

Effect of the heating rate on residual thermo-hydro-mechanical properties of a high-strength concrete in the context of nuclear waste storage

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ABSTRACT

Concrete is likely to be used in massive structures for nuclear waste long-term storage facilities in France. In the framework of vitrified waste and spent fuel management, these structures could be submitted to high temperatures. In standard conditions, ambient temperature should not exceed 60°C but in case of failure of a cooling system, concretes could be temporarily exposed to temperatures up to 250°C. Depending on the temperature rise kinetics, concretes could be damaged to a greater or lesser extent. In this context, an experimental study on the effect of heating rate on concrete thermo-hydro-mechanical properties exposed to high temperatures (110 - 250°C) was carried out at the French Atomic Energy Commission (CEA). Data analysis and interpretation provided enough arguments to conclude that, at local scale, the impact of heating rate on residual properties was real though relatively limited.

KEY WORDS: nuclear, waste management, interim storage, high-level waste, concrete structures, high-strength concrete, high temperature, heating rate, thermo-hydro-mechanics, residual properties, thermal gradient, strain, hydric stress.

INTRODUCTION

In the framework of nuclear high-level waste (HLW) management, long-term storage facility structures made of reinforced high-strength concrete (HSC) are planned to be built (Fig.1) [1]. In order to demonstrate the long-term safety and integrity of such facilities, it is essential to prove that structural materials will remain stable over extended periods of time, whatever the environmental conditions. Over several decades, HLW – vitrified waste and spent fuel – will generate an important quantity of heat which may affect the long-term behaviour of surrounding structures. In normal conditions, the average facility temperature should be below 60°C. But in case of a critical scenario – any failure of the cooling system for example – internal temperature could rise up to 250°C [2]. It is well established that due to their low porosity and permeability, HSC are very sensitive to high temperatures and that their mechanical and physical properties may be strongly affected by thermal exposures [3-9]. On the other hand, it is generally admitted that HSC behaviour at high temperature is governed by thermo-mechanical processes related to temperature gradients which generate thermal expansion gradients [10, 11] and by thermo-hydric processes linked to water movements in the material porosity [8, 12, 13]. For more than a decade, much has been learned about the behaviour of HSC at high temperature in case of fire [14]. In nuclear applications, such as long-term waste storage, concrete might be exposed to specific thermo-hydro-mechanical stresses. In structures designed to be subjected to high temperature, characteristics values – taking into account the temperature dependency of concrete mechanical and physical properties – have to be set carefully [15]. In this context, it is essential to acquire data related to the properties of HSC for low heating rates. This is the reason why, further to previous studies [16-18], the French Atomic Energy Commission (CEA) performed an experimental programme on the effect of heating rate on residual thermo-hydro-mechanical properties of a HSC submitted to temperatures between 110 and 250°C, with rates ranged from 0.1 to 10°C/min. The results of this study are shown and discussed hereafter.

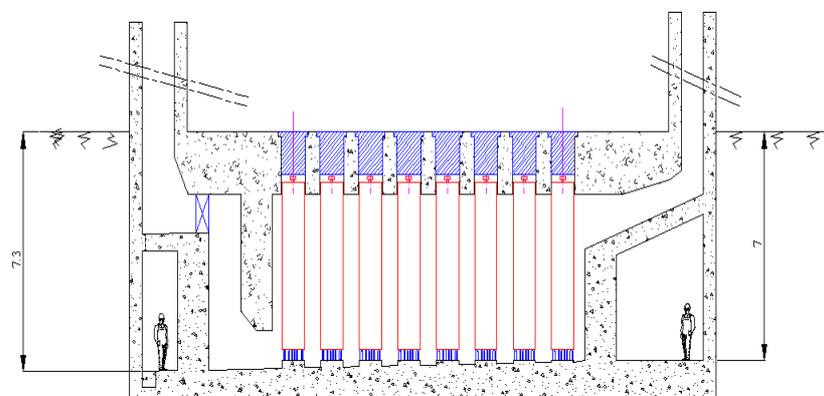


Fig. 1 Schematic cross-section of a surface vault long-term storage facility concept (after [2])

EXPERIMENTAL CONTEXT

The study was achieved with a high-strength concrete used for the MAQBETH large scale experiment – mock-up test – performed at CEA in 2001 [17,19]. This type of material, already selected in previous studies [18], was prepared with silico-calcareous aggregates and was characterized by a 0.46 w/c ratio. The concrete was provided by a Lafarge Company concrete batching plant. Concrete properties (specific density ρ , total water porosity ϕ_w , effective gas permeability k_g , intrinsic permeability k , thermal conductivity λ , compressive strength f_c , tensile strength f_t and elastic modulus E) are given in Table 1. In this table, dynamic mechanical properties – dynamic elastic modulus E_{dyn} and Poisson's ratio ν – obtained through ultrasonic measurements are also given.

After a 9-month curing period in sealed bags, concrete probes were heated – until stable mass loss – at 110, 150, 200 and 250°C with heating rates of 0.1, 1 and 10°C/min. Thermal treatment effects were examined through the evolution of above-mentioned parameters. Residual HSC properties were measured after specimen (French normalised cylindrical probes "11x22", 220-mm high, 113 mm-diameter) cooling (0.2°C/min) at $20 \pm 1^\circ\text{C}$. Before heating, the initial water saturation degree of the HSC was about 84 %.

Table 1. HSC initial properties (20°C)

ρ (kg.m ⁻³)	ϕ_w (%)	k_g (m ²)	k (m ³)	λ (W.m ⁻¹ C ⁻¹)	F_c (MPa)	F_t (MPa)	E (GPa)	E_{dyn} (GPa)	ν
2375 ± 12	13.3 ± 0.5	$4.3 \pm 5.4 \cdot 10^{-19}$	$2.8 \pm 3.2 \cdot 10^{-19}$	2.2 ± 0.1	66.2 ± 5.9	4.6 ± 0.6	43.4 ± 1.0	45.5 ± 0.8	0.25 ± 0.01

RESULTS

Mass loss

Examples of mass loss curves are shown in Fig. 2. These curves proved that the mass loss plateau was reached for each thermal loading. Through this, we checked that there was no significant amount of water unremoved for each temperature level. Slight differences, observed mainly at 150°C, are probably related to discrepancies in the initial water content of the specimens. Mass loss values evolution obtained after thermal treatments are plotted in Figure 3. This parameter evolves linearly with the temperature. On average, at 110, 150, 200 et 250°C, mass loss values were respectively, 11.5 ± 0.1 %, 12.4 ± 0.4 %, 13.0 ± 0.2 % and 13.8 ± 0.3 %. Taking into account experimental uncertainties, we assumed that the mass loss did not evolve with the heating rate. This result confirms that the intensity of dehydration is only controlled by temperature level and that there is no artefact effect due to the heating rate.

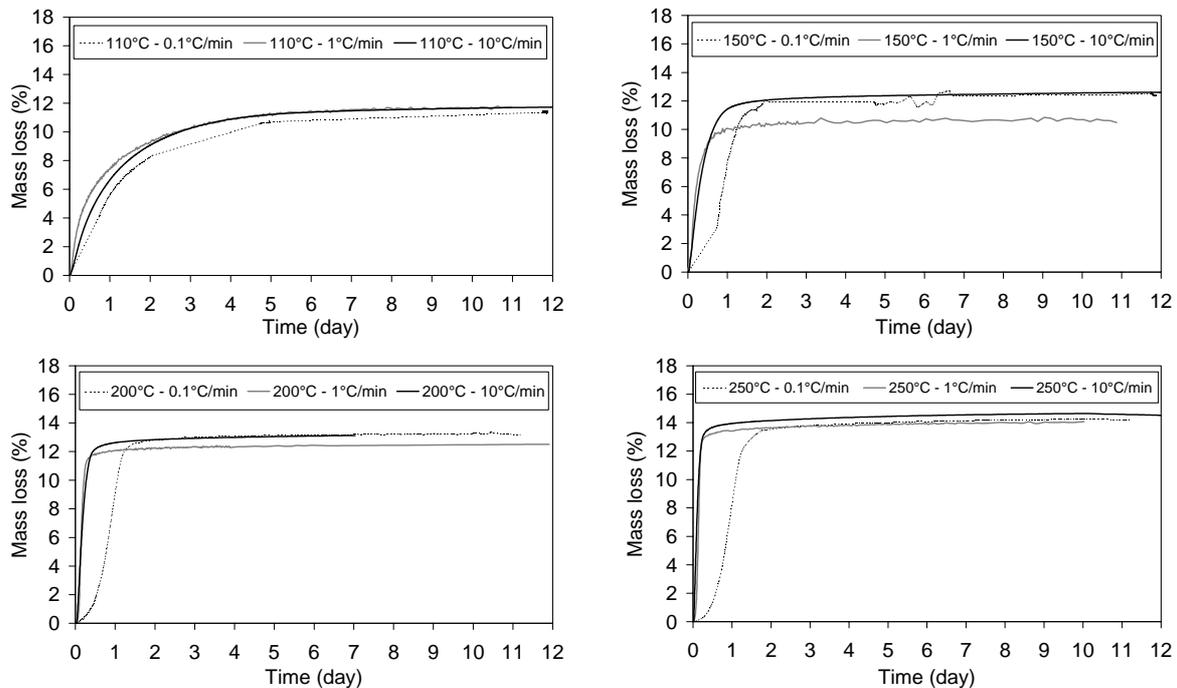


Fig. 2 Evolution of the Mass loss kinetics at 110, 150, 200 and 250°C with heating rate

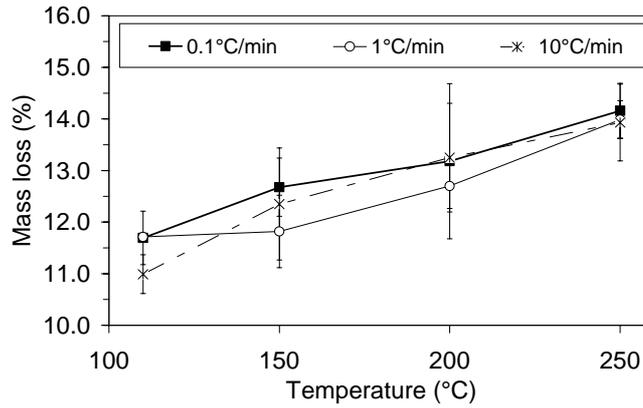


Fig. 3 Evolution of the mass loss with temperature and heating rate

Permeability

Regarding the transfer of fluids – water, vapour, air – through concrete porosity submitted to high temperature, permeability is a key parameter controlling the thermo-hydric behaviour of the material. The evolution of this parameter with the temperature and heating rate is given in Fig. 4. The results showed that there was a significant tendency to assume that a low heating rate was unfavourable to permeability. Permeability values obtained at 0.1 and 1°C/min were relatively close with an average ratio of 1.3. If we compare the 0.1°C/min data with those obtained at 10°C/min, this ratio was 2.5. Taking into account the experimental uncertainties, this ratio difference is quite significant. We can thus admit that thermal loading at 0.1°C/min generated probably more damages than thermal loading at 10°C/min.

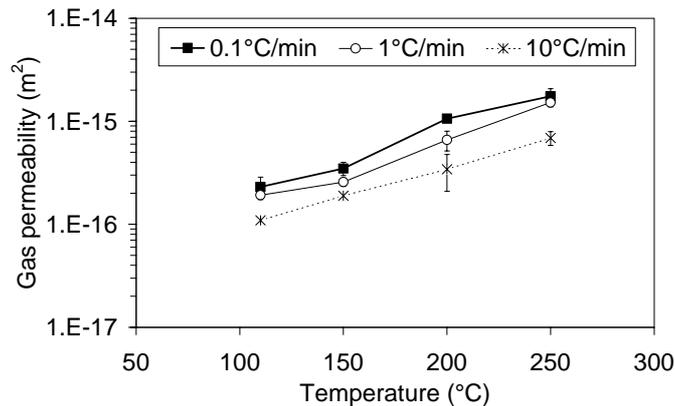


Fig. 4 Evolution of the effective gas permeability with temperature and heating rate

Mechanical properties

The influence of the heating rate on concrete mechanical properties was examined through the evolution of the compressive and tensile strength and the elastic modulus. Compressive strength average values are shown in Fig. 5. If we consider the dispersion of the experimental values, the effect of the heating rate on compressive strength remains unclear. It appeared that the lowest compressive strength values were found with the lowest heating rate (except at 110°C). However, the relative discrepancy between the average values related to the two extreme configurations – 0.1 and 10°C/min – were rather limited and inferior to 10%.

Concerning tensile strength, experimental values are produced in Fig. 6. The results showed that there was no clear hierarchy of the tensile strength values with the heating rate. Globally, for the 150 – 250°C temperature range, tensile strength differences were extremely low. Even if we consider the extreme heating rate configurations, no clear and significant tendency was detected.

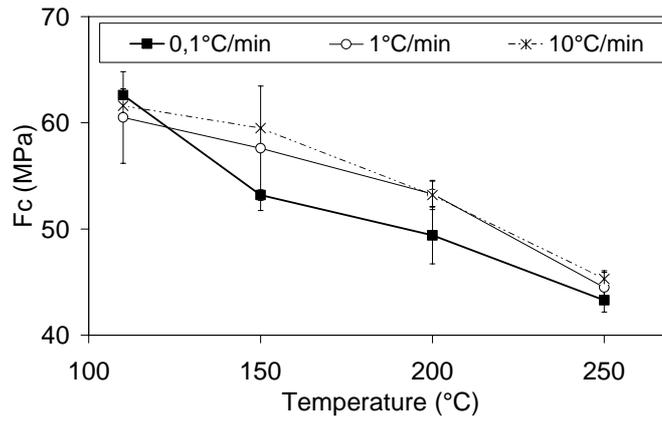


Fig. 5 Evolution of the compressive strength with temperature and heating rate

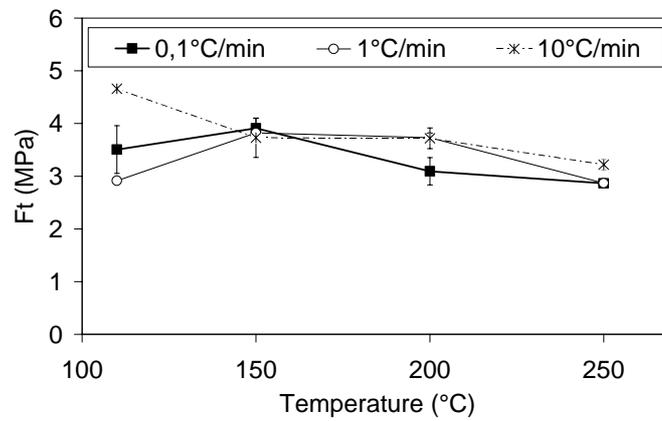


Fig. 6 Evolution of the tensile strength with temperature and heating rate

The elastic modulus data displayed a typical linear trend with temperature (Fig. 7). The dispersion of experimental data was quite low. No significant discrepancy was observed between 0.1 and 1°C/min data sets. The 10°C/min heating rate was characterized by the lowest elastic modulus values. On average, the relative variation between the elastic modulus values provided by the 0.1 and 10°C/min rates, was about 9%. As a general trend, it is possible to conclude that a high heating rate is not favourable to the elastic modulus. We can underline that an opposite trend was observed for permeability.

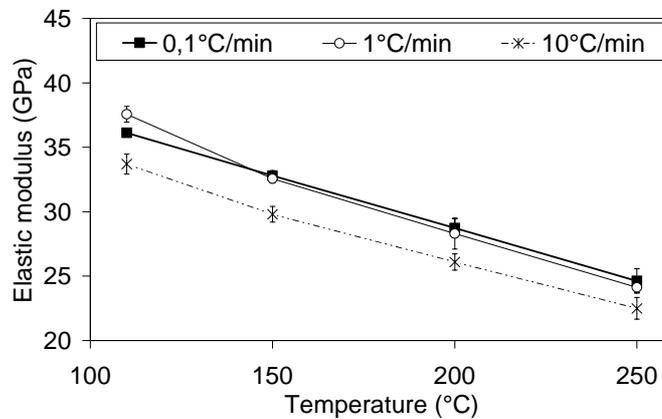


Fig. 7 Evolution of the elastic modulus with temperature and heating rate

Thermal conductivity

Concerning thermal conductivity it is essential to recall that this parameter is based on local measurements achieved with a quick thermal conductivity meter based on the principle of the heater wire. The conductivity discrepancy between the cement paste ($\approx 1\text{-}1.5\text{ W/m.C}^\circ$) and the silico-calcareous aggregates (2.5 W/m.C°) as well as the heterogeneity of the concrete itself can explain the important dispersion of the data. On average, between 110 and 250°C , thermal conductivity decreases by around 10% (Fig. 8). If we compare the 0.1 and 10°C/min rates data, the maximum relative discrepancy is about 3% . Thus, considering these tendencies and the experimental uncertainties, we can admit that there is no significant effect of the heating rate on the thermal conductivity measured on HSC specimens.

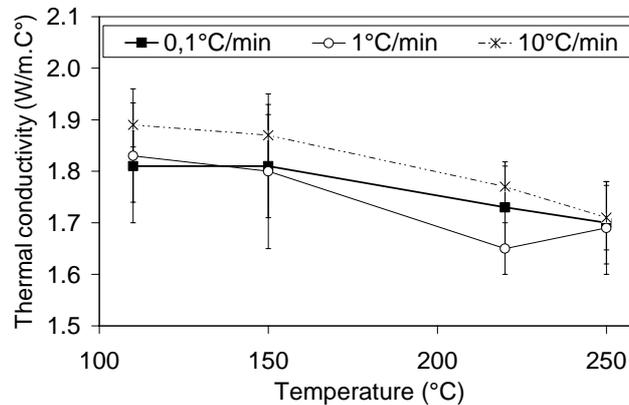


Fig. 8 Evolution of thermal conductivity with temperature and heating rate

DISCUSSION

The results obtained showed that the heating rate had a significant effect on some concrete residual properties. It should be underlined that the direction and the intensity of the variations depend on the parameter considered. On the other hand, for some parameters, data accuracy is probably of the same order of magnitude than the discrepancies we may observe. The impact of the heating rate on HSC residual properties remains difficult to analyse and is rather limited at specimen scale.

Mass loss data proved that the dehydration rate was independent from the kinetics with which the concrete probes were heated. This confirms that the quantity of water removed during thermal treatment is only controlled by the applied temperature. Slightly different conclusions were drawn in a previous study [11] dedicated to the behaviour of ordinary concrete (OC) and high-strength concrete heated up to 600°C . This study, showed that up to 350°C , the mass loss slightly increased with the decrease of the heating rate. But as concrete probes were heated during a relatively short time (one hour), it is possible that the mass loss was not totally completed.

Concerning other parameters, this work showed that a high heating rate (10°C/min) had less impact on permeability and compressive strength. On the contrary, the highest elastic modulus values were obtained with the lowest heating rate (0.1°C/min). Between the two extreme heating rates, the permeability varies by an average factor of 2.5. Compressive strength and elastic modulus relative variations were about 8 and 9 %, respectively. For thermal conductivity the maximum variation was about 3 %. No clear tendency was observed for tensile strength.

The evolution of compressive strength observed here meets the results of Noumowé's study [11] which showed that for OC and HSC heated at 350°C , the strength loss for the specimens heated at 0.1°C/min was higher than for those heated at 1°C/min (about 8.5 % relative variation). A similar work carried out on hardened paste and concrete [20] showed that between 200 and 600°C , the residual strength of a cement paste (0.67 w/c) heated at 3°C/min was lower than the one obtained when heated at 1°C/min . On the contrary, an opposite behaviour was observed for concrete (0.63 w/c). These results comply with previous works achieved by Koury [21], in which an interesting finding was that a concrete heated at 600°C at a slow rate of 0.2°C/min resulted in lower residual strength than that of an identical specimen heated at a faster rate of 1°C/min .

Further to these analyses, we can admit that the effect of the heating rate on concrete residual properties remains difficult to evaluate. In fact, the influence of this environmental factor surely depends on the material nature – hardened paste, concrete – on the constituents – aggregates – and also on temperature. In the case of concrete, the cement paste and aggregates thermal incompatibility and their differential behaviour at high temperature probably play a major role.

Moreover, it is admitted that concrete behaviour at high temperature depends on thermo-mechanical (related to thermal gradients) and thermo-hydric (related to water movements) processes. But the competition between the two types of processes and the impact on the material damaging still remains unclear. High heating rates generally lead to

higher thermal gradients and this is why low heating rates are often recommended for standard concrete thermal loading tests. On the other hand, some arguments show that the effects of low heating rates on the concrete response are not insignificant. This assumption was confirmed by the results of previous tests for which thermal gradients (Fig. 9) and mechanical strain (Fig. 10) were recorded on concrete probes during heating test up to 220°C with rates of 0.5 and 1°C/min. The thermal gradient was measured between the center and the surface of the specimens (normalised cylindrical "11x22" probes). The mechanical strain was measured at the surface of the probes using strain gauges. The highest thermal gradient – 6.3°C/cm ($\Delta T \approx 35^\circ\text{C}$) – was measured with the 1°C/min rate while for the 0.5°C/min heating rate the gradient was only 3.7°C/cm ($\Delta T \approx 21^\circ\text{C}$). On the contrary, the strain measured at the surface of probes was globally higher with the slow rate than with the high rate.

The present study showed that low heating rates can be responsible for a significant loss of property. A possible explanation is that for a low heating rate the water release through the porous network is slower. Consequently, the hydric stress related to internal vapor pressure may remain longer. Further experiments are planned to reach a more comprehensive understanding of this phenomenon.

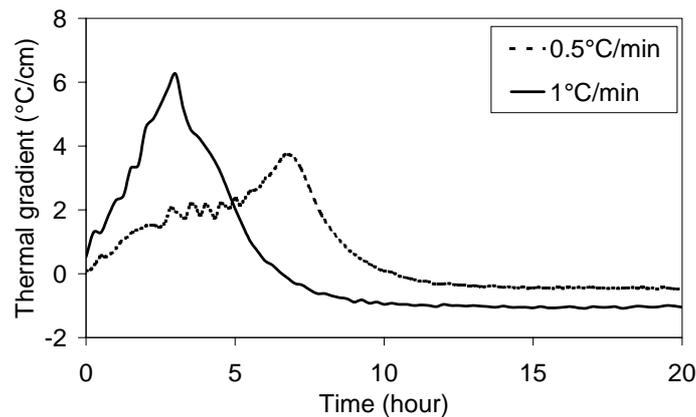


Fig. 9 Thermal gradient for HSC specimens heated at 220°C with 0.5 and 1°C/min rates

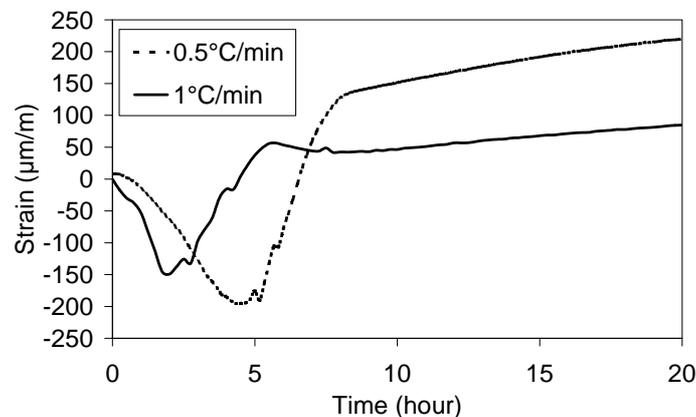


Fig. 10 Mechanical strain for HSC specimens heated at 220°C with 0.5 and 1°C/min rates

CONCLUSION

The behaviour of high-strength concretes at elevated temperatures and their residual properties, depend on many factors. Some are related to the characteristics of the material itself (e.g. aggregate, cement) ; others are environmental factors (e.g. temperature level, loading conditions, humidity, after cooling...). Both factors may greatly influence the experimental data. This study was the opportunity to investigate the impact of the heating rate on the physical and mechanical residual properties of a HSC submitted to temperatures up to 250°C. Taking into account the context of the long-term storage of nuclear HLW, several temperature levels were selected – 110, 150, 200 and 250°C – and three thermal loading rates (heating rates) were chosen, 0.1, 1 and 10°C/min. Results assessment provided interesting conclusions. Mass loss experiments showed that when the mass loss "plateau" is reached, the heating rate does not

influence the total water loss due to the thermal treatment. Concerning transport properties, gas migration tests showed that the highest permeability loss was observed when using the lowest heating rate (0.1°C/min). An average a 2.5 permability ratio was determined between the two extreme rate configurations (0.1 and 10°C/min). A similar tendency characterized the evolution of the compressive strength. This result is consistent with previous studies carried out on ordinary and high-strength concretes. On the contrary, an opposite trend was detected with the evolution of the elastic modulus for which the lowest residual values were obtained with the 10°C/min rate. For the compressive strength and the elastic modulus, the maximum relative variation induced by the heating rate effect was less than 10 %. We considered that tensile strength data did not show any clear tendency and that the effect of heating rate on thermal conductivity was extremelly limited (a few %).

This study showed that the effect of the heating rate on concrete physical and mechanical residual properies was significant but rather limited. But it also showed that low heating rates can be responsible for higher property loss than high heating rates. From the mechanical point of view (compressive strength above all), this result complies with conclusions of previous works in that domain. Additional experiments confirmed that during thermal loadings at high temperature (220°C), thermal gradients increase when the heating rate increase. On the contrary, mechanical strains are higher when using a low heating rate. A possible explanation for this phenomenon is that for a low heating rate the water release through the porous network is slower. Consequently, the hydric stress related to internal vapor pressure may remain longer. It is thus easy to understand that during a thermal treatment at high temperature, the thermo-mechanical and thermo-hydric processes taking place in the concrete, can counterbalance each other according to the heating rate. Further experiments with bigger and instrumented concrete probes are planed to reach a more comprehensive understanding of these phenomena in order to clarify the role of the heating rate on thermo-mechanical and thermo-hydric stresses.

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