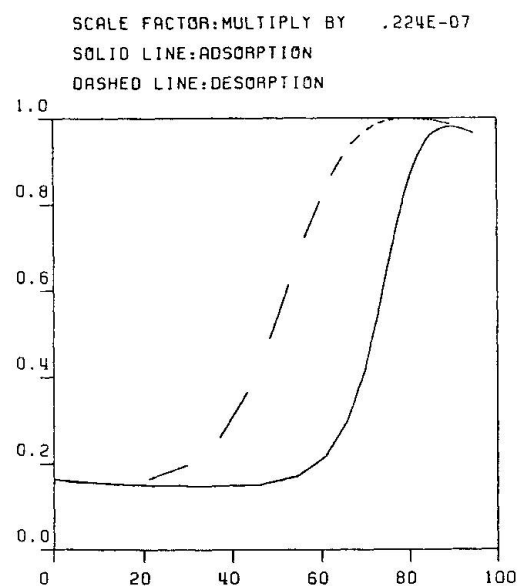


Fig. 5 Permeability coefficient.

The precise numerical values depend strongly on the chosen pore-size distribution of course. The maximum value is somewhat high as compared with experimental values. Better agreement could be reached in modifying the pore size distribution. This is not our aim, however, at the moment. The general validity of the model has been shown. The same applies for the strong increase of the permeability which lies usually around 70% for desorption. The calculated sorption isotherm and moisture capacity are shown in Figures 6 and 7.

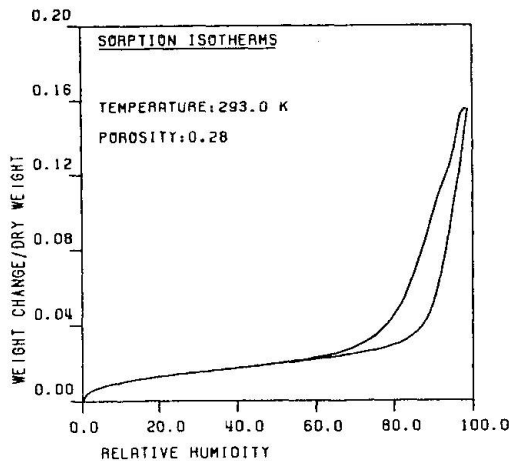


Fig. 6 Calculated sorption isotherm.

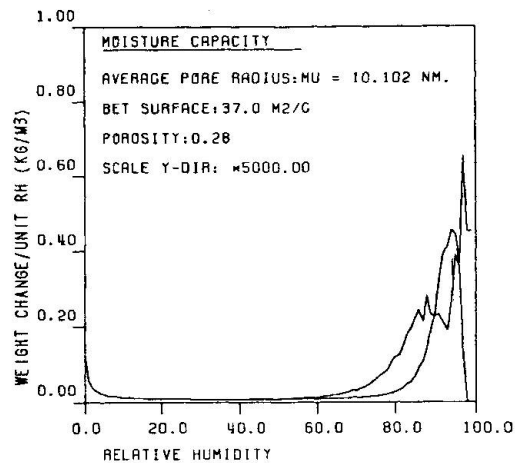


Fig. 7 Moisture capacity.

The calculated isotherms differ markedly from experimental results (see e.g. the curves published by Hundt /6/, Figure 8). Particularly desorption isotherms are much less convex at room temperature. This may be partly due to the so-called ink-bottle effect /5/; Figure 9 shows an isotherm in which this effect is taken into account. Surprisingly the calculated curves correspond much more closely to those measured at 70°; this cannot be explained on thermodynamical grounds, nor by the coarsening of cement gel, which is rather limited at this temperature.

In accordance with equation (1), the high capacity around 80% RH strongly delays drying; the phenomenon is analogous to the effect of phase changes in heat transfer problems. As a consequence, drying is predicted to be more rapid at, say, 70% RH than at 80% RH, which is in contradiction with experimental observation. Clearly, more research is necessary before a satisfactory description of the desorption isotherm is possible.

4. TRANSPORT PHENOMENA IN CONCRETE

The functions $C(H)$ and $\lambda(H)$ discussed in the previous chapter were used to simulate the drying of concrete with the methods of numerical concrete /10/ and /11/. The addition of aggregates has several consequences. The most obvious one is a reduction of permeability and moisture capacity. A second effect is the

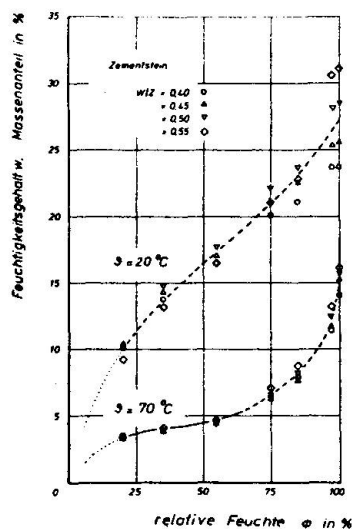


Fig. 8 Measured desorption isotherms /6/.

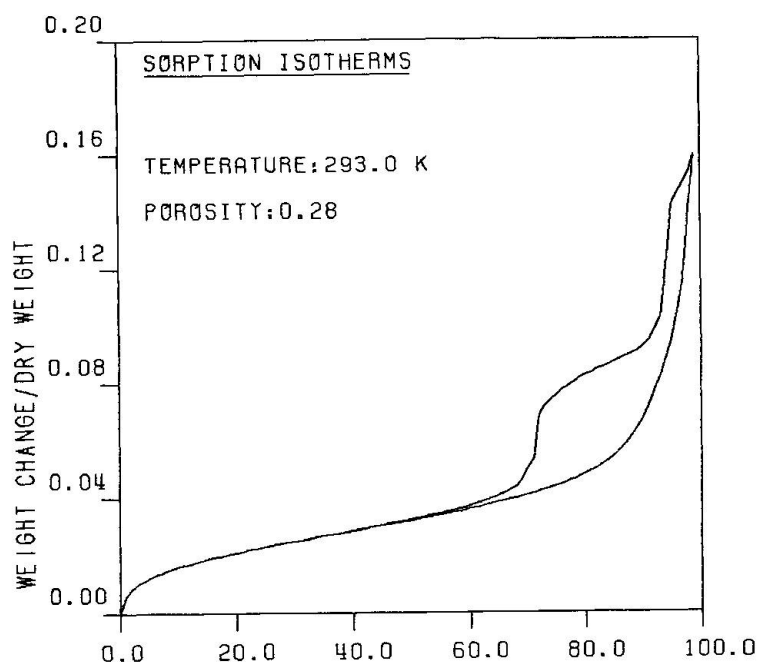


Fig. 9 Sorption isotherm, ink-bottle effects included.

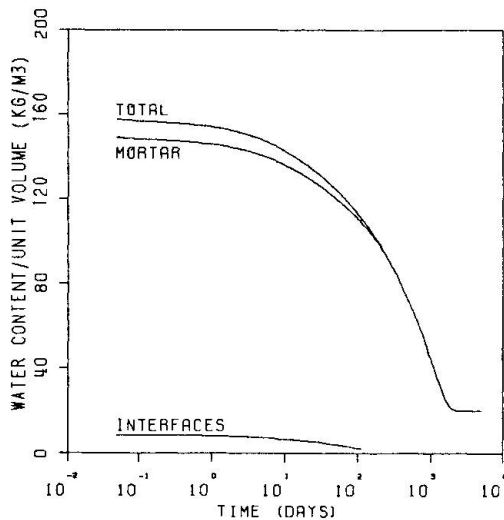
introduction of highly process interfaces between matrix and aggregates, which facilitates flow at high humidities. Finally, voids are introduced in the material during mixing. The water contained in these voids and interfaces must flow away through the dense matrix before a true equilibrium state has been reached. As the quantities of excess water are usually not well known this must falsify the determination of diffusion coefficients from weight loss curves.

Theoretical aspects of the analysis are discussed in the invited lecture of Wittmann and Roelfstra /12/ for this conference.

Bazant /2/ found a relation for the humidity dependence of the diffusion coefficient by pure data fitting. In the present paper an attempt is made to explain the physical background of this relation. In this way it will be possible to formulate generally valid material laws. These laws can be used in a numerical analysis to predict moisture movement in real concrete structures such as large span bridges, large industrial floors, and prestressed concrete pressure vessels.

For the sake of the numerical analysis, all extra porosity of the matrix was lumped in the interfaces around the large aggregates. Figure 10 shows the weight loss curve obtained with the following data :

- Volume concentration of the big aggregates: 43%
- Total water content in interfacial regions (including voids in the matrix) at 100% RH: 1800 kg/m^3 relative to the interfacial volume.
- Permeability of the interfaces: $0.8 \cdot 10^{-5} \text{ kg/m/s}$.
- Thickness of the interfaces: 50μ .
- Zero capacity and permeability of the interfaces below 98% RH.



The sharp kink in the curve is due to the convex form of the simulated desorption isotherm mentioned above which is usually not observed experimentally. With a refined model this can be overcome.

Fig. 10 Weight loss curve of concrete, drying from 100% to 50% RH.

5. CONCLUSIONS

1. A bi-porous model material has been developed in order to describe the influence of moisture content on permeability coefficient and moisture capacity. This model can be adjusted to describe different transport mechanisms in HCP and concrete realistically.
2. The influence of temperature on moisture diffusion is predicted.
3. Based on this concept material parameters for diffusion in different types of concrete and under arbitrary climatic conditions can be indicated and used in computerized structural analysis.

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