

Unraveling the temperature distribution in fire-damaged concrete by means of PFM microscopy: Outline of the approach and review of potentially useful reactions

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Concrete is a poor conductor of heat. Nevertheless, heating of concrete results in physico-chemical conditions shifting in or out the thermodynamic stability field of specific phases (minerals) in the concrete, effectuating their (dis)appearance. Metamorphic petrology is a branch of geology that deals with the study of such changes; as a basic approach to unravel the heating history of rocks, domains with phases (minerals) specific for a set of discrete physico-chemical conditions are delineated (mapped). Unravelling the heating history of concrete is important to forensic research or to determine whether a concrete structure subjected to fire and its components are still structurally sound. Analogous to metamorphic petrology, thermally triggered reactions in concrete result in changes in specific phases which may be used to trace isograds. In the case of heated concrete, these occur in response to temperature alone, and may hence be used to trace temperature variations in concrete with depth (isograds). The method involves a combination of visual inspection and the use of microscopic methods in a systematic way to study the thermally affected concrete. It is a powerful diagnostic tool that has successfully been applied in research and testing of concrete linings in tunnels, assessment of fire-damage to structures and evaluation of fireproofing in waste incinerators.

Key words: fire-damaged concrete, concrete petrography, phase changes, analysis of heating history

1 Introduction

Heating of concrete, due to fire or otherwise, may result in a variety of structural changes like cracking, spalling, debonding of aggregate and rebars, expansion, and mineralogical / chemical changes such as discoloration, dehydration, dissociation. When a concrete structure is exposed to fire, differential expansions and contractions of various components and constituents within the concrete take place. Depending on the length of exposure to the fire, considerable changes may take place in a structure, ranging from surface cracking and spalling, discoloration of the concrete, excessive expansion, buckling, warping and loss of strength of the reinforcement steel if present. Other changes may include cracking around and across aggregate, loss of bond to aggregate and

reinforcement and internal cracking within the cement paste (figure 1). With regard to the cement paste, evaporation and dissolution, dehydration and dissociation of ettringite, gypsum, calcium hydroxide, calcium carbonate and other phases in the cement paste such as the calcium silicate hydrates may occur (St. John et al. 1998).

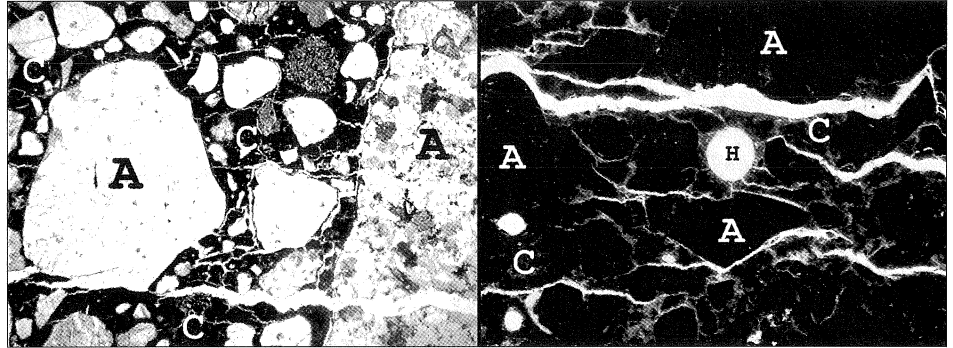


Figure 1. PFM-micrographs showing the development of cracks in and across aggregate particles into the adjoining cement paste as a result of the intense heating. A = aggregate, C = cement paste, H = air-voids. Left microphotograph = plain polarised transmitted light, right = fluorescent light. Micrograph dimensions are 2.7 mm x 1.8 mm.

The extent of alterations depends in most cases on the type and geometry of a structure, the maximum temperature reached during the exposure, the duration of heating at that temperature, and the rates of heating and cooling or quenching. In the case of tunnels, for example, the severity of the fire damage is dependent on the cross-sections or thickness of the concrete elements or linings, controlling thermal gradients developed and the differential thermal expansions of various components within the concrete including steel reinforcement, if present.

Whereas both structural and mineralogical changes (may) affect the integrity of a structure, the latter alterations may be used to deduce the heating history of that structure. Combining metamorphic petrology, which is a branch of geology that deals with the study of changes in rocks due changing physical and chemical conditions and concrete petrology provides a suitable investigative tool for this purpose (Larbi & Nijland 2001). As conditions shift in or out of the thermodynamic stability field of phases, new phases may appear whereas others disappear. A basic approach here involves mapping of the so-called isograds, which are lines of equal physico-chemical conditions, often temperature, separating the occurrence of (combinations of) phases in the field. This technique can also be used to unravel the heating history of concrete.

Analogous to metamorphic petrology, the changes in both the cement paste and the aggregate of the fire affected concrete will result in conversion of certain phases into new ones which may alter the colour of the concrete and the (original) mineralogical composition of the cement paste. Both effects may be used to trace isograds in the concrete element. Since these isograds occur in response

to virtually one single variable, that is temperature, they will roughly coincide with isotherms and hence may be used to trace the temperature variations with depth from the surface. Tracing of the temperature history of the fire-attacked concrete is therefore of vital importance in determining the depth of damage in a particular concrete member that may eventually be discarded; e.g., a reduction of up to 60 % in compressive strength may occur when concrete is heated up to 300 °C.

The primary purpose of unravelling the heating history of the concrete in a fire-damaged structure or concrete element is one of the following:

- to assess the fire resistance of a particular concrete element of a major infrastructure, such as the concrete or mortar lining of a tunnel prior to its application;
- to assist in forensic research of the cause of fire by determining the duration of exposure of concrete to elevated temperatures, etc.;
- to determine whether a structure is still structurally sound and to assess, for example if steel reinforcement in concrete has been heated up to or over its flow temperature and hence has suffered permanent loss of strength, which will affect the structure as a whole.

This paper describes the phenomena underlying the changes in concrete exposed to high temperature or fire and addresses the role of concrete petrography as a valuable tool for assessing the extent of damage to a concrete structure caused by fire and gives a review of potentially useful reactions which may provide more detailed information if required.

2 Investigation procedure

2.1 Introduction

Concrete petrography is a systematic way of examining a piece of concrete in order to characterize and determine what is wrong with it (Larbi & Heijnen 1994). It may be a follow-up of the preliminary structural or site investigation when these alone are not able to provide solutions to the problem and a detailed analysis of samples of the concrete is required. In that case, the next step in the investigation process is to drill cores from specific areas of interest of the structure for further analysis in the laboratory by means of concrete petrography. Once cores have been obtained from the structure, investigation is carried out systematically as follows: Visual inspection, followed by fluorescent macroscopic analysis (FMA) and polarising and fluorescent microscopy (PFM).

2.2 Visual & stereomicroscopic inspection of concrete cores

This cursory part of the investigation is aimed at using the alterations and colour changes in the cores as well as the surface features such as cracking, spalling and popouts, to estimate the possible temperature variations in the concrete as a function of depth from the surface. Of importance are the colour variations with depth from the surface, pattern of cracking in and around the aggregate particles, width and depth of the cracks, dissolution and loss of bonding to the aggregate particles and the integrity of the cement paste. The following is a summary of the isograds that can be traced by hand lense and/or stereomicroscope, reflecting temperatures reached in concrete during fire:

< 300 °C	Normal, no apparent macroscopic changes in concrete; colour remains grey
300 – 350 °C	Oxidation of iron hydroxides like FeOOH in aggregate and cement paste to hematite, $\alpha\text{-Fe}_2\text{O}_3$, causing a permanent change of colour of the concrete from grey to pinkish brown
573 °C	Transition of α -quartz to β -quartz, accompanied by an instantaneous increase in volume of quartz of about 5 %, resulting in a radial cracking pattern around the quartz grains in the aggregate; this phase transition itself is reversible, but the radial cracking provides a diagnostic feature that remains after cooling
> 800 °C	Complete desintegration of calcareous constituents of the aggregate and cement paste due to both dissociation and extreme thermal stresses, causing a whitish grey coloration of the concrete

The successive colour zoning that may result from these changes is schematically given in figure 2, whereas real life examples are illustrated in figure 3. These characteristic effects can be observed with the naked eye or with the aid of a hand lens or with a stereomicroscope which can then be used to give an indication of temperature variations in the fire-damaged concrete. The isograd associated with oxidation of iron hydroxides, like FeOOH, to hematite, $\alpha\text{-Fe}_2\text{O}_3$, at 300-350 °C is particularly of importance as this temperature coincides with the flow temperature of many types of reinforcement steel. At higher temperatures, traditional rebars will gradually undergo permanent loss of strength leading to buckling and warping.

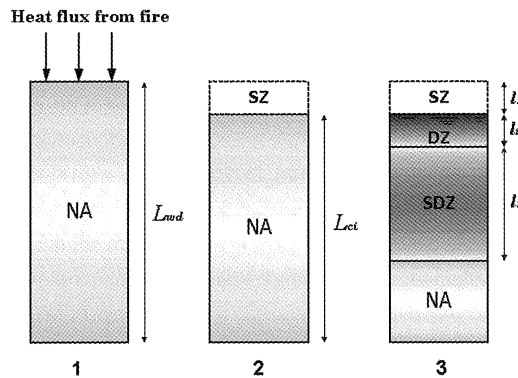


Figure 2. Simplified schematic representation of the processes of spalling, microcracking and further deterioration of the concrete elements as revealed in the cores removed from the tested concrete element. As a result of the intense heating of one surface of the element, pressure is built up in the pores of the cement paste and aggregate due to evaporating water in the concrete in the zone close to the heated surface. Excessive pressure build-up then leads to spalling, which is delineated by SZ. NA = not affected; SZ = spalled zone; DZ = almost dry or dehydrated zone; SDZ = semi-dry or -dehydrated zone. L_{wd} , L_c , l_1 , l_2 and l_3 are the respective thickness of the zones.

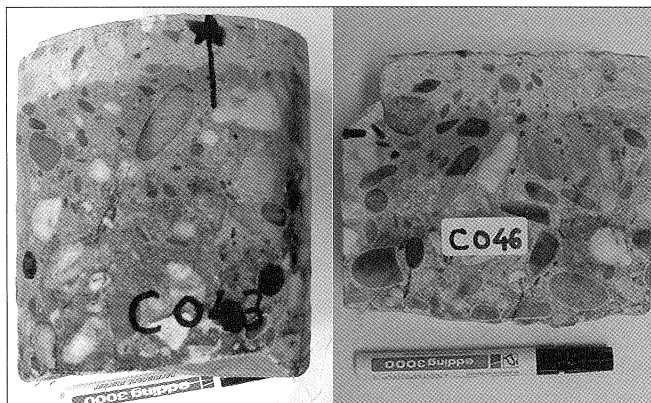


Figure 3. Colour zonation in concrete made with blastfurnace slag cement (CEM III/B). Cores show zonation from normal greenish blue to reddish beige (especially aggregate grains show iron oxidation) to whitish grey near the surface.

Remnants of fire burnt materials collected from the site can offer additional information for estimating the maximum fire temperature reached. Fire debris of materials such as timber, which chars at about 250 °C, aluminium alloys, with a melting point of about 650 °C, glass, which melts at about 850 °C and copper pipes with a melting point of about 1080 °C are quite helpful in this regard.

2.3 Fluorescent macroscopic analysis (FMA)

If detailed information regarding the distribution of cracks, including fine microcracks (cracks with widths usually less than 10 µm) and the integrity (preservation of the compactness) of the concrete with depth from the fire-exposed surface are required, flat-polished fluorescent sections can be prepared from the drilled cores and examined under UV-light, a technique previously applied by Polder & Larbi (1995) to study crack densities in marine concrete. Information on the density and the distribution of microcracks is useful in determining the thickness of concrete (from the spalled surface) that eventually needs to be removed in the case of repair work. It is also important in determining whether fire-attacked elements and components (including reinforcement steel) are still structurally sound and that the local loading conditions, in the long-term would not adversely affect the mechanical properties and the durability of the elements. The criteria used to classify the extent of microcracking in the concrete are given in table 1. Examples of polished slabs showing cracking due to heating are shown in figure 4.

Table 1: Criteria used to classify the extent of microcracking in the concrete.

Classification of micro-cracking	Description of classification
Very low	20 % or less of the cement paste contains more than 5 microcracks / surface unit ¹
Low	20-40 % of the cement paste contains more than 5 microcracks / surface unit
Moderate	40-60 % of the cement paste contains more than 5 microcracks / surface unit
High	60-80 % of the cement paste contains more than 5 microcracks / surface unit
Very high	80 % or more of the cement paste contains more than 5 microcracks / surface unit

¹ This applies to a specific unit area, for example mm² of cm², as long as the same unit is used throughout the investigation.

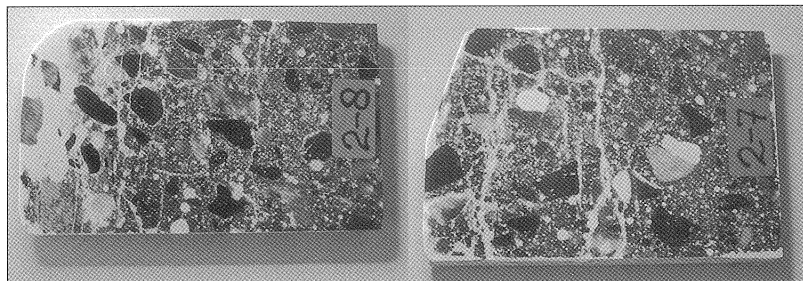


Figure 4. FMA-macrographs showing the distribution of cracks in the polished sections prepared from cores removed from reinforced concrete linings of tunnel elements subjected to fire testing; samples ca. 100 x 50 mm².

2.4 Polarising and fluorescent microscopy (PFM)

If more detailed information is required, this can be derived from polarising and fluorescent microscopy, PFM. The PFM-technique is based on examination of fluorescent thin sections with the aid of a combined polarising and fluorescent light microscope. Details of a number of methods of preparing fluorescent thin and polished sections are given in Larbi & Heijnen (1994).

The thin sections are prepared from drilled cores. Where small thin sections with a size of 50 mm x 30 mm are used, several sections may be made. Where large thin sections are made, for example sections with a size of 150 mm x 100 mm, a single thin section may be enough. Examination of the thin sections with the aid of a polarising and fluorescent microscope may reveal (dis)appearance of phases in the cement paste, secondary phases, etc., which can be used to define additional isograds

as a function of depth from the surface of the concrete. Some of the most widely applicable isograds that can be diagnosed by PFM are:

70 – 80 °C	Dissociation of ettringite, causing total depletion of ettringite in the cement paste
105 °C	Loss of physically bound water in aggregate and cement paste; this effect causes an increase in the capillary porosity and microcracking of the cement paste which can easily be recognised by fluorescent microscopy
120 – 163 °C	Dissociation of gypsum, causing its depletion in the cement paste
450 – 500 °C	Dissociation of portlandite, causing its depletion in the cement paste
573 °C	Transition α -quartz to β -quartz, resulting in a radial cracking pattern
600 – 800 °C	Dissociation of carbonates; depending on the content of carbonates of the concrete; e.g. if the aggregate used is calcareous, this may cause a considerable contraction of the concrete due to release of CO ₂ ; the volume contraction will cause severe microcracking in the cement paste
800 – 950 °C	Final dissociation of calcium silicate hydrates, C-S-H and remaining phases in the cement paste resulting in complete desintegration of the concrete, with severe microcracking

Several of these examples require, of course, knowledge whether the phase supposed to have disappeared, was originally present in the concrete. A more complete review of other potentially useful reactions is given below.

3 Review of potentially useful reactions

Above, an outline is given of the general procedure to unravel the heating history of concrete by petrographic analysis, and the most useful diagnostic features and reactions have been listed. In particular cases, and with additional investigation methods like XRD, more information may be obtained from a series of less easily detected reactions, in particular in the cement paste.

3.1 C-S-H and related phases

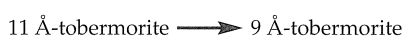
C-S-H and their more crystalline counterparts tobermorite and jennite, show increasing loss of water, contraction of the basal spacing, and finally the formation of anhydrous silicates.

At 55 – 60 °C, contraction of the crystal lattice of tobermorite, the analogue of C-S-H I, occurs:



(Taylor 1998), followed by further contraction over 100 – 300 °C

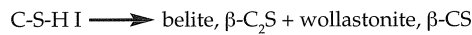
(Cong & Kirkpartick 1985):



Jennite, counterpart of C-S-H II, shows loss of water at 70 – 90 °C (Cong & Kirkpatrick 1985, Taylor 1998):



Gel-like C-S-H starts dissociating at 180 °C (Schneider 1982) and, in general, most chemically bound H₂O is liberated above 300 °C. At higher temperatures, 375 – 400 °C, a poorly crystalline phase is formed from jennite, involving loss of OH⁻ groups (Carpenter et al. 1966), with finally the formation of belite (larnite), β-C₂S, and/or wollastonite, β-CS (Carpenter et al. 1966, Cong & Kirkpatrick 1985). There are many indications that the formation of belite is gradual, and occurs over a considerable temperature range. Schneider & Herbst (1990) give temperature ranges of 120 to 500 °C for its formation from C-S-H I, 300 – 800 °C for 9 Å-tobermorite and 650 – 900 °C for C-S-H II. In contrast, Khoury et al. (1985) indicate that the reaction

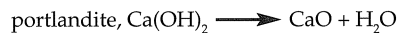


occurs above 800 °C, in agreement with 835 – 900 °C indicated by Taylor (1998); the corresponding reaction for jennite is considered to occur above 900 °C (Carpenter et al. 1966, Cong & Kirkpatrick 1985). For C-S-H, the ratio in which belite and wollastonite are formed depends on the concentration of SiO₂ present (Wedler & Hummel 1947, Yoshikane 1988, Schießl et al. 1998).

Autoclaved concretes show some additional reactions: At 650 – 700 °C, dicalcium silicate hydrate, C₂SH decomposes to not yet identified phases, whereas at 680 – 700 °C, xonotlite, C₆S₆H, will decompose to belite (Schneider & Herbst 1990).

3.2 Portlandite and calcium carbonate

Portlandite (calcium hydroxide), either present in the hardened cement paste or concrete, or newly developed as a breakdown product of other phases, dissociates in the range 450 – 550 °C, due to (Khoury et al. 1995, Schneider 1980, 1982, Maultzsch 1988, Taylor 1998, Schneider & Herbst 1990):



Some authors, however, report the presence of relic portlandite in X-ray diffractograms at 560 °C (Schneider 1973).

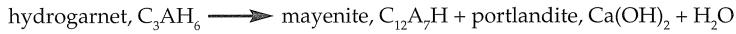
Above 800 to 900 °C (Maultzsch 1988, Schneider & Herbst 1990), calcite will decompose due to



In presence of SiO_2 , Al_2O_3 or Fe_2O_3 , calcite in cement pastes may become unstable at lower temperatures of 500 – 600 °C (Taylor 1964, Lach 1970). Generally, hardened cement pastes show an increase in free CaO with increasing temperature (Schliefl et al. 1998, Yoshikane 1988).

3.3 Ca-aluminate hydrates

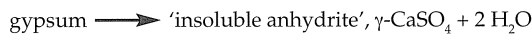
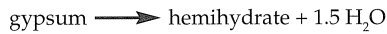
Hydrogarnet, C_3AH_6 , decomposes due to the reaction (Kunzel 1969, Passaglia & Rinaldi 1984):



Temperatures for this reaction are estimated at 200 – 250 °C (Kuzel 1969), 250 – 310 °C (Passaglia & Rinaldi 1984) and 280 – 310 °C (Maultzsch 1988). The reaction is followed by decomposition of portlandite to CaO (see above) and that of mayenite to C_3A at 810 °C (Kuzel 1969).

3.4 Sulfate-bearing phases

At 70 – 200 °C (Taylor 1998) or 90 – 160 °C (Maultzsch 1988), gypsum decomposes by the reactions:



Based on DTA/TGA studies, ettringite is considered to become unstable at 125 – 130 °C (Taylor 1998) by the reaction (Schneider & Herbst 1990):

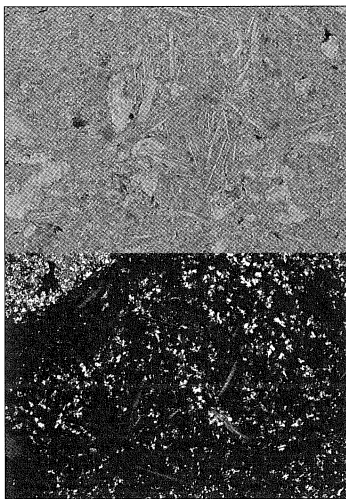
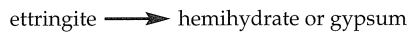


Figure 5. Microphotograph of a sample of the Enschede firework factory explosion site showing partial breakdown of ettringite in an air void to gypsum. The lenticular crystals are remnants of ettringite, whereas the finer particles represent gypsum. Microphotograph dimensions are 0.7 mm x 0.45 mm (plane & crossed polars).

(figure 5). However, often dissociation already starts at about 70 - 80 °C, when ettringite will lose all water in the channels of its structure (Pöllman et al. 1989).

At 180 – 190 °C, monosulfate, $C_4A\bar{S}H_{12}$ decomposes (Taylor 1998). Maultzsch (1988) gives slightly higher temperatures for the decomposition of C-A-S-H phases in general, viz. 250 – 270 °C.

C_4AH_x phases typically decompose to mixtures of portlandite and a sodalite-like phase, C_4A_3H at 150 – 250 °C, followed by decomposition of portlandite to CaO at 410 °C and of the sodalite-like phase C_4A_3H to a mixture of CaO and mayenite at 630 °C (Butler et al. 1959).

3.5 Reactions in aggregate particles

Over the range 300 – 350 °C, Fe-hydroxides in the aggregate (sandstones etc.), like goethite, α -FeOOH, and lepidocrocite, γ -FeOOH (and, if of suitable composition, Fe in the cement paste) oxidize to hematite, α -Fe₂O₃, resulting in pinkish to reddish brown coloration (Fig. 3).

At 573 °C, SiO₂ shows a phase transition from α -quartz to β -quartz, which is accompanied by an instantaneous increase in volume. This transition itself is reversible, but its past occurrence may be traced by the occurrence of radial cracks around the quartz grains (Larbi & Nijland 2001). Upon further heating, above 1050 °C α -cristobalite is formed, thus overstepping tridymite (Mosesman & Pitzer 1941, Heaney 1994). Depending on temperature, upon heating, opal, SiO₂•nH₂O, will change into either α - or β -quartz.

If limestone or other carbonate-bearing aggregate is present, it will show decomposition similar to that of carbonates in the cement paste (see above).

3.6 Extreme temperature conditions

At extreme temperatures, hardened cement pastes and concrete will show incipient melting. Melting starts at about 1150 – 1200 °C (Schneider 1982).

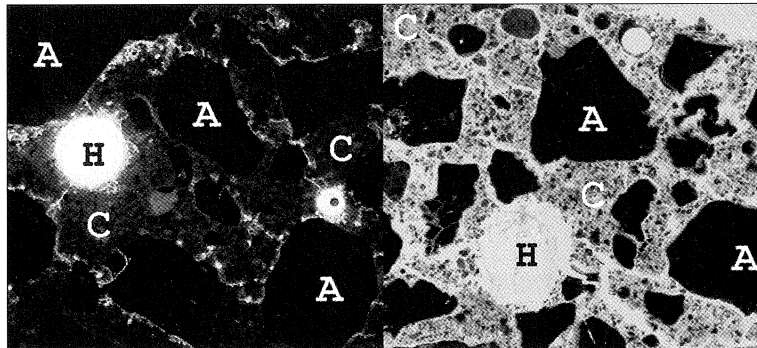


Figure 6. PFM-micrographs showing the effect of the intense heating on the capillary porosity of the cement paste. There is a clear increase in the capillary porosity of the cement paste that was exposed to intense heating (right) compared to that of the reference concrete (left). Cores have been removed from reinforced concrete linings of tunnel elements subjected to fire testing. A = aggregate, C = cement paste, H = air-voids. Micrograph dimensions are 2.7 mm x 1.8 mm.

3.7 General remarks

A review of all potentially useful reactions discussed above is given in table 2. From that table, it becomes evident that several of these reactions result in an increase in capillary porosity of the cement paste. An example of such an increase is given in figure 6.

Tabel 2: Phase changes in hardened cement paste and concrete with increasing temperature. Note that their occurrence depends not only on temperature and duration of heating, but also on cement and aggregate composition, and, possibly, partial pressures of H₂O and CO₂.

°C	Reaction	Macro/microscopically diagnostic features
55 - 60	14 Å-tobermorite → 11 Å-tobermorite	
70 - 80	Dissociation of ettringite	Absence of ettringite in cement paste
70 - 90	Jennite → metajennite	
70 - 200	Gypsum → hemihydrate or γ -CaSO ₄	Absence of gypsum in cement paste
100 - 300	11 Å-tobermorite → 9 Å-tobermorite	
180	Dissociation of gel-like C-S-H	Increase in capillary porosity
180 - 190	Decomposition of monosulfate	
200 - 310	Hydrogarnet → mayenite + portlandite	
300	Loss of chemically bound water	Increase in capillary porosity
300 - 350	Oxidation of FeOOH to α -Fe ₂ O ₃	Change in color to pink or reddish brown
450 - 550	Portlandite → CaO + H ₂ O	Absence of portlandite in cement paste
573	α -quartz → β -quartz	Radial cracks around aggregate particles
650 - 700	Decomposition of C ₂ S-H	
680 - 700	Xonotlite → belite	
800 - 900	Decomposition of carbonates	Desintegration of carbonate grains
800	C-S-H I → belite + wollastonite	Complete desintegration of cement paste
900	Jennite → belite + wollastonite	Complete desintegration of cement paste
1050	β -quartz → cristobalite	Appearance of cristobalite
1150 - 1200	incipient melting	Presence of quenched melt / melt textures

4 Concluding remarks

It is shown that a combination of visual examination and microscopic analysis provides a powerful tool for diagnosis of concrete exposed to fire. This approach has successfully been applied in research and testing of various cement-based materials and concrete as lining for underground tunnels in the Netherlands and other parts of Europe, for evaluation of fireproofing in waste incinerators and for assessment of fire-damage in tunnels, structures and large buildings, and in support of forensic investigation of fire cases at explosion sites.

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