Pergamon

Cement and Concrete Research 32 (2002) 1497–1504

CEMENT AND CONCRETE RESEARCH

Permeability and diffusivity of concrete as function of temperature

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Received 19 December 2001; accepted 3 April 2002

Abstract

Tests have been carried out on 11 types of concrete—ranging from normal-strength to high-strength concrete, to polymer-modified and self-compacting concrete—establishing permeability and diffusivity of concrete as a function of temperature between 20 and 80 °C. The results show a considerable increase of these properties with temperature. Theoretical prediction on the basis of thermodynamics shows reasonable to good agreement between theory and experiment. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Concrete; Permeability; Diffusion; Temperature; High-performance concrete

1. Introduction

Transport processes in porous materials like concrete have been investigated for several decades and numerous publications show a high level of knowledge (e.g., Refs. [1–4,6,8]). However, the correlation between transport coefficients and temperature has received only little attention. This is surprising since water transport processes are often accompanied by temperature variation. The investigation was initiated by the design and construction of hot water storage of solar energy. Water is heated up by solar collectors and pumped into a buried storage vessel. The water has a maximum temperature of 95 °C; the stored energy should be saved for the colder season of the year. The question arose about the permeability and diffusivity of concrete at temperatures between 20 and 80 °C. From the thermodynamic theory, it should be expected that the transport properties increase with temperature. This will be shown in one of the sections of the paper. This paper presents the testing methods and results of the investigation as well as some theoretical considerations.

2. Permeability

2.1. Measuring device

Since test procedures for the measuring of the permeability of water are not standardized in Germany, a test device was designed and constructed. The test device consists of the test cell and the control unit. The test cell consists of a housing made of aluminium, a contact pressure tube made of rubber, a seal pressure sleeve made of polyurethane as well as a measuring capillary with a scaling at the inlet and the outlet end (Fig. 1). The lateral pressure on the specimen amounts to 1.5 MPa.

The testing set-up has originally been designed by Graef and Grube [2,3] for isothermal condition of 20 °C. The system has been adjusted to an option up to 80 °C with special attention on the sealing materials. Up to the required temperature of about 80 °C, all parts of the system work without difficulties. To simulate higher temperatures of 50 and 80 °C, the test cells were put into a large climatic chamber with a temperature tolerance of ± 1 °C.

2.2. Test specimens

Until the beginning of the test, the test specimens were stored in water. Water storage is necessary to receive comparable results because the permeability coefficient extremely varies depending on the moisture content of the concrete; a rising moisture content, for example, causes a

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PII: S0008-8846(02)00812-8

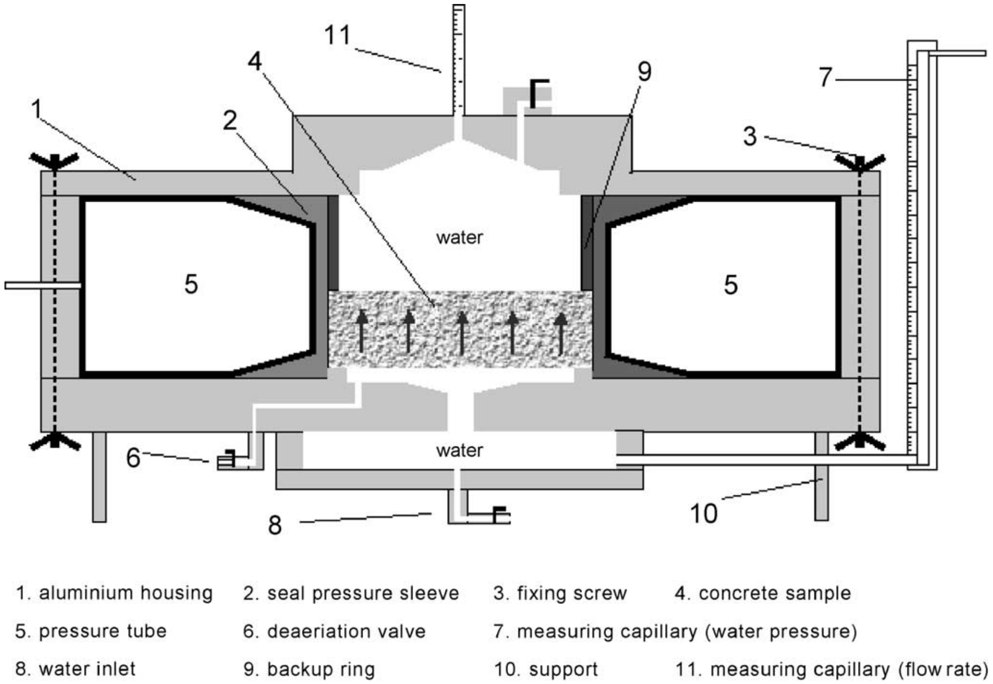

Fig. 1. Schematic illustration of the test cell for the determination of the permeability coefficient of concrete.

decline of the permeability coefficient. The test specimens themselves are circular slices with a diameter of 150 mm and a thickness of 30 mm, which were ground flat on both sides and sealed on the perimeter surface.

3. Diffusion

The diffusion tests followed standardized procedures according to DIN 52615. For these tests, the dry cup method as well as the wet cup method were used.

3.1. Dry cup method

In the dry cup method, the diffusion coefficient D and the water vapour resistance μ are determined between 0% and 50% relative humidity. The salt solutions that were used for constant humidity are specified in Table 1.

3.2. Wet cup method

In the wet cup method, the diffusion coefficient D and the water vapour resistance μ are determined between 50% and 93–100% relative humidity. Therefore, depending on temperature and humidity, various salt solutions or drying agents come into operation (see Table 1).

3.3. Measuring device

The actual measuring device consists of the small cup, on which the concrete samples are attached, the desiccators and the climatic chamber. The cups have a diameter of 100 mm, a height of approximately 10 mm and consist either of aluminium or stainless steel. The desiccators have an internal diameter of 300 mm. By installing several floors, the desiccator could be used to full capacity and approximately 12 diffusion samples per desiccator could be tested at the same time (see Fig. 2).

To ensure a uniform moisture distribution, a ventilator was attached to the upper tubes. This ventilator assured the air velocity of $0.02 < v_L < 0.3$ (m/s) given in DIN 52615. To simulate temperatures of 50–80 °C, the desiccators were put into a large climatic chamber with a temperature tolerance of ± 1 °C. Dry cup method means that the salt solution in the specimen cup guarantied constant humidity, which is lower than in the desiccator that had a humidity of about 50%. The

Table 1
Salt solutions used in diffusion tests for a constant humidity

Relative humidity range [%]	Temperature [°C]		
	20	50	80
0–3	Silica gel/phosphorus pentoxide (P_2O_5)	Phosphorus pentoxide (P_2O_5)	Phosphorus pentoxide (P_2O_5)
50	Sodium dichromate ($Na_2Cr_2O_7 \cdot H_2O$)	Sodium bromide (NaBr)	Sodium bromide (NaBr)
93–100	Ammonium dihydrogen phosphate or H_2O	Ammonium dihydrogen phosphate or H_2O	Ammonium dihydrogen phosphate or H_2O

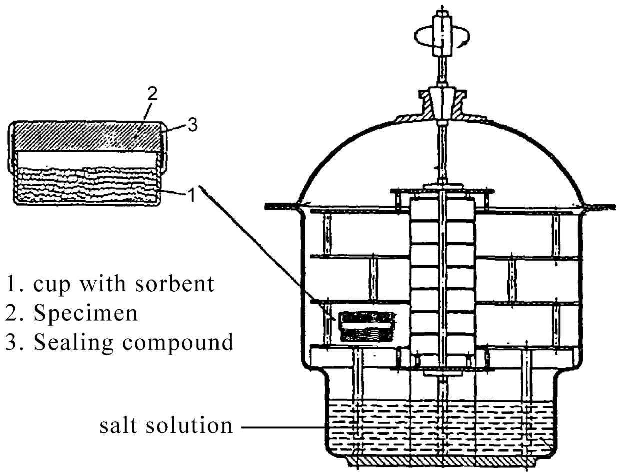

Fig. 2. Illustration of the test set-up for the determination of the water vapour diffusion coefficient of concrete in the dry cup method and the wet cup method.

wet cup method is the opposite in the sense that the moisture in the cup was higher than in the desiccator. In the first case, moisture diffuses through the specimen into the cup, whereas in the second case, moisture flows from the cup into the desiccator. The arrangement consisting of specimen, cup and sealing and was weighted in regular intervals of 4 days. From the weight changes, the moisture difference and the specimen thickness, the coefficient of diffusion has been calculated.

3.4. Test specimens

Test specimens with a diameter of 100 mm and a thickness of approximately 20–25 mm were tested. That way, all points stipulated in DIN 52615 are accommodated. All test specimens were sawn after about 28 days and were stored in the desiccator at 50% relative humidity for another 3–5 weeks until testing. During the installation, the samples were in moisture equilibrium.

4. Experimental investigations and results of high-performance concrete (HPC)

4.1. Types of concrete

Various types of concrete mixtures have been produced and tested. The main goal was the improvement of impermeability. A reference concrete, which complied with a conventional waterproof concrete according to DIN 1045, was used as a reference concrete. The specific compositions of the concretes are given in Table 2.

Table 3 shows some specific properties of fresh and hardened concrete. Other properties are described in Ref. [5]. For most tasks, three specimens were used and the scatter of the results was rather small.

4.1.1. Reference concrete B 35

This concrete was used as a reference for comparison with the other types of HPC.

4.1.2. Copolymerisate concrete

The copolymerisate dispersion that was used consists of styrene and acryl acid ester. It has a good compatibility with concrete and should improve workability. Its use opens the possibility of the reduction of water/cement (w/c) ratio. Through the addition of copolymerisate, properties like compressive and tensile strength and durability should improve. By addition of water, the copolymerisate particles swell and reduce the pore space without producing an inner pressure. Two mixtures with different quantities of copolymerisate (30 and 45 kg/m^3) were investigated.

4.1.3. HPC

A standard mixture that was optimized under the aspects of impermeability, workability and cost in the course of the tests was the starting point. As in other types of concrete, several components (type of cement, additives, aggregate)

Table 2
Composition of the concrete mixtures used

Mix	w/c ratio	Cement [kg/m^3]	Aggregate [kg/m^3]	Additive and admixture
Reference	0.45	330 (CEM I 32.5 R)	1788	FA 30 kg
Copoly-30	0.45	330 (CEM I 32.5 R)	1790	Copolymerisat—dispersion 30 kg, FA 30 kg
Copoly-45	0.45	330 (CEM I 32.5 R)	1788	Copolymerisat—dispersion 45 kg, FA 30 kg
HPC no. 35	0.33	400 (CEM I 32.5 R)	1896	MS 60 kg, FM 30, 2.8%
HPC no. 36	0.33	400 (CEM I 32.5 R)	1895	MS 60 kg, FM 30, 2.6%
HPC no. 37	0.33	400 (CEM I 32.5 NW/HS)	1895	MS 60 kg, FM 30 2.6%
HPC no. 40	0.37	360 (CEM II/A-L 32.5 R)	1948	MS 40 kg, FM 30 2.0%, FA 40 kg
HPC no. 41	0.37	360 (CEM II/A-L 32.5 R)	1956	MS 40 kg, FM 30 2.5%, FA 40 kg
RPC	Premix with: 700 kg of CEM, 225 kg of silica fume, 990 kg of sand, 210 kg of quartz flour, 45 kg of plasticiser + 195 kg of water + 200 kg of steel fibers			
SCC 1	0.45	320 (CEM II/A-L 32.5 R)	1735	4 kg of FM/BV 375, UWC 0.30 kg, FA 180 kg
SCC 2	0.41	370 (CEM II/A-L 32.5 R)	1691	5.5 kg of FM/BV 375, UWC 0.30 kg, FA 170 kg

FM: superplasticiser; BV: plasticiser; MS: microsilica; FA: flyash; UWC: underwater compound.

Table 3
Properties of the concrete mixtures used

Mix	Density [kg/m^3]	Air content in fresh concrete [%]	Compressive strength after 28 days [MPa]	Bending strength (four-point) after 28 days [MPa]
Reference	2.41	1.4	45	5.8
Copoly-30	2.34	1.8	44	6.0
Copoly-45	2.34	0.9	51	5.9
HPC no. 35	2.47	1.4	89	9.7
HPC no. 36	2.49	0.7	87	8.8
HPC no. 37	2.51	0.6	87	8.1
HPC no. 40	2.46	1.3	86	7.9
HPC no. 41	2.45	1.0	93	8.1
RPC	2.48	–	190	19.8
SCC 1	2.37	0.7	56	5.9
SCC 2	2.36	1.3	71	7.4

were varied during the tests. Altogether, five different mixtures of HPC were produced for the test program.

4.1.4. Reactive powder concrete (RPC)

RPC is a concrete with steel fibers, which is characterized by an extremely high strength and very low permeability. With a maximum grain size of approximately 250 μm, a cement content of about 700–800 kg/m^3 and a w/c ratio of <0.20, it differs from conventional types of concrete in regard to the composition. The production is completely different from conventional types of concrete. RPC cannot be compacted by vibration and has a honey-like consistency at the end of the mixing.

4.1.5. Self-compacting concrete (SCC)

SCC is very special with respect to fluidity, self-leveling ability and ventilation. It does not need compaction by vibration. The composition is different from normal concrete due to a reduced content of coarse aggregate, a lower w/c ratio and an extreme low viscosity caused by highly effective water-reducing agents (e.g., polycarboxylates).

4.2. Permeability measured

The following diagrams (Figs. 3 and 4) show the dependency of water permeability on temperature. In all diagrams, the permeability coefficient k is given as function of temperature.

4.2.1. Copolymerisate concrete

Fig. 3 clearly shows the increase of permeability depending on temperature. Depending on the type of concrete, the increase of permeability varies. The permeability rises approximately by 10–14% between 20 and 50 °C in the reference concrete and concrete with 30 kg/m^3 copolymerisate, and by approximately 7–11% between 50 and 80 °C.

In the same temperature range, however, the copolymerisate concrete with 45 kg/m^3 shows an increase of about 80%, but on a much lower level. The effect of the copolymerisate content on the permeability is clearly visible.

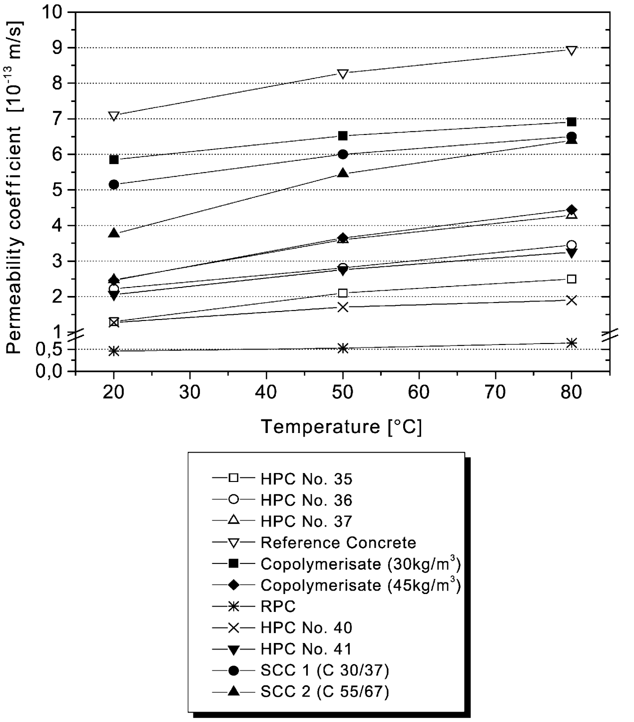

Fig. 3. Permeability coefficient of all tested types of concrete vs. temperature (after 1 h).

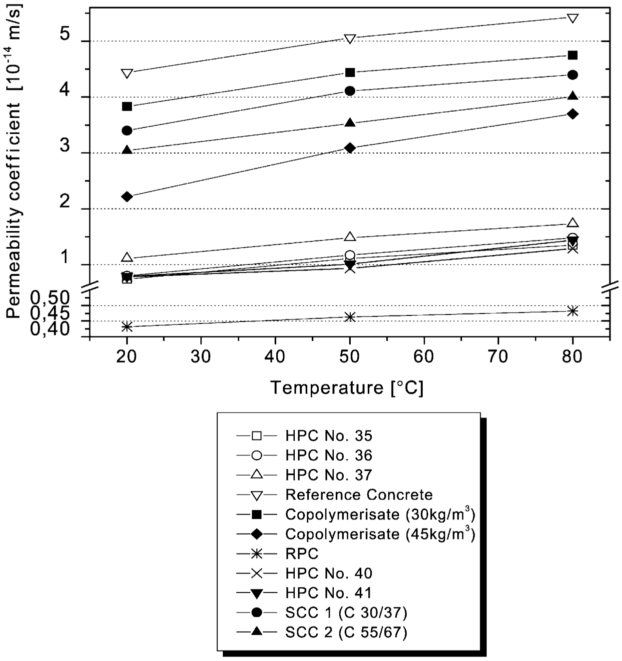

Fig. 4. Permeability coefficient of all tested types of concrete vs. temperature (after 48 h).

4.2.2. HPC

In Fig. 4, the permeability coefficients *k* of HPC and conventional concrete are compared with each other. In comparison to Fig. 3, the reduction of permeability is even more pronounced. Mixture nos. 35–37 have a permeability that is three to six times lower than that of the reference concrete at 20 °C, and a permeability that is still about two to four times lower at 80 °C, whereas the permeability difference within the series of HPCs is still considerable.

Like in Fig. 3, an increase of permeability as function of temperature can also be noticed for HPC. The percentage of the increase between 20 and 50 °C, as well as between 50 and 80 °C, is higher than for conventional concrete and copolymerisate concrete.

In Fig. 3, the coefficient of permeability after 1 h of all tested types of concrete is represented in one diagram. The group of SCCs roughly behaves according to the selected strength category. Nevertheless, SCC 1 lies distinctly below the reference concrete of the same strength category (B 35) after 1 h.

The permeability coefficient of SCC 1 is approximately 38% below that of standard concrete. The rise in permeability between 20 and 80 °C is nearly 30%. Like for other types of concrete, an increase of permeability depending on temperature can also be seen for RPC.

Since the permeability depends strongly on time, the permeability coefficient after 48 h is given in Fig. 4. This reflects the fact that the permeability coefficient dropped to about a tenth of the value determined after 1 h. The dependency on temperature remains. During the tests, it turned out that a reduction of the flow rate to about one eighth of the initial flow rate was already reached in the first hour. Even after 48 h, RPC still has a substantially lower permeability than the high-performance types of concrete. However, a temperature dependence does likewise exist in RPC, which amounts to about 10–20% between 20 and 80 °C.

4.3. Diffusion

4.3.1. Wet cup

In Fig. 5, the diffusion coefficients of the tested concretes are plotted. Like for permeability, the diffusion coefficients seem also to depend on temperature. The concrete with 30 kg/m^3 copolymerisate is approximately 15% above the reference concrete. The concrete with 45 kg/m^3 copolymerisate, in contrast, is only a little lower. However, the increase of copolymerisate from 30 to 45 kg/m^3 is not considered as significant. Between 20 and 80 °C, the diffusion coefficient shows an increase of 17–25%, whereupon the rates of increase in the three mentioned types of concrete are about the same. However, in comparison with that, the difference between the HPC and all types of copolymerisate concrete and the reference concrete is evident. A reduction of the diffusion coefficient to about one third can be observed here. Within the group of HPC, in regard to tightness, the same order can be recognized as in the permeability tests. Mixture no. 40 shows the lowest while mixture no. 37 shows the highest diffusion coefficient; a ratio of about 1:1.5 exists between the two mixtures that were just mentioned. A dependence of the diffusion coefficient on temperature is apparent in this group, too. The increase between 20 and 80 °C is substantially higher than for all types of copolymerisate concrete and reference concrete.

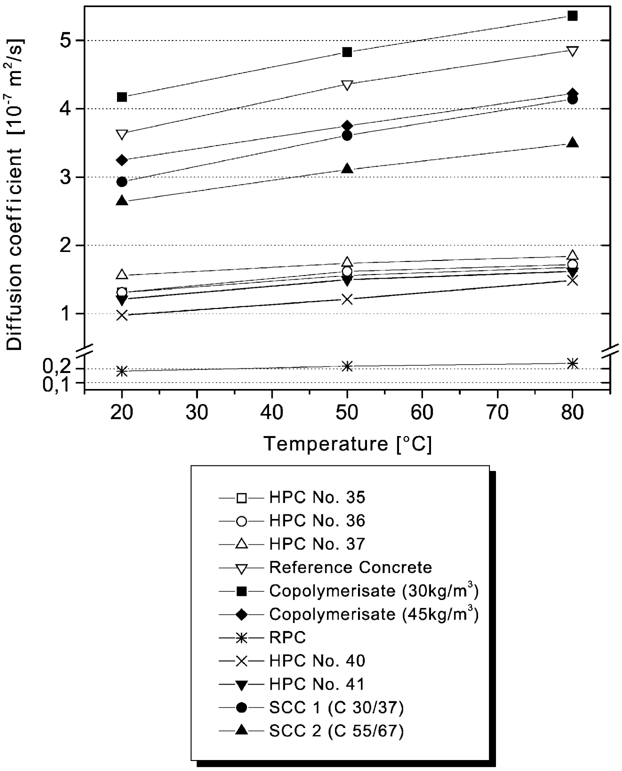

Fig. 5. Diffusion coefficient of all tested concretes vs. temperature (wet cup method).

RPC lies under the break in the *y*-axis and is thus about one order of magnitude below all types of HPC. Concerning RPC, samples with and without fibers were tested. However, no measurable difference could be determined with regard to fibers. SCC 1 turns out to have the same temperature dependence that has already been observed in the other types of concrete. The rise amounts to approximately 30% between 20 and 80 °C. Deviations because of the new generation of high water-reducing agents, which are used in SCC, cannot be noticed. Even for SCC 2, exceptional qualities in regard to temperature behaviour and the increase of the diffusion coefficient cannot be noticed. According to the strength category (B 65), the diffusion coefficient lies between the values of the reference concrete and the HPC. SCC 2 has a nearly linear increase of the diffusion coefficient between 20 and 80 °C. The rise in this temperature range is approximately 35%.

4.3.2. Dry cup testing

In the dry cup method, the division into various tightness categories (conventional, copolymerisate, HPC and RPC) is possible as well (see Fig. 6). According to their strength category, the SCCs are in between. However, the behaviour of all types of copolymerisate concrete is completely different. The diffusion coefficient is yet approximately 20–60% higher than in the reference concrete. All types of copolymerisate concrete have the largest diffusion coefficient. The high-performance types of concrete have almost the same value, so that a clear distinction is hardly possible. Also in the dry cup, RPC is well below the other types of concrete and, with a diffusion coefficient that is about 10 times smaller, again has an exceptional position in this comparison.

Like the wet cup method, in the dry cup method, SCC 1 lies between the copolymerisate and conventional types of concrete and the high-performance types of concrete as well. The increase amounts to approximately 30% between 20 and 80 °C. In the dry cup method, the values for SCC 2 are close to the HPC; only the increase depending on temperature is much stronger. In the dry cup method, the diffusion coefficient of SCC 2 is approximately 40% of the value of SCC 1 (80 °C). In the wet cup method, on the other hand, the diffusion coefficient of SCC 2 reached approximately 85% of the value of SCC 1, likewise measured at 80 °C.

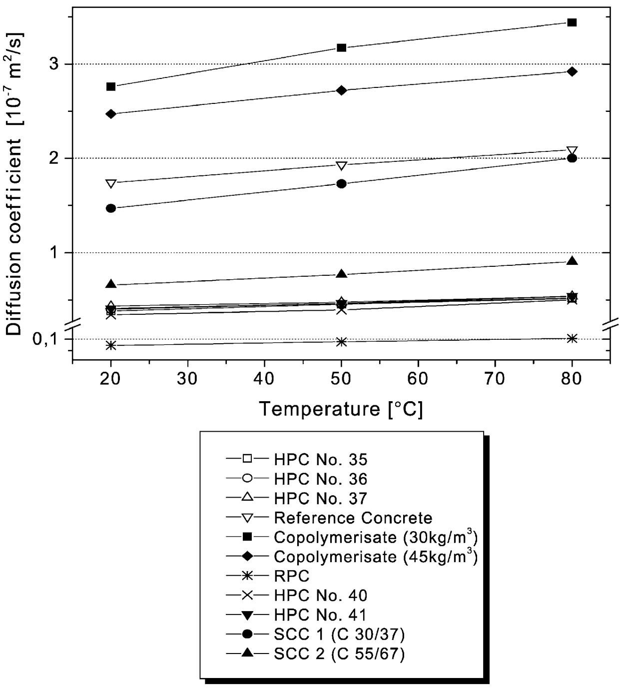

Fig. 6. Diffusion coefficient of all concretes vs. temperature (dry cup method).

5. Discussion of the results

5.1. Influence of temperature on permeability

The influence of temperature on the permeability can be predicted by theoretical considerations. The permeability depends on the viscosity of water and, to a smaller extent, on the density of water. The viscosity drops from 1.00 MPa s at 20 °C to 0.35 MPa s at 80 °C. The density is reduced from 1.00 to 0.97 kg/m^3. On the other hand, the dependence of these quantities on temperature is not linear. If a factor χ is calculated, which represents the influence of temperature on the permeability, it reads:

$$\chi(T) = \frac{k_{\mathrm{w}}(T)}{k_{\mathrm{w},20^\circ\mathrm{C}}} = \frac{\rho_{\mathrm{w}}(T)}{\rho_{20^\circ\mathrm{C}}}\frac{\eta_{20^\circ\mathrm{C}}}{\eta(T)}. \tag{1}$$

Inserting the appropriate values into Eq. (1), one gets the factor $\chi(T)$ in Table 4.

The experimental results are given in Table 5.

According to this, the permeability between 20 and 80 °C should rise distinctly. Regarding Fig. 3 in comparison to that, a minimum increase of the permeability coefficient of 18% (copolymerisate concrete with 30 kg/m^3) and a maximum of 92% (HPC no. 35) can be recognized between 20 and 80 °C. The average is approximately 65%. So it is lower than it could be expected from theory. This could be due to interactions of water and concrete that likewise depend on temperature and were not noticed so far. In this context, for example, the hydration process, which is accelerated at higher temperatures, has to be mentioned. Transferring this context on hydration of cement, a faster blocking of the pores because of the formation of hydrate crystals at higher temperatures can be assumed. This is also true for other processes like, e.g., swelling. Regarding the thermal expansion of the copolymerisate in comparison to the concrete, it is approximately 10 times as large ($\alpha_{\mathrm{T,copolymerisate}} = 10 \times \alpha_{\mathrm{T,concrete}}$). Because of the increase on temperature, the synthetic material extends much more than the concrete and thus blocks the pores.

5.2. Influence of temperature on diffusion

The influence of temperature on the diffusion coefficient can also be predicted from theory. Assuming that water behaves as an ideal gas and that there is no interdependence with other materials, the diffusivity depends on the mean

Table 4
Factor $\chi(T)$ as function of temperature

Temperature [°C]	20	50	80
Factor $\chi(T)$	1.00	1.80	2.74

Table 5
Influence of temperature on permeability, experimental results

	Temperature [°C]		
Concrete	20	50	80
Reference	1.0	1.16	1.26
Copoly-30	1.0	1.15	1.18
Copoly-45	1.0	1.48	1.80
HPC no. 35	1.0	1.62	1.92
HPC no. 36	1.0	1.26	1.55
HPC no. 37	1.0	1.46	1.78
HPC no. 40	1.0	1.35	1.50
HPC no. 41	1.0	1.35	1.58
RPC	1.0	1.13	1.40
SCC 1	1.0	1.16	1.26
SCC 2	1.0	1.15	1.70

velocity of the gas molecules $\bar{c}$ and on the free path length λ according to Ref. [7]:

$$D = 0.599\bar{c}\lambda. \tag{2}$$

The mean velocity of gas molecules is (Eq. (3)):

$$\bar{c} = \left(\frac{8RT}{\pi M}\right)^{1/2} \tag{3}$$

with $R=(pV)/(nT)$ and $V=V_0(T/T_0)$ with the following quantities:

T	Test temperature [K]
T_0	273 [K]
M	Mole mass [g/mol]
V_0	Volume of the gas at 273 [K] (22,414 cm^3)
V	Volume of the gas at test temperature
n	Mole rate, n=(m/M), regarded as constant within the scope of the test temperature of 20–80 °C

In this context, the volume change of a gas at constant pressure with increasing temperature, which is given by the Gay–Lussac law, is particularly important. The mean velocity is then given by:

$$\bar{c} = \underbrace{\left(\frac{8pV_0}{n\pi M}\right)^{1/2}}_{\text{const.}}\left(\frac{T}{T_0}\right)^{1/2}. \tag{4}$$

The free path length becomes:

$$\bar{\lambda} = \frac{\sqrt{2}}{2\pi(L/V)d^2} = \underbrace{\frac{\sqrt{2}V_0}{2\pi L d^2}}_{\text{const.}}\frac{T}{T_0} = \text{const.}\left(\frac{T}{T_0}\right) \tag{5}$$

Table 6
Influence of temperature on diffusivity

Temperature [°C]	20	50	80
Influence of diffusivity [%]	0	16	32

With the reference temperature of 20 °C.

with L=Avogadro constant and d=diameter of the molecule.After inserting Eqs. (4) and (5) in Eq. (2), the dependence of the diffusion coefficient on temperature becomes finally (Eq. (6)):

$$D = \text{const.}\left(\frac{T}{T_0}\right)^{3/2}. \tag{6}$$

Table 6 shows the predicted influence of the diffusivity as function of temperature.

Table 7 shows the experimental results of the diffusivity with respect to the reference temperature of 20 °C.

According to Table 6, the diffusivity between 20 and 80 °C should rise by 32% at 80 °C. Regarding the experimental results, which are given in Table 7, a minimum increase of the diffusion coefficient of 18% (HPC no. 40) and a maximum of 92% (HPC no. 41) can be recognized. So the experimental results show nearly the same dependence that could be expected from theory.

The assumption of an ideal gas can only be partly true since the transport of water vapour is always a combined transport of vapour and liquid [9]. Especially at high relative humidity, the fluid transport becomes more dominate.

First, the storage of water vapour takes place in a dry material that is impinged on with water vapour. During this process, the pore walls are coated with a molecule layer. In case of a further increase in moisture, the narrow parts are filled with water as a result of capillary condensation. More water vapour diffusion takes place in the pores while a transport of liquid water already can be noticed in the narrow parts. If there is a further rise of the moisture content, the sorbate layer that is deposited at the pore walls becomes so thick that the transport can totally be effected in the liquid phase. This causes an intense growth of the transport volume. Therefore, the mass flow that was determined in the wet cup method consists of two combined

Table 7
Influence of temperature on diffusivity (in percent), experimental results

	Temperature [°C]	
Concrete	50	80
Reference	11	20
Copoly-30	15	35
Copoly-45	14	32
HPC no. 35	20	36
HPC no. 36	15	30
HPC no. 37	10	23
HPC no. 40	10	18
HPC no. 41	21	41
RPC	15	25
SCC 1	18	36
SCC 2	17	38

According to Table 6, the diffusivity between 20 and 80 °C should rise by 32% at 80 °C.

effects: the original water vapour diffusion and a transport of liquid water. The following equation describes the combination of both effects (Eq. (7)):

$$\dot{m} = \dot{m}_{\mathrm{D}} + \dot{m}_{\mathrm{Fl}} = -\underbrace{\frac{D_{\mathrm{D}}}{R_{\mathrm{D}}T}\frac{\mathrm{d}p_{\mathrm{D}}}{\mathrm{d}x}}_{\text{Transport by diffusion}} - \underbrace{D_{\mathrm{W}}\frac{\mathrm{d}w}{\mathrm{d}x}}_{\text{Fluid transport}}. \quad (7)$$

The correlation between relative humidity and temperature can be demonstrated by the Kelvin–Thomson equation (Eq. (8)):

$$\varphi = \exp\left[-\frac{2\sigma\cos\theta}{r\rho_{\mathrm{W}}R_{\mathrm{D}}T}\right]. \quad (8)$$

It can be seen from this that the relative humidity increases depending on a rising temperature. This promotes the transport of liquid water. Since—as mentioned above—this is the more efficient transport mechanism, the diffusion coefficient also increases. Unfortunately, it is not possible to quantify the different effects. Only the total mixed transport could be studied.

6. Summary

Tests have been performed with the aim to establish the dependency of permeability and diffusivity of concrete on temperature. It has been shown that the permeability increases by 13–62% when the temperature is raised from 20 to 50 °C and by 3–55% by an additional increase to 80 °C. There is a considerable variation depending on the types of concrete. The diffusivity increases by 10–21% from 20 to 50 °C and by 8–21% from 50 to 80 °C. There is also a considerable variation with respect to the types of concrete. The increase of permeability and diffusivity with temperature is predicted from the theory of thermodynamics. The theoretical prediction is in close agreement with the average values of the experiments.

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