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The Diffusion of Carbon Dioxide and Oxygen in Aerated Concrete

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SUMMARY

A knowledge of the diffusion coefficients for CO_2 and O_2 is of considerable importance for a quantitative assessment of the carbonation of concrete and of the corrosion of the reinforcements. CO_2 induces the carbonation of concrete leading to a suppression of the immunity of the reinforcement embedded in an uncarbonated concrete. The phenomenon of corrosion of the reinforcement then depends, among other parameters, on the ready availability of oxygen; consequently, on its rate of diffusion. We have developed an equipment, which allows the simultaneous determination of the diffusion coefficients of the two gases as a function of the relative humidity. Measurements have been carried out with a "model" material, i.e. aerated concrete. The method chosen gives reliable results. The influence of gas solubility and pore size distribution is discussed.

1. INTRODUCTION

It is wellknown that carbonation has a major influence on durability of concrete structures. The actual rate of carbonation depends on number of different parameters which may be classified into two categories :

- Parameters describing the quality of the material such as concrete composition, cement content, water/cement ratio, etc.

- Environmental conditions such as CO_2 content of the air, presence of other agressive gases or liquids, frost action, etc.

The complex interaction of all these different processes cannot be studied easily. In a first attempt Houst, Roelfstra and Wittmann (Ref. 1) have tried to apply modern numerical simulation methods to this problem. This work has been persued and Brieger and Wittmann will report on further results of numerical simulation methods during this meeting (Ref. 2). Numerical simulation methods are nowadays very powerfull tools. All obtained results, however, depend on the material parameters which have to be introduced. So far only few experiments have been carried out to study the influence of moisture content on gas diffusion through porous materials. It is the aim of this contribution to outline an experimental method which allows us to study gas diffusion through cementitious materials at variable moisture content. So far tests have been run on aerated concrete. This is a more porous and more stable structure than hardened cement paste. For this reason it has been chosen as a model material.

2. DIFFUSION OF GASES THROUGH POROUS SYSTEMS

Phenomenologically diffusion of gases through porous systems can be described by Fick's law :

$$J = -D \frac{dc}{dx}$$

In this equation J stands for the flux of the gas and dc/dx describes the gradient of the concentration. D is the coefficient of diffusion. In reality diffusion processes are complex and we have to distinguish at least three different mechanisms (Ref. 3) :

- In large pores normal gas diffusion is observed.

- As soon as the pore dimensions get comparable to the mean free path length of the gas the diffusion process is disturbed and we have the so-called Knudsen diffusion.

- In addition to the two just mentioned mechanisms, adsorbed gases can be transported through diffusion along the internal surface of a porous material.

The contribution of Kamp and Wittmann describes the mechanisms of moisture diffusion through porous materials in more detail (Ref. 4). Schwiete, Ludwig and Zagar have developed an apparatus to measure the diffusion coefficient of oxygen and carbon dioxide through porous ceramic materials (Ref. 5, 6). Later Därr and Ludwig used similar apparatus to measure the permeable porosity of materials (Ref. 7). Engelfried has then developed a method which allowed hit to measure the diffusion of CO_2 through protective films applied on porous substrates (Ref. 8). More recently Hurling determined the influence of water/cement ratio and cement content as well as storing conditions on the diffusion coefficient of oxygen through concrete (Ref. 9). None of the just mentioned methods allows us to follow systematically the influence of variable moisture contents on diffusion coefficient.

3. EXPERIMENTAL METHOD AND RESULTS

The experimental set up is schematically shown in Figure 1. The heart of this apparatus is a measuring cell which is divided into two rooms by the porous disc. Before the porous discs are placed in the measuring cell, they are equilibrated with a given relative humidity. Once built into the measuring cell, nitrogen coming from flask 2 in Figure 1 is first of all equilibrated with a given moisture content with the by-pass system (valves 6 and 5 in Fig. 1) and then streaming successively through the upper and the lower rooms of the measuring cell. The moisture content of the streaming gas can be varied by the relative flow through valve 6 and valve 5. The proportion of gas streaming through valve 5 is water saturated in the flask 9. The flow of pure nitrogen is continued until no oxygen and no carbon dioxide can be measured any more in the lower room of the measuring cell. Then valves 4 and 20 are closed. From flask 1 a pre-conditioned gas mix is then entering first of all the moisture conditioning circuit. The moisture content is measured by a special gauge in the element 10. This pre-conditioned gas is only entering the upper part of the measuring cell. Oxygen and carbon dioxide migrate now through the porous disc into the lower part of the measuring cell. There the relative concentration is measured as function of time. Two typical examples are given in Figure 2. Figure 2 a) shows the time dependence of oxygen and carbon dioxide diffusion into the lower part of the measuring cell through a disc of aerated concrete which had been in equilibrium with a relative humidity of 0%. In Figure 2 b) similar results are shown and in this case a identical disc of aerated concrete has been used which had been in equilibrium with 86% relative humidity.

It has been shown elsewhere how a diffusion coefficient can be deduced from the curves shown in Figure 2 (Ref. 10).

4. DISCUSSION AND CONCLUSIONS

The diffusion coefficients obtained in this way are shown in Figure 3 as function of the relative humidity in which the porous discs of aerated concrete had been equilibrated. Aerated concrete is a material with a total porosity of around 80%. Most of this porosity is concentrated on very large spherical pores. In Figure 4, the pore size distribution as determined by Prim and Wittmann is shown (Ref. 11). Even at relative humidities which are close to 90% only relatively few pores are water filled. This can be clearly shown from the sorption isotherms which have been measured earlier by Houst, Alou and Wittmann (Fig. 5 and Ref. 12). Due to this fact, it is reasonable to assume that moisture content in this type of porous material has no significant influence on the diffusion coefficient. This hypothesis is verified by the experiments with oxygen diffusion as shown in Figure 3. The diffusion coefficient for carbon dioxide seems to go down substantially beyond 60% relative humidity. It is possible that in this region part of the carbon dioxide is dissolved in the capillary condensed water. This assumption, however, needs further verification.

The pore size distribution of hardened cement paste is very much finer (Ref. 13). A typical example of the influence of water/cement ratio on the pore size distribution of hardened cement paste is shown in Figure 6. In such a material a considerable percentage of the pores is water filled by capillary condensation at relative humidities above 70%. These water filled pores are of course blocked and they cannot contribute to the gas diffusion process. As a consequence we have to expect that in hardened cement paste the diffusion coefficient of gas will substantially decrease at higher relative humidities.

It has been shown that the experimental method to determine diffusion coefficients of gas through porous materials gives us reliable results. So far measurements have been carried out on aerated concrete exclusively which has been used as a model material. Experiments will be extended to cover hardened cement paste and mortar. These results will be introduced directly in the numerical models developed in our laboratory and presented during this meeting by Brieger and Wittmann. (Ref. 2)

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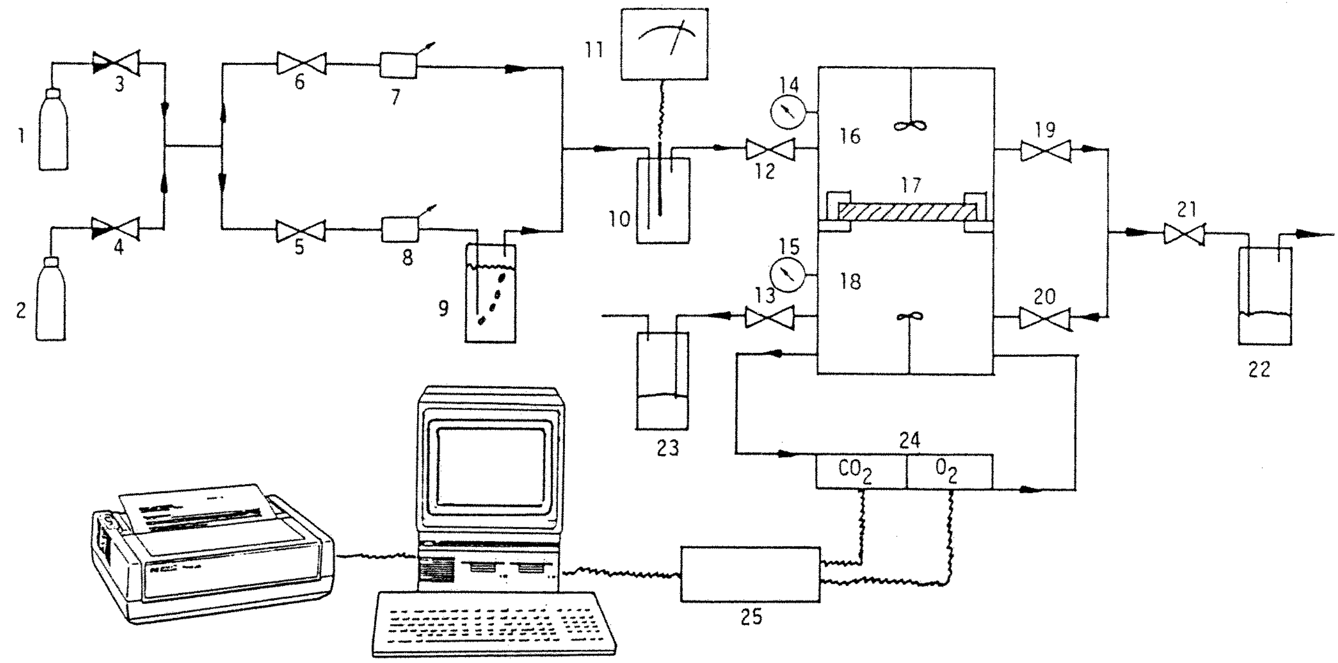

1 Gas : 78% N_2 - 20% O_2 - 2% CO_2
2 Gas : 100% N_2
3-6 valves
7-8 flowmeters
9 humidification to 100% relative humidity
10 measure RH of the mixte gases
11 RH indicator
12-13 valves
14-15 manometers
16 upper room
17 sample
18 lower room
19-21 valves
22-23 bottles to prevent introduction of ambiant air
24 gas analysers
25 data acquisition unit

Fig. 1: Schematic representation of the experimental set up.

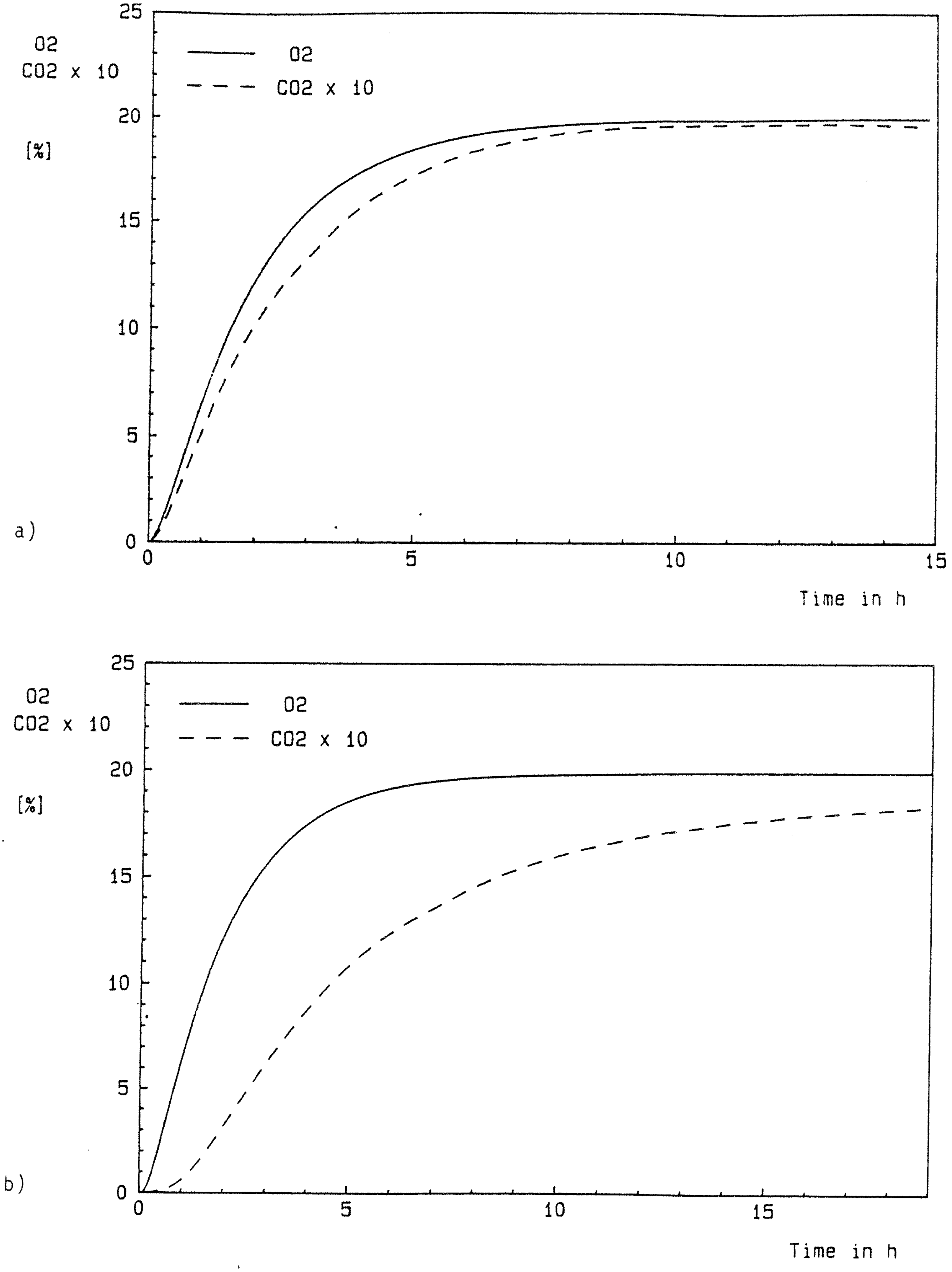

Fig. 2: O_2 and CO_2 concentration as function of time on aerated conerete type H and a RH of a) 0% and b) 86%.

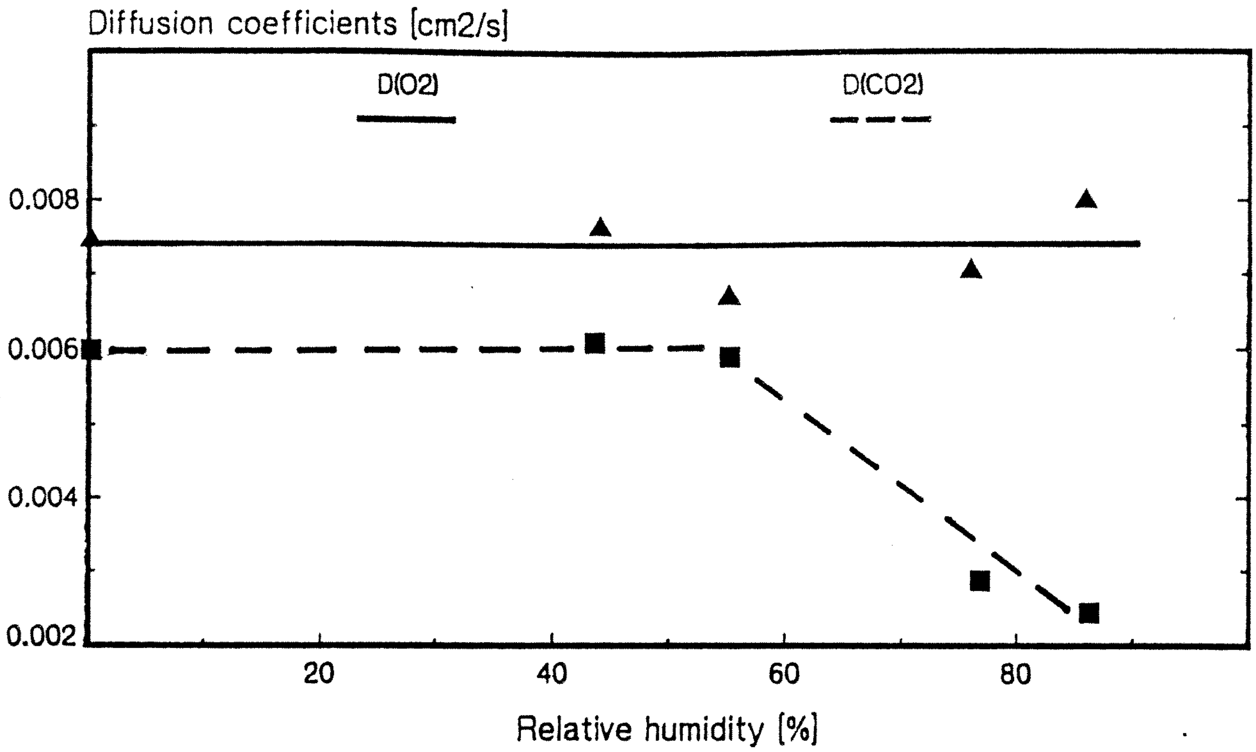

Fig. 3: Diffusion coefficient as a function of relative humidity.

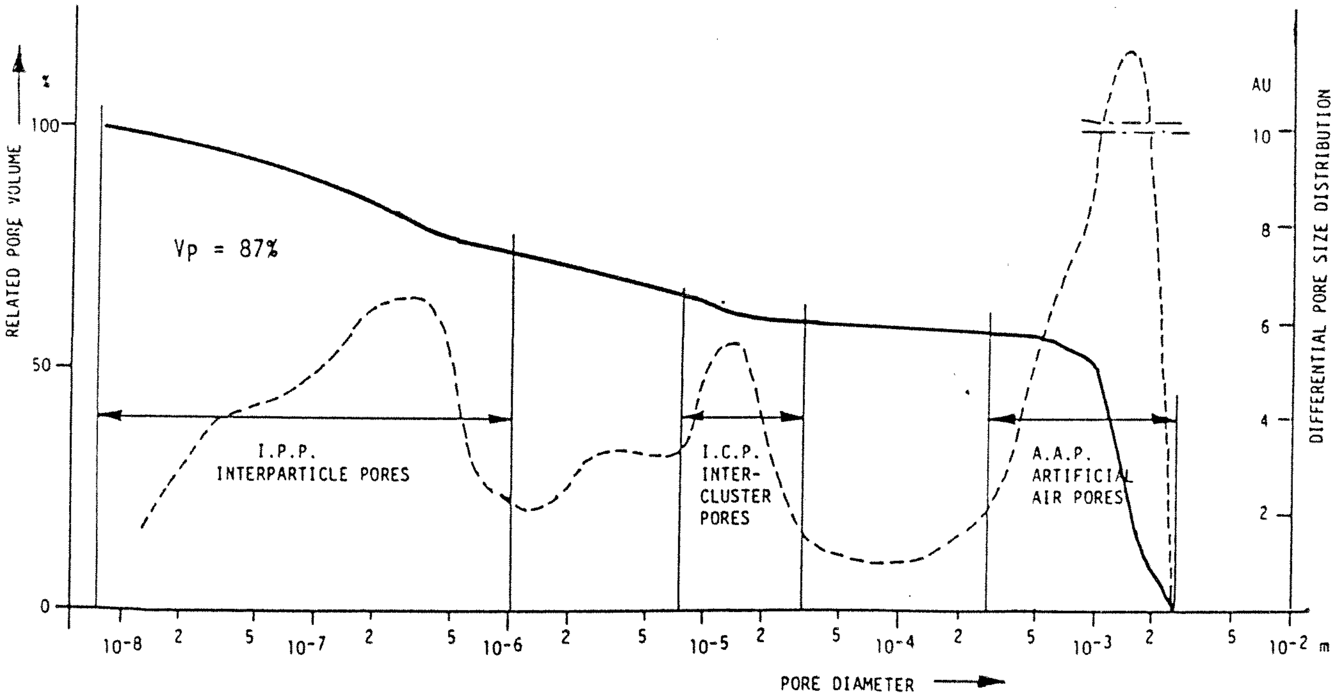

Fig. 4: Integral and differential pore size distribution compiled on the basis of results of different test methods for aerated concrete, type L (Low compressure strength) (Ref. 13).

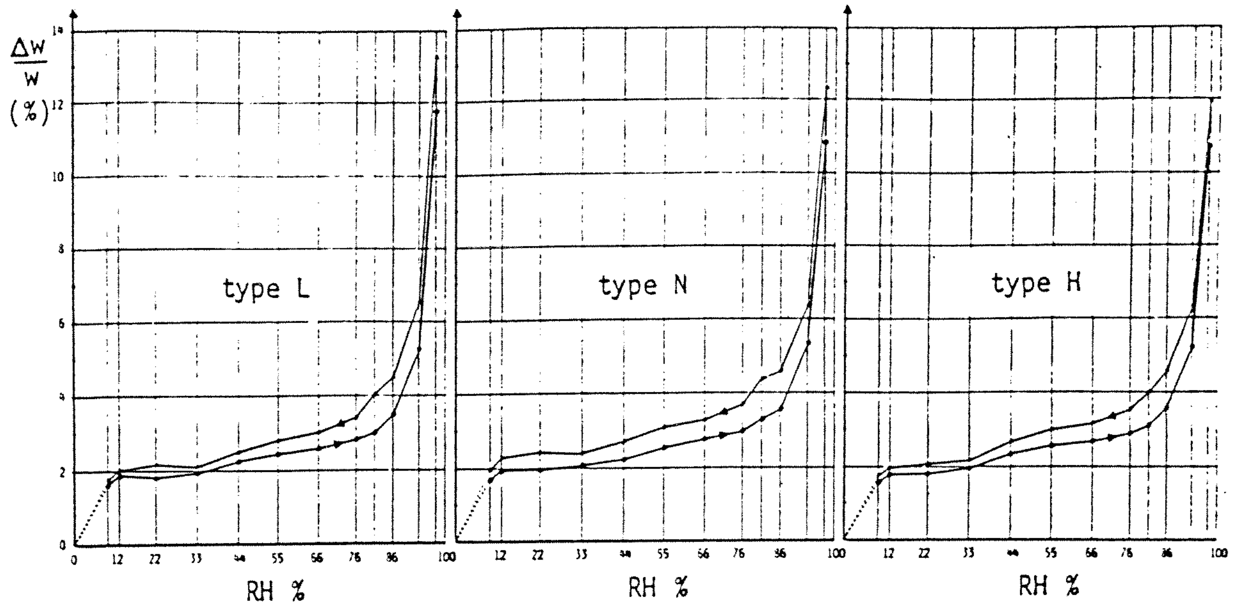

Fig. 5: Sorption isotherms of aerated concrete. The sorption and desorption branches are marked with an arrow (ΔW/W : variation of mass %) (Ref. 12).

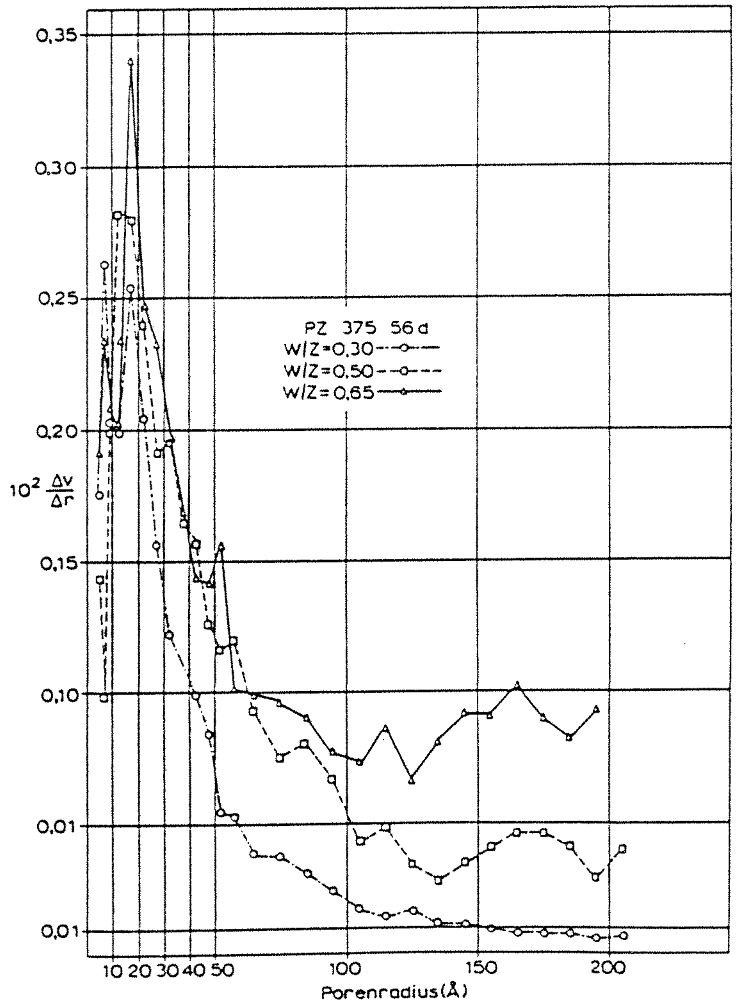

Fig. 6: Differential pore size distribution compiled on the basis of water adsorption isotherms for cement pastes with three different water/cement rations (Ref. 13).