Water Repellent Treatment of Building Materials
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Determination of Penetration Profiles of Water Repellent Agents by Neutron Absorption Technique

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Abstract

Nowadays water repellent treatments are often carried out with silicon-organic compounds. For the performance of an impregnation the penetration profile of the active substance, i.e. silicon resin is decisive. The penetration profile depends strongly on the transport of the water repellent agent by capillary suction into the cement-based material. In this contribution the fundamentals of neutron radiography will be briefly presented. It will be shown that this method is a powerful tool to characterize transport processes in situ. The transport of an aqueous silane emulsion and an undiluted silane (100%) into hardened cement paste has been investigated. First results will be presented which show that the penetration profiles depend strongly on the used type of water repellent agent.

Keywords: penetration profile, neutron radiography, FT-IR-spectroscopy

1 Introduction

The water absorption of porous building materials can be significantly reduced by surface treatment with water repellant agents. Nowadays, silanes and siloxanes are comonly used for the impregnation of concrete. The uptake and the penetration depth of the active substance are crucial for the performance of an impregnation. Therefore, the transport of the silicon organic compound into the concrete must be studied in detail to characterize the factors which influence the penetration profiles. Neutron radiography is one of the few method for non-destructive examination of transport processes of hydrogen-containing liquids into porous materials [1]. Peterka, Böck and Pleinert used neutron radiography for the study of water absorption in bricks treated with water repellent agents [2].

Nemec, Rant, Apih and Glumac studies the transport and polymerization of two silicone-based water repellent agents in clay bricks [3].

So far, no detailed studies were made to investigate the influence of the way of application of the water repellent agents on the penetration of cement-based materials.

We have studied the process of transportation of water-diluted and undiluted water repellent agents into hardened cement paste. Investigations are still in progress, but first results are presented here.

2 Fundamentals of neutron radiography

While X-rays interact with the electrons of the sample atoms, depending roughly on the third to fourth power of the atomic number, neutrons interact solely with the nuclei with no obvious regularity across the periodic system.

Neutrons can either be absorbed or scattered in the sample. Other than X-rays, neutrons can penetrate most metals and heavy elements well, while hydrogen has a very high scattering cross section for thermal neutrons. A neutron radiography will therefore show a very high contrast for even small amounts of hydrogen contained in most materials.

Neutrons can only be detected by a nuclear reaction. A suitable isotope will absorb neutrons and will either split into fragments or form a new isotope, emitting gamma or beta rays. The reaction products in turn can excite the emission of scintillation light in a suitable surrounding scintillator.

We have used a commercially available 6LiF +ZnS(Ag) screen. The kinetic energy of the reaction products of

$$6Li + n \rightarrow 4He + 3H + 4.7\ \text{MeV} \quad (1)$$

is transformed into roughly 177,000 photons in the silver-doped zinc sulfide scintillator. We used a cooled CCD camera to view the scintillator screen via a mirror [4-6]. The active area is currently set to about 20 x 20 cm² and is projected onto a 512 x 512 pixel CCD matrix, rendering a pixel resolution of approx. 0.4 mm.

The system is installed at the cold neutron radiography facility of the Munich Research Reactor FRM. The available beam is 25 cm high by 2.5 cm wide, so the whole setup has to be scanned across the beam in order to irradiate large areas. At a typical flux of $3*10^7$ n/cm²/sec and a scanning speed of 1 cm/sec, the typical fluence is $7.5 * 10^7$ n/cm2.

3 Experiments

3.1 Preparation and conditioning of test specimens

For the experiments hardened cement paste was used. The hardened cement paste is prepared with portland cement CEM I 42.5 and a water/cement ratio of 0.32 using a high speed mixer (10 000 r/min.). After preparation all specimens are stored at 20 °C under water for 4 monthes. The dimensions of the specimens have been chosen to be 100 x 70 x 10 mm^3. Afterwards specimens are dried at 50 °C until the weigth is constant. The area of the sucking surface is 70 x 10 mm^2.

3.2 Neutron radiography of the specimens

The specimens were put into a small dish with about 2-3 mm fluid level of water repellent agent over a thin plastic net and were allowed to soak for 24 hours. After that the specimens are immediatly placed in the neutron radiography set-up. The specimens were put on a sample holder in front of the detector screen and were scanned across the neutron beam.

A full quantitative analysis of the hydrogen content has to take into account the scattered neutrons hitting the detector in the wrong place [1], but the simple transmission images are sufficient for a qualitative study of the agents behaviour.

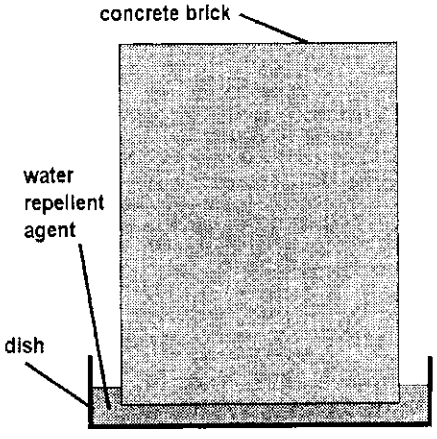

Figure 1: Set-up for the capillary suction experiments

For polymerization of the silanes the specimens have been stored in a closed box after the neutron radiography procedure.

3.3 Analysis of the impregnated specimens by FT-IR-spectroscopy

For the determination of the penetration profile the FT-IR-spectroscopy has also been used. Fundamentals of this method are described in [7]. For analyses the specimens have been treated 28 days after the neutron radiography procedure in the following way. Starting from the surface which was in contact with the water repellent agent thin layers of 2 mm respectively 5 mm have been cut. The sliced pieces were dried at 105 °C and after that the pieces are ground. For FT-IR-spectroscopy samples are prepared by using the KBr-technique. Therefore, 1000 mg potassium bromide is mixed with 40 mg of the ground and dried sample in a mortar. For making a transparent sample 250 mg of this mixture are compressed under vacuum with 250 bar in a specially designed mould. With these samples FT-IR-spectra with 20 scans in the range of 2800 cm^{-1} to 3100 cm^{-1} are taken. The FR-IR-spectra are evaluated by the baseline-method, which is implemented in the FT-IR-spectrometer software.

4 Results and Discussions

4.1 Evaluation of the neutron radiography images

The neutron radiography is based on the fact that neutrons are adsorbed by hydrogen-containing substances such as water, alcohols or silicon-organic compounds. The degree of adsorption depends on the concentration of the hydrogen-containing molecules in the observed material. For example, the absorption of water saturated concrete is significantly higher as compared to the same concrete after drying at 105 °C. Results of these measurements are grey-scaled images. Typical images of neutron radiography are shown in fig. 2. The specimen shown in fig. 2a was in contact with water for two hours. The black coloured zone represents the water saturated cement paste and the grey coloured zone represents the remaining moisture content of the hardened cement paste after drying at 50 °C. It is obvious that the capillary rise can be measured directly in this way. Also, the shape of the water front can be observed easily. The specimen, shown in fig. 2b was in contact with an undiluted silane for 24 hours. In this case the black coloured zone represents the silane saturated cement paste. The capillary rise has been determined to approximately 43 mm.

Both images show that the hydrogen containing liquids, i.e. water and silane, can be detected by neutron radiography. In fig 2c the image of a specimen is shown which was in contact with an aqueous silane emulsion for 24 hours. The front of the emulsion cannot be observed clearly. It seems, that the zone under the dotted line is sligthly darker coloured as compared to the rest of the specimen. Under this assumption the penetration depth may be estimated to be 4.5 mm.

The presented results show that neutron radiography is suitable to characterize the rising front of water or silanes in a porous material. Therefore, neutron radiography is a powerful tool for studying the performance of new application technologies or new types of water repellent agents in a nondestructive way.

4.2 Determination of penetration profiles by FT-IR-spectroscopy

The penetration profiles have also been determind by means of FT-IR-spectroscopy. Results of these analyses are shown in fig. 3. For silane (100%) the content of active substance increases from the surface to a depth of 10 mm. From a depth of 10 mm to 20 mm the values decrease from approxi-

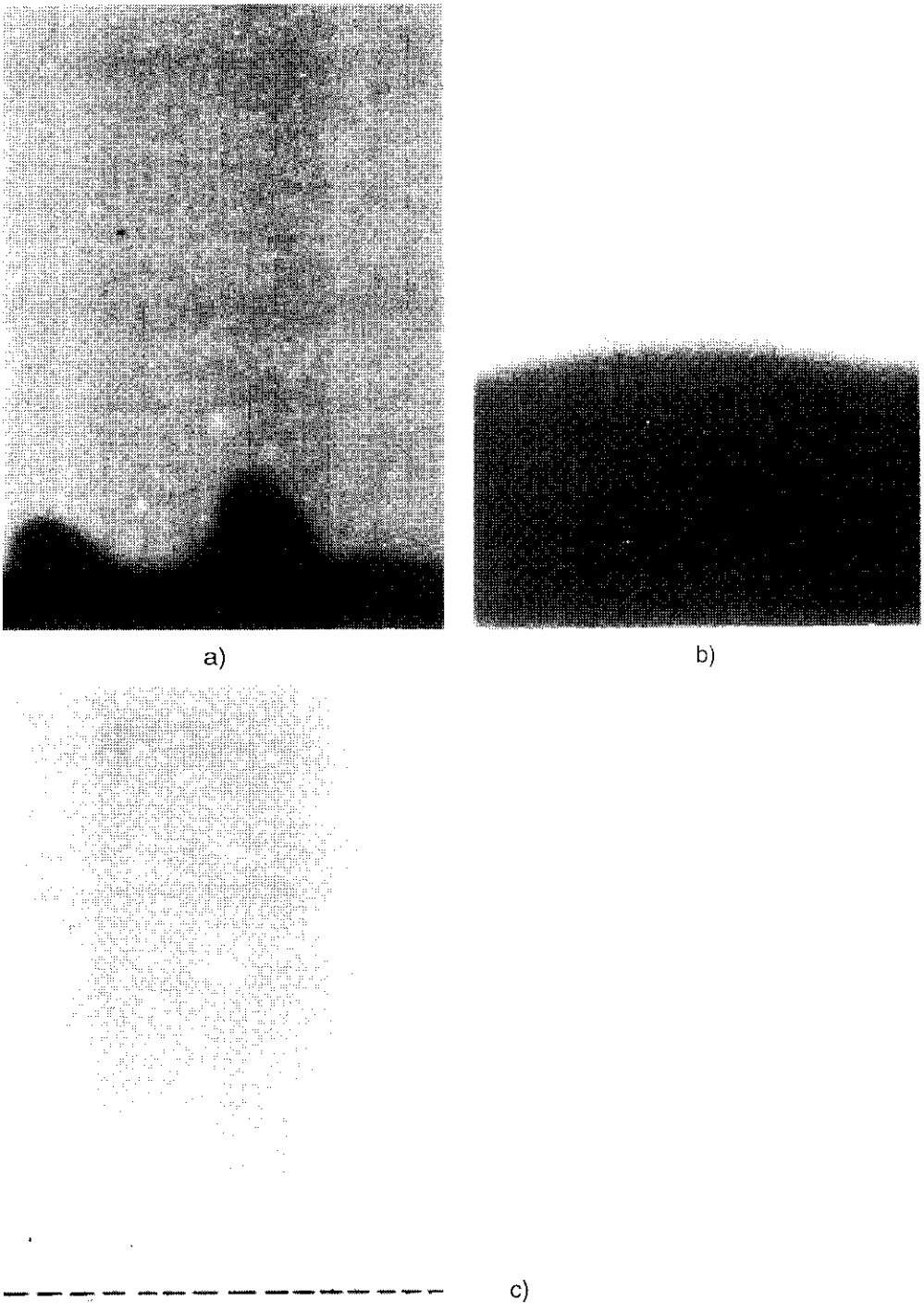

Figure 2: Neutron radiography images a) water. b) silane (100%), c) aqueous silane emulsion

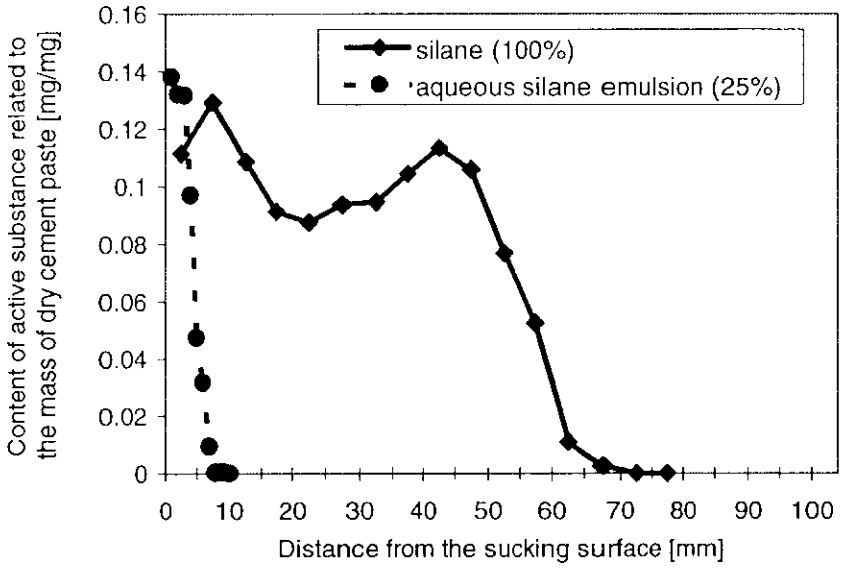

Figure 3: Penetration profiles determined by FT-IR-spectroscopy

mately 0.130 mg/mg cement paste to values of approximately 0.095 mg/mg cement paste. From 20 mm to 45 mm an increase to 0.120 mg/mg cement paste can be observed. From 45 mm to 65 mm the content of active substance drops down significantly. In a depth of 70 mm no active substance can be detected anymore.

For the aqueous silane emulsion the content decreases from a value of 0.14 mm in the first millimeter to non detectable value in a depth of 7 mm.

4.3 Calculation of the penetration depth

The absorbed amount of liquid and the penetration depth can be approximately calculated with the following equations:

$$m_l = A \cdot \sqrt{t} \tag{2}$$

$$x = B \cdot \sqrt{t} \tag{3}$$

$$B = \frac{A}{\psi \cdot \rho} \tag{4}$$

m_l= mass of the absorbed liquid [kg]
A= liquid absorption coefficient [kg/m^2 h$^{0.5}$]

t= time [h]
ρ= density [kg/m^3]
ψ= liquid capacity [m^3/m^3]
B= liquid penetration coefficient [m/h $^{0.5}$]
x= penetration depth [m]

With the values of adsorbed mass of liquid and the contact time the liquid absorption coefficient A has been calculated for the silane and the aqueous silane emulsion respectively (equation (2)). The calculated values of A are shown in table 1. The liquid absorption coefficient A can be transformed with equation (3) to the liquid penetration coefficient B. With equation (4) the penetration depth x has been calculated. The results are also shown in table 1.

In table 2 the values of penetration depth determined by neutron radiography or FT-IR-spectroscopy and the calculated penetration depth are listed.

The values determined by FT-IR-spectroscopy are higher as compared with the results of neutron radiography and the calculation described above. These differences can be explained by diffusion which is well-known for liquid transport in porous media. The neutron radiography images carried out after soaking show a sharp silane front after a contact time of 24 hours. Due to the concentration gradient a transport of silane from the saturated volume to the unsaturated volume takes place and leads to an increase of the penetration depth. The shape of the penetration profile determined by FT-IR-spectroscopy of a depth between 45 mm to 70 mm confirms this postulated effect.

Table 1: Calculated transport coefficients and penetration depth

	Silane (100%)	**Aqueous silane emulsion**
Liquid absorption coefficient A [kg/m^2 h $^{0.5}$]	1.49	0.219
Liquid penetration coefficient B [m/h $^{0.5}$]	0.01	0.00164
Penetration depth x [mm]	49	8.2

Table 2: Penetration depth determined by different methods

	Silane (100%)	Aqueous silane emulsion
Neutron radiography	43	4.5
FT-IR-spectroscopy	65	7
Liquid absorption	49	8.2

5 Conclusions

From the results presented in this contribution the following conclusions can be drawn:

Neutron radiography can be used to characterize the transport of water repellent agents into cement based materials.

The combination of FT-IR-spectroscopy with neutron radiography leads to interesting results concerning transport process of water repellent agents in cement-based materials.

The uptake of the aqueous silane emulsion is restrained. The reason for this has to be further investigated. The penetration depth of silane (100%) is one order of magnitude higher than the penetration depth of the aqueous silane emulsion.

Neutron radiography is useful for studying the performance of new application technologies or new types of water repellent agents.

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