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Scientific paper

Combined Effects of Temperature and Irradiation on Concrete Damage

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Abstract

Aggregate radiation-induced volumetric expansion (RIVE) is a predominant mechanism in the formation of mechanical damage in the hardened cement paste (hcp) of irradiated concrete under fast-neutron flux (Giorla *et al.* 2015). Among the operating conditions difference between test reactors and light water reactors (LWRs), the difference of irradiation flux and temperature is significant. While a temperature increase is quite generally associated with a direct, or indirect (e.g., by dehydration) loss of mechanical properties (Maruyama *et al.* 2014), it causes a partial annealing of irradiation amorphization of α -quartz, hence, reducing RIVE rate. Based on data collected by Bykov *et al.* (1981), an incremental RIVE model coupling neutron fluence and temperature is developed. The elastic properties and coefficient of thermal expansion (CTE) of irradiated polycrystalline quartz are interpreted through analytical homogenization of experimental data on irradiated α -quartz published by Mayer and Lecomte (1960). The proposed model, implemented in the meso-scale simulation code AMIE, is compared to experimental data obtained on ordinary concrete made of quartz/quartzite aggregate (Dubrovskii *et al.* 1967). Substantial discrepancy, in terms of damage and volumetric expansion developments, is found when comparing irradiation scenarios assuming constant flux and temperature, as opposed to more realistic test reactor operation conditions.

1. Introduction

The structural significance of irradiated concrete is of primary importance for extended long-term operation (LTO) of LWRs (Graves *et al.* 2014; Le Pape *et al.* 2014; Rosseel *et al.* 2014). Although historical data collected from the literature (Hilsdorf *et al.* 1978; Field *et al.* 2015) show a significant loss of mechanical properties correlated with RIVE at high neutron fluence (i.e., $> 10^{19}$ n.cm⁻²), the separation of the irradiation effects and thermal effects remains problematic. Indeed, irradiation experiments of concrete are typically performed in the range of 40 °C to 250 °C. Most irradiation experiments reaching fluences above 2×10^{19} n.cm⁻² were conducted at temperatures, even sporadically, above 100 °C (Field *et al.* 2015). Understanding the combined effects of temperature and irradiation on concrete is therefore required to estimate the correspondence between test reactor data and LWR conditions, i.e., with in-service temperature limited, by design, to 65 °C.

At the concrete scale, i.e., at the macroscopic scale, temperature affects the properties of unirradiated concrete, e.g., (Naus 2005, 2010), impacts the moisture transport kinetics (Luikov 1975) and, causes dimensional change. At the cement scale, i.e., at the microscopic scale,

rise in temperature leads to decomposition of the cement hydrates (Zhang and Ye 2012) and the development of cracking (Maruyama and Sasano 2014) due to differential deformations between the paste and the aggregates. Most aggregates are not subject to thermal damage below 300 °C.

Irradiation causes physical changes in the cement paste and the aggregates: (1) γ radiation induces radiolysis of water in the hcp, causing gas release and increasing shrinkage at high temperature (Elleuch *et al.* 1972), and, (2) neutron radiation induces amorphization of crystalline minerals contained in the aggregates, leading to very high RIVE as well as a loss of mechanical properties (Hilsdorf *et al.* 1978; Field *et al.* 2015).

However, the potential synergetic effects of temperature and irradiation on aggregate RIVE and CTE have received limited attention, although aggregate expansion (amplitude and rate) is a predominant effect in the formation of damage in irradiated concrete (Dubrovskii *et al.* 1967; Elleuch *et al.* 1972; Seeberger and Hilsdorf 1982; Le Pape *et al.* 2015; Giorla *et al.* 2015). RIVE is generally modeled as a function of the neutron fluence only, neglecting a potential contribution of the temperature. Studying combined temperature and irradiation effects requires a significantly large and well-documented database of irradiation test results conducted on the exact same material. In that regard, the variations of chemical composition within the broad classification of aggregates (e.g., limestone, granite, sandstone, etc...) introduce an interpretation bias. Additionally, the neutron kinetics energy spectrum or threshold, for a given experiment, is frequently not well-documented (Field *et al.* 2015), adding another possible interpretation difficulty.

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Neutron irradiation experiments of quartz at different temperatures have been solidly reported by Bykov *et al.* (1981). Dubrovskii *et al.* (1967) and Pedersen (1971) also measured the irradiated properties of concrete made with quartz aggregates. Notably, Dubrovskii *et al.* provide the temperature history for some of their samples, thus allowing a detailed analysis of that particular experiment. The data collected by Bykov *et al.* is interpreted in this article to develop a RIVE model for quartz accounting for temperature and neutron flux. This model is complemented with an analysis of the evolution of the mechanical properties of quartz as a function of its density, using experimental measurements by Mayer and Lecomte (1960) and analytic homogenization schemes. This provides the sufficient information to perform a preliminary numerical analysis of Dubrovskii *et al.* experiment with the mesoscale model previously developed by the authors in Giorla *et al.* (2015). Different scenarios in terms of neutron flux and temperature variations are simulated to identify the potential impact of these variables on the concrete RIVE and damage, and determine whether coupling between temperature and irradiation is required in further modeling efforts for irradiated structures.

2. A model for the effects of temperature and neutron irradiation on quartz expansion

Figure 1 presents the experimental volumetric expansion collected by Bykov *et al.* (1981) on quartz samples under neutron irradiation at temperature ranging from 20 °C to 600 °C. It is observed that with increasing temperatures, the expansion amplitude and rate decrease monotonically at any given fluence. The sigmoidal nature of the expansion curves remains unchanged. The final amplitude, i.e., $\approx 17.8\%$ at full amorphization, is not affected by temperature. “The relation between the value of the volume expansion and the degree of impairment of the lattice periodicity, according to the data of X-ray diffractometry [...] is independent of the irradiation temperature. This confirms that the type of defects originating and the nature of the damage caused are essentially unchanged with increase of temperature” (Bykov *et al.* 1981). The effects of temperature on quartz expansion is explained by the annealing of point defect during irradiation.

RIVE of quartz can be modeled by the empirical equation developed by Zubov and Ivanov (1966) under isothermal conditions:

$$\varepsilon^*(\Phi, T = cte) = \kappa \varepsilon_{\max} \frac{e^{\delta \Phi} - 1}{\varepsilon_{\max} + \kappa e^{\delta \Phi}} \quad (1)$$

The terms $\varepsilon_{\max}$ corresponds the maximum volumetric expansion, i.e., $\varepsilon_{\max} = 17.8\%$, for quartz. κ (dimensionless parameter homogeneous to a strain) and δ

(inverse of a fluence) govern the shape of the sigmoid. In particular, the inflexion point of the sigmoid, i.e., $\partial^2 \varepsilon^* / \partial \Phi^2 = 0$, corresponds to a fluence of $\ln(\varepsilon_{\max} / \kappa) / \delta$ and a swelling of $(\varepsilon_{\max} - \kappa) / 2$ (Le Pape 2015). At the same inflexion point, the rate of swelling is given by the expression $(\varepsilon_{\max} - \kappa) \delta / 4$. Hence, the main characteristics of Zubov's curve, except for the final amplitude of swelling, do not show a one-to-one dependency on Zubov's model parameters. Eq. (1) can also be interpreted in a slightly modified form, which corresponds, mathematically speaking, to the form adopted by several researchers, e.g., (Larive 1997; Saouma and Perotti 2006), to model the expansion of concrete subjected to alkali-silica reaction (ASR) as a function of time. Substitution of time by the fluence Φ leads to:

$$\varepsilon^*(\Phi, T = cte) = \varepsilon_{\max} \frac{1 - e^{-\frac{\Phi}{\Phi_c}}}{1 + e^{-\frac{\Phi - \Phi_L}{\Phi_c}}} \quad (2)$$

with $\Phi_c = 1/\delta$ the *characteristic fluence* that primarily governs the rate of expansion at the inflexion point of the sigmoid, and $\Phi_L = \ln(\varepsilon_{\max} / \kappa) / \delta$ the *latency fluence*, i.e., the fluence at the inflexion point, which is located at about half of the total expansion, when $\kappa \ll \varepsilon_{\max}$.

As shown in **Fig. 1**, Zubov and Ivanov's equation provides a good approximation of the trend lines plotted originally by Bykov *et al.* although it tends to overestimate the expansion rate above $\approx 15\%$ expansion.

Fig. 1 Volumetric expansion of neutron-irradiated quartz at different temperatures (Adapted and modified from (Bykov *et al.* 1981)). (a)-(k) indexes and symbol colors correspond to different ranges of temperature ranked by increasing order. (a): 25-30 °C; (b): 30-45 °C; (c): 40-50 °C; (d): 60-80 °C; (e): 80-85 °C; (f): 85-90 °C; (g): 90-100 °C; (h): 105-115 °C; (i): 120-160 °C; (j): 210 °C; (k): 240 °C. Source: $\square$ in APS-1 NPP reactor; ∇ in BR-5 test reactor; $\diamond$ in BR-10 test reactor. $\circ$ from literature (Wittels 1957; Primak 1958; Weissmann and Nakajima 1963). Dashed lines: Bykov et al. trend lines (no equations given). Solid lines: fit assuming Zubov and Ivanov's Eq. (1).

As noted by Bykov *et al.*, "the very strong temperature effect in the range of 30 °C to 100 °C and the reduction of this effect at a higher temperature indicates a nonuniformity of distribution of the defects with respect to the energy of activation, in particular, to a reduction of the fraction of defects with an increased energy of activation." Hence, instead of trying to interpret the data with the calibration of an apparent energy activation, temperature effects are analyzed directly on how Zubov's parameters are affected. While the plots of κ and δ (not shown here) exhibit a nonlinear relation with the temperature, the plots of the fluences Φ_c and Φ_L show a quasi linear evolutions against the temperature increase (Fig. 2). Best fit regressions provide the empirical relations:

$$\begin{aligned}\Phi_L &= a_L T + b_L \approx 2.40 \times 10^{-2} T - 6.26 \times 10^{-2} \\ \Phi_c &= a_c T + b_c \approx 2.77 \times 10^{-3} T + 0.128\end{aligned}\quad (3)$$

with T in °C and $\Phi_{L,c}$ in $\times 10^{20}$ n.cm⁻². From Eq. (3) the evolutions of $\kappa = \varepsilon_{\max} / \exp(\delta \Phi_L)$ and $\delta = 1 / \Phi_c$ with temperature can be derived, Eq. (6). Note that the default parameters suggested by Zubov and Ivanov (1966) for quartz, i.e., $\kappa = 0.27\%$ and $\delta = 4.5 \times 10^{20}$ n.cm⁻² correspond to a temperature of 34 to 41 °C assuming Eq. (3) holds, which is consistent with the irradiation temperatures from the experiments of Wittels (1957).

Figure 3 shows the validation plot of the proposed model against the experimental data, i.e., the data corresponding to marks in Fig. 1 at varied temperatures and fluences. Although good correlation between the experimental and modeled data is found, the scatter band is quite important because of the uncertainty on the actual irradiation temperature, the unreliability of Zubov's model at high expansions, i.e., above $\approx 15\%$, and the strong dependency of the model parameters to temperature.

Fig. 2 Evolution of the characteristic and latency fluences, respectively Φ_c and Φ_L as function of the irradiation temperature. ● correspond to the calculation of the fluences Φ_c and Φ_L derived from the fits of Bykov *et al.*'s trend lines (Fig. 1) using Zubov's Eq. (1). Linear fit equations (solid lines) provided directly with the plots.

From Eq. (2), the incremental variation of volumetric expansion at a given neutron fluence, temperature, neutron flux and temperature increment reads:

$$\frac{d}{dt} \varepsilon^*(\Phi, T) = \left. \frac{\partial \varepsilon^*}{\partial \Phi} \right|_T \cdot \frac{d\Phi}{dt} + \left. \frac{\partial \varepsilon^*}{\partial T} \right|_{\Phi} \cdot \frac{dT}{dt} \quad (4)$$

The isothermal RIVE rate is obtained by direct derivation from Eq. (1):

$$\left. \frac{\partial \varepsilon^*}{\partial \Phi} \right|_T = \kappa_T \varepsilon_{\max} \frac{\delta_T e^{\delta_T \Phi} (\varepsilon_{\max} + \kappa_T)}{(\varepsilon_{\max} + \kappa_T e^{\delta_T \Phi})^2} \quad (5)$$

with

$$\delta_T = \frac{1}{a_c T + b_c} \quad \text{and} \quad \kappa_T = \frac{\varepsilon_{\max}}{\exp\left(\frac{a_L T + b_L}{a_c T + b_c}\right)} \quad (6)$$

The second part of Eq. (4), i.e., in iso-neutronic condition ($\Phi = \text{cte}$), can be derived from post-irradiation thermal experiments (Mayer and Lecomte 1960; Primak 1958; Yano *et al.* 2007). Figure 4 shows the evolutions of the postirradiation expansion of quartz during an isochronal thermal heating up to about 1,000 °C at a rate of about 1,000 °Ch⁻¹ (Yano *et al.* 2007). For specimens irradiated at low fluence, annealing, or "self-healing", i.e., the evolution toward α -quartz accompanied by a reduction of the expansion to the initial value of quartz density is observed. Conversely, after irradiation at high fluence, anti-annealing is observed: quartz evolves toward more amorphous forms, called *cryptocrystalline* by Wittels, although different from vitreous silica unless fluence of the order of 10^{20} n.cm⁻² is reached (Primak

Fig. 3 Correlation between the modeled expansion and the measured expansion of quartz at varied fluences and temperatures. Experimental data from (Bykov *et al.*, 1981). Modeled data assuming Zubov's Eq. (1) and a linear dependence of the latency and characteristic fluences on the temperature (Fig. 2). ●: $\bar{T} < 65$ °C; ■: $\bar{T} \in [65; 110]$ °C; ◆: $\bar{T} \in [110; 180]$ °C; ◆: $\bar{T} > 180$ °C, with $\bar{T}$ the median temperature.

1958). Mayer and Lecomte (1960) estimate the dose threshold separating the annealing/anti-annealing behaviors at $\approx 0.65 \times 10^{19}$ n.cm⁻² at energy above 0.3 MeV (Keppens and Laermans 1996). Since limited volumetric change ($< 0.5\%$) is observed in the typical range of temperatures, i.e., < 400 °C, of test or commercial reactor operations, it appears legitimate to neglect the term $\delta\epsilon^*/\delta T|_{\Phi} \approx 0$ in Eq. (4), for those particular applications.

The temperature history depends on the test reactors operation, and can exhibit daily variation (e.g., JEEP-II in Norway) and quasi-monthly cyclic evolutions (e.g., BR-5 reactor in Russia). Accounting for temperature variations implies the estimation of the volumetric expansion $\epsilon_{i+1}^* = \epsilon_i^* + d\epsilon^*$ after a temperature step $dT = T_{i+1} - T_i$ during the time increment $d_t = t_{i+1} - t_i$. Since Bykov *et al.* observed that, at a given value of volume expansion, the degree of impairment of the lattice periodicity is independent of the irradiation temperature, it can be assumed that (1) The volumetric expansion is an *internal state variable* characterizing the degree of irradiation-induced damage. (2) The expansion rate, at a given time, appears to be a function only of the corresponding neutron flux and temperature, and not, a function of the histories of flux and temperature. Hence, the same volumetric expansion ϵ_i^* would have been obtained under an isothermal irradiation at temperature T_i to the equivalent fluence $\tilde{\Phi}_i$:

$$\tilde{\Phi}_i = \frac{1}{\delta_i} \ln \frac{\epsilon_{\max}^* (\epsilon_i^* + \kappa_i)}{\kappa_i (\epsilon_{\max}^* - \epsilon_i^*)} \quad (7)$$

The same reasoning can be applied at temperature T_{i+1} to derive the equivalent fluence $\tilde{\Phi}_{i+1}$:

Fig. 4 Post-irradiation annealing or anti-annealing of quartz. The fluence level before thermal treatment is given in brackets $\times 10^{20}$ n.cm⁻². ●: from dimensional change measurements (Primak 1958); □: from density measurements (Primak 1958); ◇: from dimensional change measurements (Yano *et al.* 2007).

$$\tilde{\Phi}_{i+1} = \frac{1}{\delta_{i+1}} \ln \frac{\epsilon_{\max}^* (\epsilon_{i+1}^* + \kappa_{i+1})}{\kappa_{i+1} (\epsilon_{\max}^* - \epsilon_{i+1}^*)} \quad (8)$$

and, as a consequence, the volumetric expansion rates at the beginning and the end of the time step read:

$$\left. \frac{\delta\epsilon^*}{\partial\Phi} \right|_{t_i} = \kappa_{i+1} \epsilon_{\max} \frac{\delta_i e^{\delta_i \tilde{\Phi}_i} (\epsilon_{\max} + \kappa_i)}{(\epsilon_{\max} + \kappa_i e^{\delta_i \tilde{\Phi}_i})} \quad (9)$$

and, the volumetric expansion increment is obtained by:

$$\left. \frac{\delta\epsilon^*}{\partial\Phi} \right|_{t_{i+1}} = \kappa_{i+1} \epsilon_{\max} \frac{\delta_{i+1} e^{\delta_{i+1} \tilde{\Phi}_{i+1}} (\epsilon_{\max} + \kappa_{i+1})}{(\epsilon_{\max} + \kappa_{i+1} e^{\delta_{i+1} \tilde{\Phi}_{i+1}})} \quad (10)$$

where ξ is a numerical parameter between 0 and 1 controlling the integration scheme, i.e., 0 for an implicit scheme, 1 for an explicit scheme or in between for any semi-implicit/explicit scheme – See Fig. 5.

3. Elasticity and thermal expansion of irradiated quartz

Quartz (aggregate) is a polycrystalline assemblage of randomly oriented α -quartz trigonal (rhombohedral) monocrystals. Hence, while α -quartz possesses anisotropic properties, polycrystalline quartz is an isotropic material and its properties can be derived by homogenization, e.g. by calculating Reuss-Voigt bounds or by using a self-consistent scheme. Under irradiation, α -quartz and vitreous silica, e.g., fused quartz evolve toward a similar isotropic structure caused by the disordering created by neutron irradiation (Wittels 1957; Primak 1958). When subjected to temperature increase, α -quartz is relatively stable until phase transformation to β -quartz (hexagonal) at 573 °C (atm. pressure) is reached.

Fig. 5 Derivation of the incremental volumetric expansion accounting for temperature variations.

3.1. Elastic properties

Trigonal α -quartz is characterized by 6 independent elastic constants, i.e., c_{ij} stiffness coefficients, including a non-zero c_{14} term, while isotropic silica only requires the determination of 2 constants. The values of c_{ij} for varied polymorphs of crystalline silica and silica glass have been obtained experimentally in the temperature range of 0 °C to $\approx 1,000$ °C by several authors, e.g., see review articles (Ballato 2008; Pabst and Gregorová 2013).

Elastic constants of irradiated α -quartz and silica have been obtained by Mayer and Gigon (1956); Mayer and Lecomte (1960); Zubov and Ivanov (1967) in the corresponding range of RIVE below $\approx 6\%$, i.e., at about a third of the maximum expansion. Tentative extrapolation for RIVE beyond 6% is discussed hereafter. The elastic properties of polycrystalline quartz can be theoretically derived by homogenization, i.e., Voigt-Reuss bounds (Voigt 1889; Reuss 1929), or self-consistent scheme (Dormieux *et al.* 2006). The results can either be plotted as a function of RIVE or density. While the implementation in a finite element frame (See section 5) is done considering properties function of the volumetric expansion (internal variable), plotting the irradiated properties of both α -quartz and silica as functions of the density make it possible to evaluate the possibilities of extrapolation beyond the study range, assuming with Mayer and Lecomte, that at full amorphization, α -quartz and silica evolve toward the same disordered state, i.e., of equal densities.

Young's modulus of irradiated α -quartz.

Figure 6 shows that, in the range of studied density, i.e., 2.55 to 2.65, irradiation affects the Young modulus of quartz although no monotonic decreasing or increasing trend can be observed. Irradiated Young modulus remains in the range of 90 to 100 GPa. Under the same irradiation conditions, the Young modulus of silica

Fig. 6 Young modulus of irradiated α -quartz and silica as a function of the irradiated materials density. (s) amorphous silica; (r) irradiated quartz, Reuss bound; (v) irradiated quartz, Voigt bound; (SC) irradiated quartz, self-consistent scheme.

slightly increases from 73 GPa to 76 GPa. Hence, there is still a significant gap between the Young moduli of α -quartz and silica at the end of Mayer and Lecomte's experiments preventing any extrapolation. It appears that Voigt-Reuss bounds tend to narrow with irradiation. This suggests that the behavior of α -quartz crystals evolves, with irradiation, towards isotropy.

Poisson's ratio of irradiated α -quartz.

The Poisson ratio of irradiated α -quartz, ν^* , shows a rapid increase from its initial value $\nu_0 = 0.08$ to reach about 0.18 at $\approx 4.5\%$ volume expansion - See **Fig. 7**. In this range of RIVE, the Poisson ratio estimated by the selfconsistent scheme is fitted by an exponential function. In the same range of fluence, the Poisson ratio of silica increases from 0.17 and seem to level off at about $\nu_s^* \approx 0.21$, which will be considered here, as the maximum limit value of irradiated α -quartz at full amorphization - See **Fig. 8**. The proposed evolution of irradiated α -quartz Poisson ratio reads:

$$\nu^* = \min\left(1 - \nu_0 + \exp\left(a(\varepsilon^*)^b\right); \nu_s^*\right) \quad (11)$$

with a and b two fit parameters found equal to 0.0063 and 1.938.

3.2. Coefficient of thermal expansion

The linear CTE of α -quartz at about 20 °C varies with the crystallographic orientation: $\approx 7.4 \mu\text{mm}^{-1} \text{ } ^\circ\text{C}^{-1}$ ($\parallel_c$, i.e., optical axis) and $\approx 1 \mu\text{mm}^{-1} \text{ } ^\circ\text{C}^{-1}$ ($\perp_c$) (Amatuni and Shevchenko, 1966). Similar values are found in natural quartz in the range of -20 °C to 60 °C (Johnson and Parsons 1944, nat. quartz, Minas Geraes, Brazil), i.e., $\tilde{\alpha} = 12 \mu\text{mm}^{-1} \text{ } ^\circ\text{C}^{-1}$. In the range of ≈ 20 °C to 350 °C, quartz CTE can be approximated by a linear law $\tilde{\alpha}(T) \approx 11.2 + 10.9 \times 10^{-3}T$, with $\tilde{\alpha}$ in $\mu\text{mm}^{-1} \text{ } ^\circ\text{C}^{-1}$ and T in °C

Fig. 7 Poisson ratio of irradiated α -quartz and silica as a function of the irradiated materials density. (s) amorphous silica; (r) irradiated quartz, Reuss bound; (v) irradiated quartz, Voigt bound; (SC) irradiated quartz, self-consistent scheme.

Fig. 8 Poisson ratio of irradiated α -quartz as a function of RIVE. Gray-colored portion: Voigt-Reuss bounds. Dashed line: self-consistent scheme. Thick solid line: Suggested fit with Eq. (11).

(Amatuni and Shevchenko, 1966) (see Kosinski *et al.* (1991) for a Taylor expansion of the CTE).

However, fused-quartz has a much lower CTE, $\approx 0.42 \mu\text{m m}^{-1} \text{ } ^\circ\text{C}^{-1}$ at 25°C (Beattie *et al.* 1941). It increases nonlinearly up to $0.5\text{--}0.6 \mu\text{m m}^{-1} \text{ } ^\circ\text{C}^{-1}$ at $\approx 200^\circ\text{C}$ before decreasing monotonically toward 0.42 to $0.54 \mu\text{m m}^{-1} \text{ } ^\circ\text{C}^{-1}$ at $\approx 1,000^\circ\text{C}$. The temperature of the peak value varies with the authors: 200 to 400°C (Otto and Thomas 1963). Also, α -quartz, obtained after phase transformation at 573°C (atm. pressure), exhibits a very low slightly negative CTE (Welche *et al.* 1998). Hence, it can be conjectured that irradiation-induced amorphization affects the CTE of quartz toward decreasing. The CTE of α -quartz at 20°C shows little modification after irradiation for fluences up to $2.2 \times 10^{19} \text{ n.cm}^{-2}$ (Mayer and Gigon 1956). Above that fluence value, Mayer and Lecomte (1960)'s post-irradiation CTEs (average value between -190°C to 0°C) show a sharp decrease for RIVE above $\approx 10\%$ of the maximum expansion. **Figure 9** shows that the CTE of α -quartz and silica appear to be reaching similar values at the end of Mayer and Lecomte's experiments. Interestingly, the CTE of irradiated silica shows a decreasing trend in the negatives. Note, however, that post-irradiation measurements of CTEs (-196°C to 20°C) on varied limestone, dolerite, andesite and hornfels at fluence above $10^{19} \text{ n.cm}^{-2}$ (fast neutron) show either little variation, moderate increase or large increase (dolomite) compared to the pre-irradiation CTEs (Kelly *et al.* 1969; Gray 1971). Hence, generalization to other-than-quartz aggregate is subject to caution.

Based on the observed trend (**Fig. 10**), the CTE of irradiated polycrystalline quartz (average value between -190°C to 0°C), α^* obtained with a self-consistent scheme can be fitted by a sigmoidal function similar to Eq. (2):

Fig. 9 Coefficient of thermal expansion of irradiated α -quartz and silica as a function of the irradiated materials density. (s) amorphous silica; (r) irradiated quartz, Reuss bound; (v) irradiated quartz, Voigt bound; (SC) irradiated quartz, self-consistent scheme.

Fig. 10 Coefficient of thermal expansion of irradiated α -quartz as a function of RIVE. Gray-colored portion: Voigt-Reuss bounds. Thick solid line: Suggested fit with Eq. (12).

$$\alpha^* = \alpha_0^* + \alpha_1 \left(1 - \frac{1 - e^{-\varepsilon^* / \varepsilon_c^*}}{1 + e^{-(\varepsilon^* - \varepsilon_L^*) / \varepsilon_c^*}} \right) \quad (12)$$

with ε_c^* and ε_L^* , two fitting coefficients found equal to 0.85% and 4.27% , respectively. The choice of the fitting function is arbitrary. ε_0^* corresponds to the CTE of fully-irradiated quartz or silica, i.e. $\approx -0.52 \times 10^{-6} \mu\text{m m}^{-1} \text{ } ^\circ\text{C}^{-1}$. $\alpha_1 + \alpha_0^*$ is the value of the pristine α -quartz CTE, i.e., $\tilde{\alpha}(T)$ assuming that Eq. (12) holds beyond the temperature range studied by Mayer and Lecomte. After fitting, α_1 is found equal to $8.63 \mu\text{m m}^{-1} \text{ } ^\circ\text{C}^{-1}$. Because the amplitude variation of fused quartz in the temperature range of 20°C to 400°C quite limited, i.e.,

Table 1 Irradiation parameters and volumetric expansion (assumed as the cube of the measured diameter expansion) for Dubrovskii *et al.*'s data. Table assembled from (Dubrovskii *et al.*, 1967, Original Tab. 1 & 3). † : estimated values.

	Group No.			
	1	2	3	4
integrated flux ($\times 10^{20}$ n.cm $^{-2}$)	3-3.3	2-2.8	1.2-1.45	0.4-0.6
maximum flux ($\times 10^{13}$ n.cm $^{-2}$ s $^{-1}$)	1.9-2.1	1.3-1.8	0.75-0.9	0.25-0.4
average flux ($\times 10^{13}$ n.cm $^{-2}$ s $^{-1}$)		0.62 [†]		
maximum heat deposition (Wcm $^{-3}$)	0.19-0.21	0.12-0.17	0.07-0.085	0.025-0.04
maximum temperature (°C)	300-350	225-275	100-150	20-50
average temperature (°C)	115 [†]	89	57 [†]	35 [†]
concrete vol. expansion (%)	16.2	>17.4	9.3	2.4
	> 19.2	>18.9	5.1	1.5
	> 17.7	>15.9	5.7	
		> 18.0		

0.4-0.6 $\mu\text{m m}^{-1} \text{°C}^{-1}$, and, remains one order of magnitude below that of pristine α -quartz CTE, the effects of temperature on α_0^* can be neglected ($\alpha_0^* \approx 0.5 \mu\text{m m}^{-1} \text{°C}^{-1}$).

4. Interpretation of irradiation experiments on quartz aggregate concrete

While quartz and quartzite sand is commonly used in concrete, coarser aggregate are usually made of other rocks, e.g., limestone, granite, basalt, etc. . . Hence, irradiation experiments on concrete are rarely conducted with pure quartz aggregate. We found only to sets of irradiation experiments on quartz aggregate concrete in the literature (Dubrovskii *et al.* 1967; Pedersen 1971). The details provided by Pedersen are unfortunately quite limited to envision a thorough interpretation through modeling. Dubrovskii *et al.* (1967) tested the effects of neutron irradiation of ordinary concrete (density 2.31) cylinder of 40 mm in height and diameter, made of Portland cement (w=c = 0.5). The mix contains quartz river sand (0.15 to 0.6 mm, c : a = 1 : 1.7) and sandstone gravel (5 to 10 mm, c : a $\approx$ 1 : 3). The volume fraction of quartz sand is about 25%. The total volume fraction of aggregate is close to 70%. Using the chemical composition of the mix provided by Dubrovskii *et al.*, and assuming a typical oxide composition for the Portland cement, we found that the molar composition of the sandstone is about 31% of silicon and 66% of oxygen, proving the highly-siliceous nature of the tested sandstone containing also traces of calcium, iron, magnesium and aluminum, supporting Dubrovskii *et al.* statement that “the total quartz content in this concrete was 70%.”

Table 1 presents the irradiation fluxes, the temperature, and the post-irradiation volumetric expansions for the four tested groups.

Quasi-linear correlations between the maximum flux, Φ_{max} , the maximum heat deposition and maximum temperature, T_{max} can be observed ($\Phi_{\text{max}} \approx 5.83 \times 10^{-3} T_{\text{max}} + 0.106$ with T_{max} in °C and Φ_{max} in $\times 10^{13}$

n.cm $^{-2}$ s $^{-1}$). Assuming that the found linear correlation is applicable in the range of temperatures corresponding to the BR-5 reactor operation, the flux history of group 2 experiments, plotted in Fig. 11, can be derived from the history of temperature provided in (Dubrovskii *et al.* 1967, original Fig. 1). Note that kinetic energies above 10 keV account for about 99.5% of the total energy (See energy distributions in (Dubrovskii *et al.* 1967, Table 2., p 1054)). Hence, the empirical parameters a_c , b_c , a_L , and b_L , derived from the interpretation of Bykov *et al.*'s data are assumed valid to model Dubrovskii *et al.*'s data. The average temperature and average flux calculated from these plots are 89 °C and 6.2×10^{12} n.cm $^{-2}$ s $^{-1}$, i.e., respectively, 2.5-3 times, and 2-2.5 times, lower than the maximum temperature and flux. Figure 12 illustrates the potential implication of different temperature and flux scenarios in terms of volumetric expansion evolutions of quartz, assuming the proposed model hold - Eqs. (2- 10): (a), i.e., the “reference scenario”: variable temperature

Fig. 11 Estimated time-evolution of the integrated energy flux (black solid line) and fluence (thick solid red line) over time - Group 2. Dashed line: fluence evolution assuming a constant flux at 6.2×10^{12} n.cm $^{-2}$ s $^{-1}$. The error bar at 501 d corresponds to the interval of fluences provided in Table 1.

(Dubrovskii *et al.* 1967, original Fig. 1) and variable flux (Fig. 11); (b): constant temperature (89 °C) and variable flux; (c): constant temperature and constant flux 6.2×10^{12} n.cm⁻² s⁻¹; and, (d): variable temperature and constant flux. At the end of the experiment, the estimated RIVE ranges between 14.7% and 17.8%. However, important scatter can be observed for times around 300 d: while scenario (b) leads to almost full expansion, i.e., full amorphization, scenario (d) leads to a RIVE under 2%. The scenario (c) assuming constant temperature and flux underestimates the volumetric expansion by <7%. Hence, the total fluence received during irradiation is not a sufficient parameter to describe the experiment. At high fluence (groups 1 and 2), RIVE was large enough to fill the space between the concrete sample and the stainless steel capsule wall. When excessive swelling occurred, the expansion data are presented in Table 1 with a > sign, since the measurements were taken after the extraction of the wedged samples. It can be observed that the volumetric expansions, calculated assuming a factor 3 on the diameter change, are close to the maximum expansion of quartz, i.e., 17.8%, for the samples from groups 1 and 2.

Dubrovskii *et al.* noted that “the nature of temperature variations for samples in the first and third groups was the same” (as the second group). Hence, the same temperature and flux histories can be used for groups 1 to 3. An approximation of the concrete expansion, ε_c , can be derived from the aggregate expansion, ε^* and its volume fraction, f_a , for important level of expansion leading to important damage in the hcp (Le Pape *et al.* 2015) (micromechanical theory):

$$\varepsilon_c \approx \frac{2f_a}{1+f_a} (\varepsilon^* + 3\tilde{\alpha}\Delta T_{\max}) \quad (13)$$

The term $3\tilde{\alpha}\Delta T_{\max}$ correspond to the maximum thermal volumetric expansion of aggregate during the irradiation experiment. Eq. (13) assumes that the cement paste suffers extensive micro-cracking, up to the point that it has no elastic stiffness. The strains in the cement paste (either due to shrinkage or thermal deformations) have therefore no influence over the concrete homogenized expansion.

In Dubrovskii *et al.*'s experiments on group 2, RIVE of $0.10\varepsilon_{\max}$ is reached approximately around 150 d (scenario (a) in Fig. 12). The maximum temperature reached during this phase is about 150 °C. Hence, the term $3\tilde{\alpha}\Delta T_{\max}$ can be estimated by $3 \times 1.2 \times 10^{-5} \times 130 \approx 0.5\%$. As a consequence, for this particular set of experiments, the thermal expansion of aggregate during irradiation can be neglected in comparison to RIVE. For a volume fraction of 70%, the expansion transfer coefficient to concrete, $2f_a/(1+f_a)$, equals 0.82. Hence, the maximum theoretical concrete expansion is $0.82 \times 17.8\% = 14.6\%$ while the measured concrete expansion for samples in groups 1 and 2 are around 17.8%, i.e., the maximum expansion of quartz. Such discrepancy may be explained by: (1) biased diameter change measurement caused by nonuniform swelling, as suggested by (Dubrovskii *et al.* 1967, Photograph of sample No. 8 in Fig. 3), resulting from the aggregate distribution in the sample, (2) displacement field discontinuities induced (micro-) cracking. This motivates the development of a

Fig. 12 Time-evolution of the volumetric expansion of quartz for group 2 specimens assuming different temperature and flux scenarios. (a): variable temperature (Dubrovskii *et al.* 1967, original Fig. 1) and variable flux (Fig. 11); (b): constant temperature (89 °C) and variable flux; (c): constant temperature and constant flux 6.2×10^{12} n.cm⁻² s⁻¹; and, (d): variable temperature and constant flux. The black solid line corresponds to the flux and temperature correlated histories.

Fig. 13 Time-evolution of the volumetric expansion of concrete for group 2 specimens assuming a variable temperature (Dubrovskii *et al.* 1967, original Fig. 1) and variable flux (Fig. 11). The boxes indicate the ranges of maximum fluences and expansions provided by Dubrovskii *et al.* (data in Table 1). Dashed lines indicate the uncertainty of the expansion measurement because the irradiated samples were wedged in the capsule (see text).

numerical model, as analytical micromechanical models have limited capabilities to capture these effects.

5. Numerical modeling

We simulate the experiments of Dubrovskii *et al.* (1967) with the model previously developed by the authors for irradiated concrete. The material is considered at the meso-scale with a two-dimensional finite element representation of its microstructure. The model calculates the deformation as well as the localization and extent of the micro-mechanical damage in the sample, accounting for the different properties and behaviors of the cement paste and the aggregates.

Post-irradiation deformations are the only experimental result available for this set of temperature and fluences, as Dubrovskii *et al.* were unable to measure mechanical properties due to the samples being wedged in the irradiation rig. The model provides an estimation of the history leading to that deformation. Rigorous validation of the simulation results cannot be achieved due to the absence of monitoring during the experiments and the uncertainties related to the data provided by Dubrovskii *et al.*. The principal purpose of the simulations is to give insights on the influence of the temperature and neutron flux variations on the expansion and damage variations, and assess the importance of considering combined or separated effects of temperature and irradiation in further modeling efforts.

The main components of the model are summarized below, and described in greater details in Giorla *et al.* (2015). The model is implemented in the C++ finite element framework AMIE, which was initially developed in the context of mesoscale simulations of ASR (Dunant and Scrivener 2010), and later applied for the study of creep (Giorla *et al.* 2014) and irradiation (Giorla *et al.* 2015).

5.1. Microstructure

We simulate a two-dimensional vertical slice of a concrete cylindrical sample. Aggregates are represented with circles randomly placed in a square box. Dubrovskii *et al.* used two different aggregate classes in their experiments: sand (0.15-0.6 mm) and gravels (5-10 mm), without intermediate sizes. In the simulation, only gravels are accounted for, to avoid using a very fine finite element mesh. While sand is not explicitly accounted for in the simulation, the surface fraction of gravels is increased to 58% in order to partly compensate for its absence (It is reminded that the actual aggregate volume fraction is 70% in Dubrovskii *et al.*'s experiment). The particle size distribution follows a Füller curve between the given minimum and maximum gravel diameters in absence of further data (Fuller and Thompson 1907).

The RIVE of the missing sand fraction is neglected for this set of simulations. The properties of the cement paste could be adapted in order to account for that additional expansion. However, to do so, one would need the ex-

pansion and degradation of the sand and cement paste mixture not only as a function of the fluence and temperature, but also as a function of the level and direction of the current stress. Mechanical restraint is likely to affect the relation between expansion and degradation, notably making them both anisotropic as it is the case in similar phenomena like ASR (Larive 1997; Multon and Toutlemonde 2006; Dunant and Scrivener 2012). The only experiment of neutron radiation under load in the literature (Gray 1971) lacks the sufficient supporting data and control measurements to be conclusive in that respect. This effect is therefore neglected here due to lack of knowledge about irradiation expansion under load.

The interfacial transition zone (ITZ) between the aggregates and the cement paste is also neglected. Assigning cohesive properties to the ITZ might control the debonding between the cement paste and the aggregate (e.g., Maruyama and Sugie (2014)). However, mechanical properties of the ITZ need to be calibrated on experimental data, which were not readily available for this experiment. In particular, there is no available data in the open literature on the effects of irradiation on the ITZ bonding properties.

The microstructure is discretized with a conforming unstructured mesh using a Delaunay triangulation. The calculations were run on a mesh containing $\approx 14,000$ linear elements. The two-dimensional representation was chosen as the model does not present strong anisotropy, in terms of mechanical properties, loadings, or geometry. However, it is recognized that this hypothesis might lead to an earlier crack percolation compared to a three-dimensional analysis.

The displacements of the bottom side of the sample are fixed in the vertical direction. The central node of the bottom side is fixed in the horizontal direction to prevent global sliding of the sample. The microstructure and mechanical boundary conditions are presented in **Fig. 14**. The temperature, relative humidity and neutron fluence fields are assumed homogeneous through the sample. The four cases detailed in section 4 (See also **Fig. 12**) are simulated: (a) variable temperature and flux, (b) variable temperature and constant flux, (c) constant temperature and variable flux, and, (d) constant temperature and flux.

5.2. Constitutive behaviors

Cement paste. The hcp is considered as a viscoelastic material with isotropic linear damage and thermal expansion (with α_p the coefficient of thermal expansion).

The creep deformation is divided into two components: a short-term recoverable creep, modeled with a Kelvin-Voigt unit, and a long-term irrecoverable viscous flow, modeled with a time-dependent dashpot placed in series (see **Fig. 15**). With this model, the long-term creep behavior of the paste follows a logarithmic function of time. The model relies on four parameters: the Young modulus E_p , the Poisson ratio ν_p , the creep viscosity η_p (measured as the slope of the creep curve in the logarithmic scale), and the creep characteristic time τ_p .

Fig. 14 Concrete microstructure with aggregates in black, cement paste in grey.

(which controls the instant at which the logarithmic behavior starts). Temperature accelerates creep with an Arrhenius-type law (with T_{act} the activation temperature), while relative humidity reduces the amplitude of the creep deformation (with h_c a coefficient controlling the loss of creep with relative humidity). Drying creep is neglected in the current model, as the samples in Dubrovskii *et al.* experiments have probably been pre-dried considering the reported weight loss. As the method of pre-drying (if any) was not reported by Dubrovskii *et al.*, no assumptions is made on the potential loss of mechanical properties and additional shrinkage due to drying.

The damage is simulated with a non-local isotropic linear damage adapted from the work of Dunant and Bentz (2015). The stress-strain curve of the material presents a linear softening branch after the peak (see Fig. 15). This behavior is characterized by the strength at the peak $f_{t,p}$ and the ultimate strain at the end of the fracture process $\varepsilon_{y,t,p}$. Failure in compression is neglected, as most of the damage will be caused by the very high tensile strains induced by the RIVE.

Aggregates. Aggregates are considered as purely elastic with thermal expansion and RIVE. The aggregates are furthermore considered homogeneous with no internal defects. The failure behavior of the aggregates is

Table 2: Constitutive parameters used in the simulations. (1) Based on creep experiments for cement paste by Le Roy (1995). (2) Assumption based on typical values for cement paste and concrete. (3) Based on tensile strength experiments for cement paste by Wittmann (1968). (4) Typical value for concrete recommended by Bažant and Steffens (2000). (5) Based on creep experiments for cement paste by Wittmann (1970). (6) Value for non-irradiated quartz obtained using the self-consistent scheme on experimental data by Mayer and Lecomte (1960). (7) Final value for the RIVE of quartz measured by Bykov *et al.* (1981).

Cement paste		
E_p [GPa]	12	(1)
ν_p [-]	0.2	(2)
α_p [$\mu\text{m m}^{-1} \text{ }^\circ\text{C}^{-1}$]	10	(2)
$f_{t,p}$ [MPa]	6	(3)
$\varepsilon_{y,t,p}$ [mm m^{-1}]	0.75	(2)
r_{nl} [mm]	0.3	(2)
η_p [GPa d]	30	(1)
τ_p [d]	20	(1)
T_{act} [K]	5,000	(4)
h_c [-]	0.2	(5)
Aggregates		
E_a [GPa]	95.7	(6)
ν_0 [-]	0.08	Eq. 11
ν_s^* [-]	0.21	Eq. 11
a [-]	0.0063	Eq. 11
b [-]	1.938	Eq. 11
ε_{max} [%]	17.8	(7)
a_L [$10^{20} \times \text{ncm}^{-1} \text{ }^\circ\text{C}^{-2}$]	0.024	Eq. 3
b_L [$10^{20} \times \text{ncm}^{-1}$]	-0.062	Eq. 3
a_c [$10^{20} \times \text{ncm}^{-1} \text{ }^\circ\text{C}^{-2}$]	2.77×10^{-3}	Eq. 3
b_c [$10^{20} \times \text{ncm}^{-1}$]	0.128	Eq. 3
ξ [-]	0	Eq. 10
α_0^* [$\mu\text{m m}^{-1} \text{ }^\circ\text{C}^{-1}$]	-0.52	Eq. 12
α_1^* [$\mu\text{m m}^{-1} \text{ }^\circ\text{C}^{-1}$]	8.63	Eq. 12
ε_c^* [%]	0.85	Eq. 12
ε_L^* [%]	4.27	Eq. 12

neglected as the compressive stresses induced by the RIVE are well below the material strength. This assumption may not be valid for heterogeneous aggregates with internal differential RIVE.

The mechanical and physical properties of the aggregates are taken as functions of the advancement of the irradiation process, as measured by the current level of RIVE, Eq. (11)-(12).

The model uses the classical hypothesis of infinitesimal strain. This hypothesis may not be valid for the later stages of the degradation, where the strains exceed one percent.

Fig. 15 Rheological model for the creep of cement paste (top), typical creep curves for a loading at $t = 0$ showing the influence of the different components of the models (using arbitrary parameter values) (middle) and stress-strain curve for a paste element (bottom). On the creep curve E can be read from the initial deformation, η from the asymptotic slope in the logarithmic scale, and τ as the intersection between the initial and final asymptotes.

5.3. Material properties

The material properties of each phase are gathered in Table 2. Notations are explained in greater details in (Giorla *et al.* 2015).

Cement paste. The material properties are taken from experimental results in the literature for cement pastes with a water/cement ratio of 0.5 as in Dubrovskii *et al.* experiments whenever possible.

The elastic and creep properties at ambient temperature are provided from the basic creep experiments on sealed cement paste samples with a water/cement ratio of

Fig. 16 Evolution of the Young's modulus and tensile strength of the cement paste as a function of the maximum temperature reached in an element, following experimental results by Fu *et al.* (2004).

0.5 by Le Roy (1995). The Arrhenius coefficient for creep is taken equal to 5,000 K as suggested in the commonly used B3 model (Bažant and Steffens 2000). The creep properties are reduced by a factor calibrated on Wittmann (1970) creep experiments on pre-dried sealed cement paste samples with a water/cement ratio of 0.4.

The tensile strength of the material is adapted from values given by Wittmann (1968) for dried cement paste samples with a water/cement ratio of 0.45 and 0.6.

The radius of the non-local averaging r_{nl} is related to the characteristic length of the heterogeneities in the material (here, the cement paste) (Bažant and Pijaudier-Cabot 1989), and is therefore taken as the radius of the largest sand particle neglected in the model. The elastic modulus and tensile strength of a cement paste element are reduced with the maximum temperature reached in that element following the experimental results of Fu *et al.* (2004) obtained on thin cement paste samples with a water/cement ratio of 0.5 (see Fig. 16). The ultimate strain at rupture is dynamically changed through the simulation to keep the fracture energy of each element constant in absence of further data.

Aggregates. The mechanical properties of the aggregates are taken as function of the irradiation, as described in the previous sections. They are assumed to be independent of the temperature in the temperature range recorded during the experiments. The Young's modulus of the quartz is taken as constant due to lack of data for higher fluences. In a previous study (Giorla *et al.* 2015), the authors found that the Young's modulus of the aggregate had no significant impact on the damage formation and propagation in the hcp during irradiation.

5.4. Results and discussion

Two main outputs are derived from the simulations: (1)

the expansion of concrete, measured from the maximum displacements variation between the opposite edges of the sample, and, (2) the damage of the cement paste, measured as the surface average of the damage scalar variable over the elements representing the cement paste. The corresponding results are plotted as a function of the fluence on **Figs. 17 and 18**, respectively. The simulations at variable temperature show an earlier increase of the expansion and damage as a function of the fluence. When the temperature is maintained constant, the expansion and damage do not seem to depend on the neutron flux, as both curves (constant and variable flux) coincide. All simulations reach a plateau value in term of damage, $\approx 65\%$, which corresponds to the loss of integrity of the sample: at that point the cement paste provides no mechanical restraint and the aggregates are free to swell.

Expansion. The volumetric expansion reaches values between $\approx 15\%$ and $\approx 18\%$ depending on the studied scenarios, which is consistent with the experimental observations from Dubrovskii *et al.*, considering that the difference of actual aggregate volume fraction in the experiment (0.70) and the numerical model (0.58). The final expansion is about 30% higher than the integral of the average strains in the sample, the later corresponding to Eq. (13) assuming a volume fraction of aggregate of 58%.

This difference is explained by the severe micro-cracking and heterogeneities of the sample micro-structure, notably induced by the presence of large aggregates near its surface. It should be noted that gradient effects are not considered in the present model, and may further increase this difference. This has a direct practical implication for the design of concrete irradiation experiments, as it indicates that the maximum swelling of a concrete sample might be higher than what would be

Fig. 17 Volumetric expansion of the concrete as a function of fluence for the Group 2 at variable temperature and neutron flux (thick solid line), constant temperature (solid line), constant flux (dashed line), and constant temperature and flux (dotted line).

expected from analytical models. The gap between the sample and the irradiation capsule wall should account for this effect in order to avoid wedging the samples into the capsule, as what happened in Dubrovskii *et al.* experiments.

Damage. Damage in the cement paste (**Fig. 18**) increases rapidly in the sample after initiation. The neutron fluence at which this rise occurs is significantly reduced when the complete history of temperature is simulated. Furthermore, the final stage of the damage propagation (from 50% of damage in the cement paste to 65%) is much sharper in the case of variable temperature than constant temperature, as if it ranged from stable propagation (constant temperature) to unstable (variable temperature).

A closer analysis to the first damage increase reveals that, when considering a variable temperature and neutron flux, the initial damage increase occurs during the first cooldown of the reactor, as shown in **Fig. 19**. During that cooldown, the flux remains strictly positive due to the hypothesis of linearity between flux and temperature (see section 4): the small increase in neutron fluence, coupled with the low temperature, leads to a sharp increase of the RIVE. This behavior is also observed, albeit with a smaller extent, during the second cooldown. Note that this effect is not a modeling artifact as quasi-identical results are obtained when assuming a neutron zero flux for temperatures below 30°C , i.e., considering a complete reactor shutdown.

Fig. 18 Average damage in the cement paste as a function of fluence for Group 2 at: (a) variable temperature and neutron flux (thick solid line), (b) constant temperature (solid line), (c) constant flux (dashed line), and, constant temperature and flux (dotted line). The microstructures show the damage in the cement paste (in grey level: black corresponds to a damage of 1.0; aggregates are represented in blue) at 10%, 25% and 50% damage in the cement paste for the case at constant temperature and neutron flux.

Fig. 19 Average damage in the cement paste as a function of time for the Group 2 at variable temperature and neutron flux (thick solid lines) and flux (dashed). Temperature variations in red.

The difference between simulations at variable or constant temperature has a critical impact on the analysis of degradation of concrete structures exposed to irradiation. Indeed, it indicates that the remaining service life of such structure might be overestimated if the temperature is averaged over time. Operation cycles, and most notably temperature cycles should be accounted for in the analysis. Cooldown phases should be examined with care, as a residual flux at low temperature might still be able to initiate damage in the microstructure. The fact that the evolution of the damage does not seem to depend on the flux might lead to the hypothesis that creep does not play a significant role in degradation caused by irradiation. However, these simulations correspond to short-term experiments compared to actual structures, with very high deformations and without mechanical restraints, and in which most of the damage occurs in the first months. This may not be applicable for conditions representative of the operation of nuclear power plants and requires further analysis (Giorla and Le Pape 2015).

6. Conclusions

The irradiation of concrete biological shields in LWRs occurs at relatively low temperature ($< 65\text{ }^{\circ}\text{C}$) while the range of irradiation temperature in test reactor varies importantly from “low” temperature, i.e., $< 40\text{ }^{\circ}\text{C}$ to “high” temperature ($> 150\text{ }^{\circ}\text{C}$). In that range of temperatures, the amorphization rate, and thus, the RIVE rate of quartz is significantly affected. Based on data collected on quartz, RIVE rate appears to be significantly accelerated when the irradiation temperature decreases.

The evolution of the mechanical properties (elastic properties and CTE) of irradiated α -quartz are expressed as a function of the current density of the material (that is, the current level of RIVE) instead of the current fluence, using analytic homogenization schemes. These properties have not been characterized in the literature for

RIVE beyond $\approx 30\%$ of the maximum swelling, which limits the predictive capabilities of the proposed model. However, it should be noted that because of the important rigidity contrast between the aggregate and the cement paste, the potential relative loss of elastic modulus in irradiated aggregate may not lead to a significant modification of the damage level in the hcp, e.g., (Giorla *et al.* 2015), as the cracking development is primarily governed by the aggregate expansion amplitudes.

Based on the extensive data collected by Bykov *et al.*, a model accounting for the coupled effects of temperature and irradiation irradiation on quartz RIVE has been developed: RIVE rate is described as a function of temperature. In the absence of neutron flux, thermal annealing can be neglected in the range of temperature observed in test reactors. After implementation in the code AMIE, the simulation of Dubrovskii *et al.*'s experiments on ordinary concrete made of quartz/quartzite aggregate showed consistency with the observed expansion, and pointed out that: (1) Measuring the dimensional change of the diameter and/or height may lead to a significant overestimation of the actual average volumetric expansion in the specimen; (2) Assuming constant neutron flux and temperature can lead to important discrepancies in term of post-irradiation damage and volumetric expansion when compared to more realistic scenarios accounting for periodic reactor shutdowns or other power cycles. Both effects certainly contribute to the experimental scatter observed in data collected in the open literature, and should be considered when analyzing these data, as well as for future simulations of irradiated structures.

While the coupled effects of temperature and neutron irradiation of quartz expansion have been thoroughly studied by Bykov *et al.* (1981), the same effects on other physical and mechanical properties need to be studied to obtain a thorough understanding of the potential effects of differential RIVE in polyphasic, i.e., poly-minerals, aggregates.

Nevertheless, qualitative outcomes in terms of engineering applications can be made: (1) Temperature-induced point-defect annealing in irradiated minerals affects greatly the RIVE kinetics and occurs even at low fluence. This effect questions the validity of using directly radiation-induced aggregate expansion curve obtained in test reactor for LWRs conditions. In that particular perspective, it suggests that some test reactor data, e.g., (Gray 1971), may be overly conservative because obtained at about $45\text{ }^{\circ}\text{C}$, while other data should be reconsidered as relevant for LWRs operation because obtained at quite high temperatures. (2) Because of the effects of temperature on RIVE kinetics, LWR operation cycles can potentially affect radiation-induced damage development in the biological shield.

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Appendices

A. Appendix: Micromechanical Estimates of Thermoelastic Properties of Quartz Polycrystal

Numerical computations at the concrete scale require the thermomechanical properties of aggregates as a function of irradiation. These properties are well documented for the quartz monocrystal Mayer and Lecomte (1960). The aim of this appendix is to upscale these properties from the quartz monocrystal to the aggregate, assumed to be made up of a polycrystalline arrangement of quartz crystals. The properties of interest are the stiffness and the coefficient of thermal expansion.

Introduction to Mean-Field Homogenization of Thermoelastic Characteristics

The general framework of homogenization of elastic then thermoelastic properties is briefly recalled here. For more details, see for example Dormieux *et al.* (2006).

A heterogeneous representative elementary volume (rev) is considered, the behaviour at each point $\underline{z}$ being linear elastic:

$$\sigma(\underline{z}) = \mathbb{C}(\underline{z}) : \varepsilon(\underline{z}) \quad (14)$$

To formulate the effective behaviour, this rev is submitted to kinematic uniform boundary conditions, $\underline{\xi}(\underline{z}) = \underline{E} \cdot \underline{z}$, where $\underline{E}$ is the macroscopic strain. The linearity of the local behaviour (14) allows to introduce the strain localization tensor $\mathbb{A}$ relating the microscopic ε and macroscopic $\underline{E}$ strain tensors:

$$\varepsilon(\underline{z}) = \mathbb{A}(\underline{z}) : \underline{E} \quad (15)$$

The macroscopic stress Σ being the spatial average of the microscopic stress field σ , the effective behaviour of the REV is found to be linear elastic:

$$\Sigma = \mathbb{C}^{eff} : \underline{E} \text{ with } \mathbb{C}^{eff} = \langle \mathbb{C}(\underline{z}) : \mathbb{A}(\underline{z}) \rangle_{\Omega} \quad (16)$$

where $\langle \cdot \rangle_{\Omega}$ denotes the spatial average of a field over the REV Ω . The local behaviour is now considered as linear thermoelastic:

$$\sigma(\underline{z}) = \mathbb{C}(\underline{z}) : (\varepsilon(\underline{z}) - \alpha(\underline{z})\Delta T) \quad (17)$$

where the field of temperature increment ΔT is assumed to be homogeneous across the REV. Alternatively, this behaviour can be written using the eigenstrain ε_L

$$\sigma(\underline{z}) = \mathbb{C}(\underline{z}) : (\varepsilon(\underline{z}) - \varepsilon_L(\underline{z})) \text{ with } \varepsilon_L(\underline{z}) = \alpha(\underline{z})\Delta T \quad (18)$$

or, introducing the eigenstress σ_p :

$$\begin{aligned} \sigma(\underline{z}) &= \mathbb{C}(\underline{z}) : \varepsilon(\underline{z}) + \sigma_p(\underline{z}) \\ \text{with } \sigma_p(\underline{z}) &= -\mathbb{C}(\underline{z}) : \varepsilon_L(\underline{z}) = -\mathbb{C}(\underline{z}) : \alpha(\underline{z})\Delta T \end{aligned} \quad (19)$$

The effective behaviour of the REV is found to be linear thermoelastic (Levin's theorem):

$$\begin{aligned} \Sigma &= \mathbb{C}^{eff} : \underline{E} + \sum_p \sigma_p \\ \text{with } \mathbb{C}^{eff} &= \langle \mathbb{C}(\underline{z}) : \mathbb{A}(\underline{z}) \rangle_{\Omega} \\ \text{and } \sum_p \sigma_p &= \langle \sigma_p(\underline{z}) : \mathbb{A}(\underline{z}) \rangle_{\Omega} \end{aligned} \quad (20)$$

$\mathbb{A}(\underline{z})$ still being the strain localization tensor, defined in (15), where $\sigma_p = 0$. The effective behaviour can also be written in terms of the tensor of thermal expansion:

$$\begin{aligned} \Sigma &= \mathbb{C}^{eff} : (\underline{E} - \alpha^{eff} \Delta T) \\ \text{with } \alpha^{eff} &= \mathbb{C}^{eff-1} : \langle \alpha(\underline{z}) : \mathbb{C}(\underline{z}) : \mathbb{A}(\underline{z}) \rangle_{\Omega} \end{aligned} \quad (21)$$

Upscaling from Crystal to Polycrystal

A specific case of rev is considered here: a polycrystal made up of many crystals, each crystal differing from the others only through its orientation. The following assumptions are considered: (1) Perfect interfaces between crystals (continuity of displacement); (2) At the crystal scale, anisotropy only regards the stiffness and thermal expansion tensors, not the crystal shape; and, (3) Isotropic orientation distribution of crystals, which yields an isotropic effective behavior.

The orientation of one crystal is described by the frame ($\underline{u}_1, \underline{u}_2, \underline{u}_3$) or the angles θ, ϕ, ψ (see Fig. 20). Each crystal stiffness tensor $\mathbb{C}_c(\theta, \phi, \psi)$ thus has the same components in the frame ($\underline{u}_1, \underline{u}_2, \underline{u}_3$) associated to the crystal. This is also the case for the tensor of thermal

expansion $\alpha_{cr}(\theta, \phi, \psi)$.

Voigt and Reuss Bounds and Estimates

The classical Voigt and Reuss bounds on the polycrystalline stiffness $\mathbb{C}_{pc}$ are obtained from an average over the whole rev of the stiffness or compliance tensor:

$$\langle \mathbb{C}_{cr}(\theta, \phi, \psi)^{-1} \rangle_{or}^{-1} = \mathbb{C}^R \leq \mathbb{C}_{pc} \leq \mathbb{C}^V = \langle \mathbb{C}_{cr}(\theta, \phi, \psi) \rangle_{or} \quad (22)$$

where the ' $\leq$ ' operators are considered in the sense of quadratic forms. The averaging operator ' $\langle \rangle_{or}$ ' considers the isotropic orientation distribution, and is computed as:

$$\langle \mathbb{T} \rangle_{or} = \int_{\theta=0}^{2\pi} \int_{\phi=0}^{2\pi} \int_{\psi=0}^{\pi} \mathbb{T}(\theta, \phi, \psi) \frac{\sin \theta}{8\pi^2} d\theta d\phi d\psi \quad (23)$$

with any fourth order tensor $\mathbb{T}$ which depends on orientation θ, ϕ, ψ only through the frame (u_1, u_2, u_3) . The tensors $\mathbb{C}_{pc}$, $\mathbb{C}^R$ and $\mathbb{C}^V$ being isotropic, the inequality (22) translates into inequalities on the effective bulk and shear moduli:

$$\kappa^R \leq \kappa_{pc} \leq \kappa^V \text{ and } g^R \leq g_{pc} \leq g^V \quad (24)$$

The classical formulas relating the bulk and shear moduli to the Young's modulus and Poisson ratio yield Voigt and Reuss estimates of the latter (but not strictly bounds).

To get an estimate of the effective tensor of thermal expansion, an estimate of the strain localization tensor is required, to use (20). The Voigt estimate considers that the strain field is homogeneous in the whole REV, $\varepsilon(\underline{z}) = E$:

$$\mathbb{A}^V(\theta, \phi, \psi) = \mathbb{I} \quad (25)$$

The Reuss estimate considers that the stress field is homogeneous in the whole rev, $\sigma(\underline{z}) = \Sigma$:

$$\mathbb{A}^R(\theta, \phi, \psi) = \mathbb{C}_{cr}(\theta, \phi, \psi)^{-1} : \mathbb{C}^R \quad (26)$$

Due to isotropy of crystal orientation, the tensor of thermal expansion is also isotropic, and its estimates read:

$$\alpha^V 1 = \mathbb{C}^{V-1} : \langle \mathbb{C}_{cr}(\theta, \phi, \psi) : \alpha_{cr}(\theta, \phi, \psi) \rangle_{or} \quad (27)$$

and $\alpha^R 1 = \langle \alpha_{cr}(\theta, \phi, \psi) \rangle_{or}$.

where the averaging operator ' $\langle \rangle_{or}$ ' is also defined by (23) for second order tensors.

Self-Consistent Estimate

Contrary to Voigt and Reuss bounds, so-called homogenization schemes provide estimates of the effective

Fig. 20 Characterization of the orientation of one crystal by angles θ, ϕ, ψ : the frame (u_1, u_2, u_3) associated to the crystal is defined from the reference frame (e_1, e_2, e_3) through the spherical frame (e_r, e_θ, e_ϕ) , appearing as dotted lines.

stiffness. These schemes allow estimate of the average strain localization tensor over each phase of the rev. Among these schemes, the self-consistent one (Kröner 1977) is particularly suited to polycrystals. Indeed, the average strain in each crystal is estimated as the strain arising in a sphere having the same stiffness, embedded into an infinite medium whose stiffness is the sought effective stiffness $\mathbb{C}^{SC}$, and submitted at infinity to a reference strain E^0 . The solution to this so-called auxiliary problem is given by Eshelby (1957):

$$\varepsilon_{cr}(\theta, \phi, \psi) = [\mathbb{I} + \mathbb{P}^{SC} : (\mathbb{C}_{cr}(\theta, \phi, \psi) - \mathbb{C}^{SC})]^{-1} : E^0 \quad (28)$$

where $\mathbb{P}^{SC}$ is the Hill tensor of a sphere in a medium of stiffness $\mathbb{C}^{SC}$. The strain E^0 at infinity can be related to the strain E at the boundary of the rev using the average strain rule $\langle \varepsilon_{cr}(\theta, \phi, \psi) \rangle_{or} = E$. The strain localisation tensor on crystals oriented along θ, ϕ, ψ is thus estimated by:

$$\mathbb{A}_{cr}(\theta, \phi, \psi) = [\mathbb{I} + \mathbb{P}^{SC} : \dots (\mathbb{C}_{cr}(\theta, \phi, \psi) - \mathbb{C}^{SC})]^{-1} : \dots \langle [\mathbb{I} + \mathbb{P}^{SC} : (\mathbb{C}_{cr}(\theta, \phi, \psi) - \mathbb{C}^{SC})]^{-1} \rangle_{or} \quad (29)$$

The self consistent estimate of the effective stiffness is then obtained from (16):

$$\mathbb{C}^{SC} = \langle \mathbb{C}_{cr}(\theta, \phi, \psi) : [\mathbb{I} + \mathbb{P}^{SC} : (\mathbb{C}_{cr}(\theta, \phi, \psi) - \mathbb{C}^{SC})]^{-1} \rangle_{or} : \dots \langle [\mathbb{I} + \mathbb{P}^{SC} : (\mathbb{C}_{cr}(\theta, \phi, \psi) - \mathbb{C}^{SC})]^{-1} \rangle_{or} \quad (30)$$

The latter is a non linear tensorial equation, as the Hill tensor $\mathbb{P}^{SC}$ depends on the sought self-consistent stiffness $\mathbb{C}^{SC}$. As the latter is isotropic, and as the average operator yields isotropic tensors, this tensorial equation can be reduced to two non linear equations over κ^{SC} and g^{SC} , by projection over $\mathbb{J}$ and $\mathbb{K}$. The self-consistent estimate α^{SC} of the coefficient of thermal expansion is obtained from (21) and (29).