

Computing creep-damage interactions in irradiated concrete

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Abstract

Among the various degradation mechanisms possibly affecting the long-term operation of nuclear power plants, the effects of induced expansion and internal degradation that occur in concrete exposed to high-flux neutron radiation require additional research. Notably, the utilization of short-term test-reactor data to assess the long-term structural significance of light water reactor concrete biological shields necessitates to properly capture the concurrent time-dependent effects, e.g., creep and damage caused by radiation-induced volumetric damage. As this poses significant numerical challenges, a creep-damage algorithm was developed to account simultaneously for the progress of damage and visco-elastic processes in the concrete microstructure. The algorithm uses a time-adaptative scheme in which the instants at which damage occurs are explicitly searched for. This provides a non-local continuum damage procedure with very low sensitivity to the time or loading step. The proposed method is then used to simulate creep and restraint effects on radiation- induced degradation in concrete.

Keywords: irradiation, concrete, numerical model, meso-scale, creep, damage, radiation-induced volumetric swelling

1 **Introduction**

² Potential applications for subsequent license renewal of the U.S. commercial

- ³ nuclear power plants has modified the perspective of continuous operation be-
- ⁴ yond 60 years. Aging of non replaceable large components, such as the concrete
- ⁵ containment building or the concrete biological shield (CBS), have become a

⁶ focal point of attention. Among the varied concrete aging mechanisms under
⁷ consideration, lacks of knowledge on irradiated concrete were identified (Graves
⁸ et al., 2014).

Under neutron irradiation, concrete mechanical properties are affected by fluence exceeding $\approx 10^{19} \text{ n.cm}^{-2}$ (Hilsdorf et al., 1978; Field et al., 2015). 10 Radiation-induced volumetric expansion (RIVE) of aggregate appears as a first-11 order mechanism explaining irradiation damage of concrete (Seeberger and Hils-12 dorf, 1982; Field et al., 2015; Le Pape et al., 2015). Neutron high-attenuation, 13 structural restrains and RIVE amplitude (order of magnitude 1%) result in elas-14 tic stresses in the CBS exceeding the strength of irradiated concrete (Le Pape, 15 2016). Two concurrent mechanisms can relax the developed stresses: damage, 16 i.e., cracking, and viscous or quasi-viscous effects, i.e., creep. Irradiation experi-17 ments in test reactor are conducted at a rate of about 1 to 2 orders of magnitude 18 higher than in light water reactors (LWRs), i.e., in actual commercial nuclear 19 reactors (Maruyama et al., 2013; Remec et al., 2013). Because of the lack of 20 experimental data at low irradiation flux, some rate effects and structural con-21 straints are analyzed using numerical simulations at the microstructure level. 22

This raises the question of computing coupled creep and damage processes 23 in a finite element software. The typical procedure consists in using finite differ-24 ences to account for time-dependent processes (*i.e.* creep), and then Newton-25 Raphson iterations in-between the time steps to characterize the damage. This 26 may lead to a strong sensitivity to the time or loading step as well as slow 27 convergence rates. Furthermore, such staggered solving procedure assumes that 28 there is a separation between the time scales of the two processes which is 29 violated here. In the present paper, a numerical algorithm is presented to 30 solve creep-damage problems in finite element codes. The algorithm finds when 31 damage occurs, notably in the case when it occurs in-between the prescribed 32

time steps. Therefore, the time-sensitivity is greatly reduced and the coupling between visco-elastic and damage processes is directly obtained through the algorithm. Also, the algorithm directly accounts in its formulation for timedependent mechanical properties, which makes it particularly useful for the simulation of aging processes.

The algorithm is applied to the study of irradiated concrete, using a model previously validated (Giorla et al., 2015b) against literature data (Elleuch et al., 1972). Different test conditions are simulated in order to study the difference between short-term and long-term testing, as well as a preliminary investigation of the influence of external restraint on the irradiation-induced expansion and damage.

44 2 Damage algorithm

This section describes in details a time-adaptative algorithm to simulate the 45 simultaneous progress of damage (using the framework of continuum damage 46 mechanics) and visco-elastic processes. The present approach uses space-time 47 finite elements to compute the time- and history-dependent response of quasi-18 brittle visco-elastic materials. While the idea of finite elements in space and 49 time has been proposed as early as 1969 (Argyris and Scharpf, 1969; Fried et al., 50 1969), it is not until the 2000 that it received some attention in the context of 51 solid mechanics (Idesman et al., 2001; Bajer and Dyniewicz, 2009; Dumont and 52 Jourdan, 2012; Giorla et al., 2014), notably for their ability to represent spatial 53 domains that undergo some change over time, like mechanical contact (Adélaide 54 et al., 2003) or growth of expansive inclusions (Giorla et al., 2015a). 55

Space-time finite elements have several merits that make them preferable to more traditional approaches (finite elements in space combined with finite differences in time) in the context of viscoelastic quasi-brittle materials (Giorla ⁵⁹ et al., 2014):

The time-stepping procedure is independent of the rheological model used
 for the visco-elastic material (for example generalized Maxwell or Kelvin Voigt chains), while finite difference schemes must be written separately
 for each rheology.

• In space-time finite elements, the displacement field and its derivatives (strain, strain rate, stress, etc) are described as function of time, while with a finite difference scheme, they would be known at discrete instants only. The continuous variations of these fields allows to explicitly find *when* they reach critical values.

• There is a direct and simple relation between the length of the time step and space-time elementary matrices. Updating the time step has therefore a very low computational cost, while for finite difference schemes it necessitates to re-assemble the global system matrix and vector of nodal forces (e.g. Zienkiewicz et al. (1968)). This facilitates the use of timestep-adaptative strategies like the one presented here.

75 2.1 Description of the problem

The solid is considered as a visco-elastic material described by an arbitrary
assembly of springs and dashpots and susceptible to continuum damage. The
constitutive behavior is described with the following components:

• The displacement field **u** and several internal displacement fields \mathbf{a}_j . For a Maxwell or Kelvin-Voigt material, these internal displacement fields are the displacements associated with each dashpot in the model. ϵ and α_j are the strains derived from these displacement fields. In the following,

 \mathbf{x} denotes the concatenation of the displacement field and the internal 83 displacement fields: $\mathbf{x} = [\mathbf{u}, ..., \mathbf{a}_i, ...]$

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• A set of partial differential equations relating the stress σ to ϵ , the α_j , and 85 their respective rates $\dot{\epsilon}$ and $\dot{\alpha}_i$. This set of partial differential equations can 86 be written as a symmetric matrix for any spring-dashpot assembly (Biot, 87 1954). Examples for generalized Kelvin-Voigt, generalized Maxwell, and 88 Burgers materials were given by the authors in (Giorla et al., 2014). 89

• A damage variable d which affects the different material properties of the 90 constitutive equations defined above. In this paper, only the case of an isotropic scalar damage variable varying from 0 to 1 is considered, but 92 the algorithm could be extended to other forms of damage or irreversible 93 phenomena, including orthotropic damage or visco-plasticity. 94

• A failure criterion $\mathcal C$ which dictates at which strain or stress damage increases. In the general case, \mathcal{C} can be written as a function of the extended displacements of the material \mathbf{x} , the damage variable d, and material parameters. Since all these variables vary in time, the criterion is also described as a function of time $\mathcal{C}(t)$. By convention, the failure surface is described by $\mathcal{C} = 0$, and the physically admissible domain by $\mathcal{C} \leq 0$. \mathcal{C} must be a continuous and monotonic function of the displacements. A fully damaged element (d = 1) is considered always below its failure surface. The irreversibility conditions are summarized by:

$$\mathcal{C} \cdot \dot{d} = 0, \ \mathcal{C} \le 0, \ \dot{d} \ge 0 \tag{1}$$

The following condition is also required, which is true for quasi-brittle

materials without hardening:

$$\frac{\partial C}{\partial d} \le 0 \tag{2}$$

• Finally, the solid is subject to external forces \mathbf{f}_e on one section of its boundaries, imposed displacements $\hat{\mathbf{x}}$ on another, and internal body forces \mathbf{f}_b (e.g. thermal expansion). The initial values of the displacement fields $\mathbf{x}(t=0)$ are prescribed.

⁹⁹ All fields are functions of space and time, including material properties like ¹⁰⁰ the stiffness or mechanical strength of the solid. The space-time coordinates ¹⁰¹ (x, y, z, t) are omitted for clarity.

¹⁰² 2.2 Space-time discretization

¹⁰³ The solid is considered over a time interval $[t_S, t_F]$. In the following, $_S$ denotes ¹⁰⁴ the start of the time interval, $_F$ its finish, and Δt its length. The history of the ¹⁰⁵ system up to t_S is already known (either from the initial conditions or from the ¹⁰⁶ result of a previous time step).

The spatial domain is decomposed in a set of polygonal subdomains. The space-time finite element mesh is obtained by extruding each of these subdomains along the time direction between t_S and t_F . Each element has then two corresponding sets of nodes and shape functions, one set located at t_S and the second one at t_F , so that for each node in one set there is another node at the same spatial position in the other set.

The shape functions are taken as linear in time. This corresponds to the quasi-static approximation: acceleration effects are neglected.

Using this discretization, one can write a system of linear equations in the form of:

$$\left[\mathbb{K}(d) + \mathbb{L}(d, \Delta t)\right]\mathbf{x}_F = \mathbf{f}_F(d) - \mathbb{L}(d, \Delta t) \mathbf{x}_S \tag{3}$$

With \mathbb{K} the assembled stiffness matrix, \mathbb{L} the assembled viscosity matrix, \mathbf{x}_S and \mathbf{x}_F the vector of nodal displacements at t_S and t_F respectively, and \mathbf{f}_F the vector of nodal forces at t_F .

The use of prismatic space-time elements ensures the following properties are true if the material properties are constant in time:

- K is independent of the time step.
- L is inversely proportional to the time step.

Proof is given in the appendix of (Giorla et al., 2014) and not repeated here for sake of brevity. The case of time-dependent material properties is addressed in section 2.6.

127 2.3 Damage algorithm

The system (3) is in principle non-linear. However, it is linear if d is set between 128 $[t_S, t_F]$ for all elements, as in this case the material undergoes no increase of 129 damage. A sequence of linear systems could then constructed by incrementing d130 using a step by step procedure similar to that proposed by Rots and Invernizzi 131 (2004) or Zhu and Yvonnet (2015), accounting for the time dimension. Dunant 132 et al. (2011) proposed a renormalisation which made such algorithm work when 133 multiple materials are considered at once. The key difficulty is finding the 134 instants at which damage should be incremented. 135

The algorithm is based on the fact that damage in an element can not increase before that element has reached its failure criterion C. Since the solid is at equilibrium at the beginning of the time step, there is an interval after t_S during which damage does not increase. The goal of the algorithm is to find that interval and the next point of equilibrium. This is similar to the work of Dunant and Bentz (2015) which searches iteratively for these points of equilibrium. A proof of convergence of such algorithms is provided in the latter paper for quasistatic problems.

The algorithm adjusts the time step such that damage events occuring where C is reached happen at the beginning of the step. As opposed to other timeadjusting procedures, the current algorithm moves the *start* of the time step instead of the end to ensure damage occurs only when the elements reach their failure criteria, and not before. This is important as the algorithm ensures that the elements are all in a possible state at all times, making computing their viscous behavior correct.

The algorithm is described using the following steps. Figure 1 shows a flowchart of the entire procedure.

153 1. Initialization

The iterations are initialized with $t_0 = t_S$ and $\mathbf{x}_{S,0} = \mathbf{x}_S$. Damage is assumed constant during the time step and equal to its value at the beginning of the time step: $d_0 = d_S$.

157 2. Resolution

 $t_i, \mathbf{x}_{S,i}$ and d_i are known and such that all elements at t_i are strictly below their failure surface ($C(t_i) < 0$ in all elements), and a system similar to (3) has been built. Resolution of that system gives the displacements at the end of the time step $\mathbf{x}_{F,i}$.

¹⁶² 3. Failure criterion

From $\mathbf{x}_{S,i}$, $\mathbf{x}_{F,i}$, d_i and the space-time shape functions, the failure criteria for all elements can be calculated as a function of space and time. Notably, for all elements the first instant at which their failure criterion is reached can be obtained. This can be directly evaluated for simple criteria (constant strength
or limit strain), but may necessitate a bisection over time in the general case.

¹⁶⁸ 4. Damage increment

 t_{i+1} is the first instant at which an element \mathcal{E} reaches its failure criterion. Condition (2) ensures that $t_{i+1} > t_i$ by continuity. The damage in \mathcal{E} is then increased at t_{i+1} by an fixed amount δd . This action must be repeated until \mathcal{E} is strictly below than its failure surface at t_{i+1} . This defines d_{i+1} .

173 5. Time step adjustment

The irreversibility conditions (1) are satisfied over the interval $[t_i, t_{i+1}]$, so 174 the solution calculated at step 2. gives the actual solution of the problem on 175 that interval and does not need to be calculated again. Therefore, the next time 176 step is $[t_{i+1}, t_F]$. This can be achieved by moving all nodes located at t_i to t_{i+1} . 177 $\mathbf{x}_{S,i+1}$ is obtained as the values of the extended displacements at t_{i+1} from 178 $\mathbf{x}_{S,i}, \mathbf{x}_{F,i}$, and the space-time finite element shape functions. K and is not 179 affected, and \mathbb{L} must be scaled according to that new time interval. Finally, 180 the elementary contributions of the damaged element \mathcal{E} to \mathbb{K} , \mathbb{L} and \mathbf{f} must be 181 re-evaluated. This yields a new assembled system with the same structure as 182 (3).183

184 6. Iteration

Go back to 2. using the new t_{i+1} , $\mathbf{x}_{S,i+1}$ and d_{i+1} .

186 7. Exit

The algorithm has converged when no element reaches its failure surface before t_F (step 3.). This always occurs after a finite number of iterations due to the fixed damage increment δd .

¹⁹⁰ 2.4 Non-locality

The damage pattern obtained from a given stress-strain relationship depends 191 strongly on the mesh if the relationship has been defined from the local stress 192 and strain of the elements. This in turns means that given a domain and 193 boundary conditions, the energy dissipated by the initiation and propagation of 194 damage is not constant with the mesh. Problematically, this energy converges 195 to 0 as the mesh is refined as the elements damaged coalesce into a single band 196 of vanishing width. To palliate this defect, Pijaudier-Cabot and Bazant (1987) 197 proposed to transform the formulation of the stress-strain relationship from a 198 local to a non-local one. 199

To make the proposed algorithm non-local, the strains are smoothed using 200 a kernel of radius r_{nl} when calculating the failure criterion C. The radius is 201 characteristic of the size of the fracture process zone, a consequence of the un-202 derlying microstructure of the material. For example, the characteristic distance 203 for concrete models is frequently taken as 1 to 2 times the largest aggregate di-204 ameter. This corresponds to the width of observed networks of microcracks 205 forming around a main crack when the concrete is failing in tension (Otsuka 206 and Date, 2000). 207

The variable to be smoothed can in principle be chosen freely. However, in practice, it is preferable to smooth strains, as these are defined independently of the material properties and can therefore be meaningfully scaled and added (Jirásek, 1998). The internal strain fields associated with the dashpots are also smoothed using the same kernel as they represent a certain fraction of the strain history of the material. The stresses are then evaluated using the non-local strains and the local mechanical properties.

The equilibrium problem solved is local, but the failure criterion is non-local. This is physically explained by t Smoothing the failure criterion ensures that damage forms over the prescribed radius. Figure 2 shows the damage pattern obtained for a plate subject to uni-axial tension for different mesh sizes. The material is a generalized Kelvin-Voigt chain with one module, with a linearsoftening strain-stress relation. The width of the damage band is independent of the characteristic size of the elements in the mesh.

This procedure is numerically more efficient than solving the complete nonlocal problem by introducing the smoothing couplings in the system of equations. Indeed, we find that only the criterion needs to be non-local using the solving strategy described in this paper. Nonetheless, the problem is discretized not only in space but also in time, and the sensitivity to this latter discretisation should also be assessed.

228 2.5 Time-step sensitivity

The algorithm is adaptative in time and is not affected by the prescribed the 229 time (or load) step. This property is shown on a simple case with a single 230 square element on which a tensile displacement is applied at a constant rate. 231 The material is the same as the one used in the previous test case, with a 232 stress-strain curve presenting a linear softening branch. The time step is varied 233 between 0.001 and 0.5 in arbitrary units, with the larger time step chosen so 234 that the first step occurs beyond the peak. The resulting stress-strain curves 235 with the different time steps are shown in Figure 3. For all time steps, the 236 algorithm gives as a result the exact behavior of the element for the considered 237 strain. Furthermore, the damage algorithm (not shown in the graph) always 238 finds the exact peak of the material behavior and initiates damage at this time. 239 This illustrates the robustness of the algorithm to large time (or load) steps. 240

Of course, when the material has a time-dependent behavior, the time steps must be chosen to be able to capture it.

243 2.6 Time-dependent material properties

The algorithm remains valid with little modifications when the material properties are function of time, due to (for example) ageing, irradiation or temperature
effects.

 K only depends on the elastic properties at t_F as it is a representation of the instantaneous response of the material.

• L depends on the rate of the viscous properties. However, it can be divided into two parts: the first is inversely proportional to the time step, and independent of the rate of the viscous properties, while the second is independent of the time step and depends only on the rate of the viscous properties. The two parts can be stored independently in the memory in the memory so that the adjustment of L with the time step remains simple.

• If the strength (or limit strains) of the material are time-dependent, the algorithm can also be used as written. In this case, the failure criterion becomes a more elaborate function of time but retains its continuity properties. Therefore, the instant at which an element reaches its failure criterion can still be obtained with an appropriate bisection, by evaluating *C* at different instants.

²⁶² 2.7 Matrix conditioning

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Since \mathbb{L} is inversely proportional to the time step and \mathbb{K} is constant, the conditioning number of the final assembled matrix $\mathbb{K} + \mathbb{L}$ increases when the time step decreases. Therefore, resolution of (3) with a conjugate gradient solver becomes increasingly difficult after each iteration of the damage algorithm (Gilbert and Nocedal, 1992). However, when the time step $(f_F - t_i)$ becomes much smaller than the characteristic time of the viscoelastic processes, the problem may be considered as "instantaneous" and rate effects neglected.

When this occurs, the viscoelastic variables \mathbf{a}_j can be fixed to their last computed values, leaving only the displacements \mathbf{u} as unknown in (3). The contribution of \mathbb{L} to the global system then vanishes, unless the material behavior depends explicitly on the strain rate $\dot{\epsilon}$ (this would be the case, for example, of a single Kelvin-Voigt unit). This approximation greatly improves the conditioning number of the global system matrix for very small time steps, and therefore improves the resolution of (3).

²⁷⁷ In the present work, this threshold is taken equal to 1 second.

²⁷⁸ 3 Application to irradiated concrete

The algorithm is applied to meso-scale simulations of concrete specimens subject to irradiation-induced expansion and degradation. Concrete is represented at its meso-level with aggregates embedded in a cement paste matrix. The simulations are carried out in two dimensions using plane strain assumption to limit the computational time. The model, previously validated in (Giorla et al., 2015b) on irradiation experiments from the literature (Elleuch et al., 1972), is briefly summarized here.

286 3.1 Microstructure

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The aggregates are generated as random polygons following the work of Beddow and Meloy (1980). The polar radius r of each vertex is calculated as an harmonic function of the polar angle θ and the average radius of the particle r_0 :

$$r = r_0 \left[1 + \sum_{j=1}^{m} e^{-p\log(j) - b} \cos(j\theta + \alpha_j) \right]$$
(4)

With p and b parameters describing the shape of the particles, and α_j random numbers uniformly taken between 0 and 2π . p and b are taken equal to 0.9 and 1.9 respectively from the work of Wang et al. (1999), while the radii are generated using a Fuller and Thompson (1907) curve between 8 and 0.5 mm. The number of vertexes in each aggregate is randomly taken between 6 and 10. Only the largest aggregates are represented; the cement paste mechanical properties are increased to account for the missing aggregate fraction.

The aggregates are randomly placed (from the largest to the smallest) in a 10 \times 10 cm sample with a minimum interdistance of 0.5 mm. The sample is then meshed with a conforming Delaunay triangulation so that the node density is relatively constant throughout the sample. The relative size of the sample and the aggregates were chosen so that the microstructure was representative of the material, and avoid the variability induced by the presence of unusually large aggregates as observed in a previous works (Giorla et al., 2015b).

The microstructure and a detail of the finite element mesh is shown in Figure 4. The mesh is composed of 19,174 linear elements, for a computational time ranging between 24 and 48 hours.

³⁰⁹ 3.2 Constitutive behaviors

In the following, $_p$ denotes properties of the cement paste, and $_a$ the aggregates.

311 Cement paste

³¹² Cement paste is simulated as a viscoelastic quai-brittle damage material ³¹³ with thermal deformation. Creep is simulated with an aging logarithmic model ³¹⁴ assuming a separation of the short-term recoverable creep ϵ_r and the longterm non-recoverable creep ϵ_c . The rheological model is thus composed by an elastic spring in series with a Kelvin-Voigt unit (short-term creep) and a time-dependent dashpot (long-term creep) as with the B3 model (Bažant and Baweja, 2000) or the model more recently proposed by Hilaire et al. (2014). This assembly is shown in Figure 5.

³²⁰ The corresponding set of constitutive differential equations is:

$$\sigma = (1-d) \mathbb{C}_e : [\epsilon - \epsilon_r - \epsilon_c - \epsilon_{imp}]$$
(5)

$$\sigma = (1-d) \left[\mathbb{C}_r : \epsilon_r + \mathbb{E}_r : \dot{\epsilon}_r \right]$$
(6)

$$\sigma = (1-d) \left[1 + \frac{t}{\tau_c} \right] \mathbb{E}_c : \dot{\epsilon}_c$$
(7)

With \mathbb{C}_e the elastic stiffness tensor of the material, \mathbb{C}_r and \mathbb{E}_r the stiffness and viscosity tensors of the Kelvin-Voigt module, \mathbb{E}_c the initial viscosity tensor of the time-dependent dashpot, ϵ_{imp} the imposed deformation (shrinkage, thermal strains, etc), and τ_c the characteristic time of the logarithmic creep. This system can be made symmetric by substracting the elastic equation (5) to each viscous relation (6-7).

To limit the number of calibration parameters, the different creep properties are assumed equal: $\tau_c \mathbb{C}_r = \mathbb{E}_r = \mathbb{E}_c$. Also, the Poisson ratio of all springs and dashpots are constants and equal, following experimental observations on biaxial long-term creep tests by Charpin et al. (2015).

The cement paste is assumed to fail only in tension. The failure criterion C_p is written so that the material exhibits a linear softening branch after the peak:

$$C_p = \|\bar{\epsilon}\| - \frac{\mathbf{E}_{soft}}{\mathbf{E}_{soft} + \mathbf{E}_{inst}} \epsilon_{y,t}$$
(8)

333 With $\|\bar{\epsilon}\|$ the maximum principal component of the averaged (non-local)

strains, E_{soft} the slope of the softening branch (a material parameter), E_{inst} the current instantaneous modulus, accounting for the damage and the evolution of the creep processes, and $\epsilon_{y,t}$ the strain at the end of the softening branch. In practice, E_{soft} is obtained from the tensile strain at the peak ϵ_t , the tensile strain at the end of the softening branch $\epsilon_{y,t}$, and the Young modulus of the material E.

340 Aggregates

The aggregates mechanical behavior is taken as a purely elastic and made of homogeneous phases undergoing the same RIVE. The strain ϵ_{Φ} imposed by the irradiation-induced swelling is given as a function of the neutron fluence Φ by Zubov and Ivanov (1966) equation:

$$\epsilon_{\Phi} = \frac{\kappa \ \epsilon_{max} \left(e^{\delta \Phi} - 1 \right)}{\epsilon_{max} + \kappa \ e^{\delta \Phi}} \tag{9}$$

With ϵ_{max} the final deformation, and κ and δ two parameters controlling the shape of the sigmoid curve.

Cracking of aggregates is neglected as it is expected that the much less resistant cement paste will fail first. This hypothesis will be verified a posteriori by measuring the stresses in the aggregates.

350 3.3 Material properties

The mechanical properties, given in Table 1, are chosen to represent a prototypical concrete used for the construction of nuclear power plants in the 70s. In order to provide a basis of comparison, the mechanical properties of the cement paste and the aggregates are calibrated so that the elastic and strength macroscopic properties of the sample are similar to the one used by Le Pape (2016) for a structural analysis of a biological shield (Young's modulus E = 34 GPa, ³⁵⁷ compressive strength $f_c = 40$ MPa).

The creep properties are provided by the analysis of uniaxial creep experiments on pure cement paste samples by Le Roy (1995) (water/cement ratio 0.5). The Poisson ratio is assumed to be constant in time. The creep properties are adjusted with the temperature using an Arrhenius-type law. The activation energy E_{act} is taken equal to 5000 K according the recommendations of Bažant and Baweja (2000).

Thermal deformations are accounted for using the thermal expansion coefficients θ_p and θ_a from the radiation experiments of Elleuch et al. (1972). Shrinkage is neglected as the effect of irradiation is not well understood yet.

Table 1: Material properties for the cement paste and the aggregates. (1) backcalculated using the target Young's modulus and compressive strength; (2) typical value for concrete; (3) calibrated on the creep experiments of cement pastes with a water-cement ratio of 0.5 of Le Roy (1995); (4) arbitrary parameter; (5) from the experimental data of Elleuch et al. (1972).

Cement paste				
Elastic properties	E_{p}	20	[GPa]	(1)
	ν_p	0.2	[-]	(2)
Creep properties	η_c	40	[GPa.d]	(3)
	$ au_c$	2	[d]	(3)
	E_{act}	5000	[K]	(2)
Failure properties	ϵ_t	0.4	[mm/m]	(1)
	$\epsilon_{y,t}$	0.5	[mm/m]	(1)
	r_{nl}	0.5	[mm]	(1)
	δd	0.1	[-]	(4)
Thermal properties	$ heta_p$	9	$[10^{-6} \ 1/\mathrm{K}]$	(5)
Aggregates				
Elastic properties	\mathbf{E}_a	60	[GPa]	(2)
	$ u_a$	0.2	[-]	(2)
Thermal properties	θ_a	7.5	$[10^{-6} \ 1/\mathrm{K}]$	(5)

367 3.4 Test conditions

The model is tested in 8 different conditions in order to assess the relevance of creep in the analysis of irradiation-induced damage. The conditions are the combination of three different factors:

- With/Without creep. For the simulation without creep, the differential equations (6-7) are ignored and ϵ_c and ϵ_r are both set to 0. Otherwise, the same numerical procedure is adopted.
- Slow/Fast irradiation. The neutron fluence is taken as a linear function of time up to 4 × 10¹⁹ n/cm². The fluxes are chosen to represent fast-flux experiments in test reactors (duration of irradiation 47 days) of pressurized water reactors operation (duration 80 years) respectively.
- Free/Restrained. In the later case, the vertical displacements of the bot tom and top edges of the sample are blocked, but the sample is still free
 to deform in the horizontal direction. In both cases the middle point of
 the bottom edge is fixed to avoid global displacement of the sample.

The temperature is supposed homogeneous throughout the sample and equal to 65 °C which is the design operating conditions in commercial test reactors (Field et al., 2015). Effect of irradiation on relative humidity is neglected due to lack of supporting data.

386 4 Results and Discussion

The expansions and average damage in the cement paste were calculated at each step of the simulations. The values of the expansions correspond to maximum displacement variation between two opposite edges of the sample, as if they were measured with an external extensometer or a micrometer. Damage is obtained as the average of the damage scalar d over the elements representing the cement paste. Note that in Fig. 6-9, the marks (circles) correspond to the actual simulation results, while the curves are obtained using the **loess** smoothing function from the R statistical software package (R Core Team, 2012).

Figure 6 shows the respective evolutions of concrete volume increase as a function of the neutron fluence for all studied cases.

When unrestrained, the expansion is isotropic and its evolution with fluence is identical for the four studied cases (underlying grey curve) This result was expected as the RIVE imposes an elastic deformation to the aggregates which does not depend on the neutron flux.

When restrained, the expansion can only develop laterally. The vertically-401 restrained RIVE results in the creation of significant stresses, which, in turn, 402 cause the development of creep. Creep presents a significant effect on the evo-403 lution of the volumetric expansion and internal damage. An important and 404 sudden deformation increase is observed corresponding to the formation and 405 opening of a compressive fracture through the sample. When creep is consid-406 ered, the development of this fracture occurs at a higher fluence and in a more 407 progressive manner. Once this fracture opened, the remaining of the microstruc-408 ture produces a micro-cracking pattern similar to that developed in unrestrained 409 conditions. During this later phase, the volume increases at a similar rate for 410 all cases, although the rates and amplitudes appear smaller when expansion is 411 restrained. Creep increases the expansion during this later stage by an amount 412 that seems to be higher for low-flux irradiation. 413

The damage as a function of the neutron fluence is plotted, respectively, in Figure 7 for the unrestrained condition and Figure 8 for the restrained case. The damage evolutions in the simulations neglecting creep are similar regardless of the time scale of the irradiation, i.e., slow or fast flux, which demonstrates the

stability of the damage algorithm. At a given fluence level, creep contributes to 418 the reduction of average damage. It appears that creep causes a latency effect on 419 the initiation of damage. This latency effect, i.e., apparent shift in fluence, de-420 creases with the neutron flux because lower flux permits the relaxation through 421 a viscoelastic process. This observation illustrates the competition between the 422 two mechanical energy dissipation playing a role in the stress relaxation: the 423 formation of cracks and the viscous relaxation. Notably, in the restrained case 424 accounting for creep effects, the initiation and propagation of damage is much 425 smoother, and vertical cracks can be observed in the sample. 426

Finally, Figure 9 shows the volume increase as a function of the damage for 427 the restrained case. The results of the simulations in the unrestrained case are 428 plotted in grey solid lines. When creep is considered, the material exhibits a 429 much larger expansion for the same degree of damage for a long-term irradiation 430 exposure. This also indicates that for the long-term scenario, damage in the 431 material would be much lower for the same level of expansion when calculated 432 with creep than what would be obtained with a purely elastic simulation. This 433 indicates a strong need for accounting for visco-elastic effects in the analysis of 434 the long-term durability of CBS. 435

436 4.1 Creep of Irradiated Cement Paste and Concrete

The current model assumes that the creep properties of the material remains unaffected by irradiation. This is a rather simplistic approximation which is dictated by the poor understanding of the effects of irradiation on shrinkage and creep: (1) The mechanistic understanding of unirradiated creep is still controversial despite decades of research, and, (2) Very limited data on shrinkage and creep of irradiated cementitious materials are available in the literature (Gray, 1971; McDowall, 1971).

Under gamma irradiation for 10 months at a dose of 0.114 kGy h^{-1} , Mc-444 Dowall (1971) found that the creep rate of concrete (10 MPa) decreases while 445 the shrinkage rate is increased. The specimens were sealed in copper foils, al-446 though venting radiolytic gas is permitted through a gas bubbler filled with 447 water. While moisture transport through vapor diffusion is not allowed, gas 448 transport could lead to a partial, though limited, 'drying' of the specimens. 449 Hence, it can be assumed, by lack of better data, that the effects on creep and 450 shrinkage rates are primarily attributed to gamma-ray exposure. 451

Gamma irradiation induced hydrogen production of absorbed water and 452 nano-confined water are respectively two and one order(s) of magnitude higher 453 than that of bulk water in controlled-nanopore (8-300 nm) boro-silicate glasses 454 (Rotureau et al., 2005; Le Caër et al., 2005). The microstructure of calcium-455 silicate hydrates (C-S-H) suggests that similar radiolytic effects could be ob-456 served in cement pastes potentially causing change primarily at the nanoscale. 457 A possible irradiation-induced drying mechanism of absorbed and nano-confined 458 water could result in the collapse of C-S-H as it is observed after drying under 459 sustained moderate temperature (Jennings et al., 2007; Maruyama et al., 2014). 460 Hence, such a mechanism could possibly cause the increase of the apparent 461 viscosity, and thus, the decrease of the creep rate under gamma irradiation. 462 Concurrently, radiolysis of absorbed water at the C-S-H surface can result in 463 releasing the disjoining pressure (Beltzung and Wittmann, 2005), and hence, 464 increasing the shrinkage rate. 465

⁴⁶⁶ Under concurrent neutron and gamma irradiation, shrinkage and, subse-⁴⁶⁷ quently, creep (6.9 MPa) of Portland cement grout specimens were tested by ⁴⁶⁸ Gray (1971) at varied temperatures ranging from 20 °C to nearly 95 °C. For ⁴⁶⁹ about 15 d, the creep specimens were subjected to irradiation in Herald test ⁴⁷⁰ reactor (UK). Fast neutron fluence is not specifically reported but can be es-

timated at $\approx 0.75 \times 10^{19} \text{ n.cm}^{-2}$ with a flux of $\approx 5 \times 10^{12} \text{ n.cm}^{-2} \text{ s}^{-1}$. The 471 deformation history reconstructed from Gray's data is presented in Figure 10. 472 Creep kinetics at 60 to 95 °C (average \approx 70 °C) under concurrent irradiation is 473 one order of magnitude higher than creep of specimens out-of-pile before or after 474 irradiation in a similar range of temperature. The relative humidity and loss of 475 mass of the specimens were not monitored. Neutron-induced radiolytic effects 476 on water are similar in nature to gamma ray effects (Kontani et al., 2010), and 477 thus, should lead to a decrease of the creep rate following the observation of Mc-478 Dowall. This indicates the possibility of an additional mechanism, even though 479 experimental data support that neutron irradiation has a limited effect on the 480 macroscopic mechanical properties on cement paste (Gray, 1971; Elleuch et al., 481 1972). The possible mechanisms of neutron-induced damage on the solid phase 482 of cement hydrates remain poorly understood and requires further investigation. 483

484 4.2 On the Role of Temperature

The effects of temperature were not specifically studied in this article but need 485 to be discussed as an additional source of discrepancy between test reactor 486 data obtained at 40 to 250 °C and the actual behavior of concrete in LWRs at 487 < 65 °C. In addition to the known effects of temperature (Naus, 2010), specific 488 interactions between temperature and irradiation exist: (1) Dehydration caused 489 by combined drying and radiolysis results in important shrinkage strains in the 490 cement paste (Elleuch et al., 1972). (2) Thermal expansion aggravate the RIVE 491 of aggregate leading to the development of cracks in the paste (Le Pape et al., 492 2015). (3) Temperature increase allows the annealing of neutron-irradiation-493 induced point defects. Hence, RIVE rate is higher at lower temperature (Bylov 494 et al., 1981). These effects are not accounted for in the present model and 495 require further research. 496

497 5 Conclusion

A numerical algorithm was presented in this paper to compute creep and contin-498 uum damage with space-time finite elements. The algorithm adapts the length 499 of the time step to the rate of the damage process, thus allowing an accurate 500 characterization of the viscoelastic dissipation in the material. This decreases 501 greatly the sensitivity of the procedure to the time or loading step, even when 502 the material is directly loaded after its peak in the stress-strain curve. The 503 method is non-local by considering average strains during the evaluation of the 504 instant at which failure occurs. 505

The algorithm was applied to the study of radiation-induced expansion and 506 degradation. Concrete was simulated at the meso-scale with aggregates repre-507 sented by polygonal inclusions embedded in a cementitious matrix. The expan-508 sion was driven by the radiation-induced swelling of the aggregates, while the 509 damage propagated in the viscoelastic cement paste. Different scenarios were 510 evaluated to investigate the rate and restraint effects on the overall expansion 511 and damage. Imposing a restraint on the expansion causes an increase of the de-512 formation in the lateral direction, and fracture formation was observed through 513 the sample during the early stages of the irradiation. However, accounting for 514 creep in the simulation reduces the damage onset and propagation for the same 515 level of expansion. The final volumetric expansion rate as a function of the 516 fluence seemed independent of the conditions and rate of irradiation. These 517 results are of practical interest for the interpretation of irradiation experiments 518 conducted in test reactor, i.e., fast flux and unrestrained expansion, in the per-519 spective of analyzing the effects of neutron irradiation on the concrete biological 520 shield in actual light water reactors (LWRs) conditions. In other words, the ob-521 tained results suggest that creep can play a favorable role in terms of LWRs 522 long-term operation by delaying the initiation of damage to a higher fluence 523

524 exposure.

Still, several questions remain open on the effects of radiation on creep, 525 including the role of gamma radiation, as well as the strong coupling with tem-526 perature. Confirmatory experimental results complementing those obtained by 527 McDowall (1971) and Gray (1971) are required to better understand interac-528 tions between creep and radiation-induced swelling and damage in concrete. 529 The proposed algorithm can serve as a numeric tool to carry out this analysis 530 at the concrete mesoscale and formulate a more complete material model. A 531 set of probabilistic simulations could then be performed with varying condi-532 tions in order to express a numerical macroscopic model that could be used in 533 a structural analysis. 534

The algorithm itself has a generic and highly flexible formulation, which 535 makes it suitable for the analysis of several creep-degradation phenomena in 536 concrete. It was used in previous publications in the context of alkali-silica 537 reaction (Giorla et al., 2015a), radiation-induced volumetric expansion (Giorla 538 et al., 2015b), or snap-back analysis of visco-elastic materials (Dunant and Hi-539 laire, 2015). It could be applied to analyze early-age cracking, loading rate 540 effects on concrete failure, or other long-term durability phenomena such as 541 delayed ettringite formation. 542

543 6 Acknowledgements

This research is sponsored by the U.S. Department of Energy (DOE) Light Water Reactor Sustainability Program. This manuscript has been authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to ⁵⁵⁰ publish or reproduce the published form of this manuscript, or allow others ⁵⁵¹ to do so, for United States Government purposes. The Department of Energy ⁵⁵² will provide public access to these results of federally sponsored research in ac-⁵⁵³ cordance with the DOE Public Access Plan (http://energy.gov/downloads/ ⁵⁵⁴ doe-public-access-plan).

555 7 Notations

- ⁵⁵⁶ The following symbols are used in this paper:
- $\mathbf{a}_j = \text{internal viscous displacement of the dashpots of the material}$
- b = parameter controlling the shape of the aggregate particles (4)
- $_{559}$ \mathbb{C} = fourth-order elastic stiffness tensor
- 560 C = failure criterion (8)
- $_{561}$ d = scalar damage variable
- 562 $\mathbb{E} =$ fourth-order viscosity tensor
- E = elastic Young's modulus
- $E_{act} = activation$ temperature of the Arrhenius law for creep
- $_{565}$ **f** = forces applied on the system
- 566 $\mathbb{K} = \text{global assembled stiffness matrix (3)}$
- $_{567}$ \mathbb{L} = global assembled viscosity matrix (3)
- p_{568} p = parameter controlling the shape of the aggregate particles (4)
- r = radius of the aggregate particles (4)
- $r_{nl} = radius of the non-local averaging$
- $t_F = time$ at the end of the time step
- $t_S = time$ at the start of the time step
- $_{573}$ **u** = displacement field
- $\mathbf{x} = \text{global unknown of the problem (3)}$
- $\alpha_j = \text{second-order internal viscous strain tensors, deriving from } \mathbf{a}_j$
- 576 $\Delta t = \text{time step}$
- $\delta = \text{scale parameter for the Zubov and Ivanov curve (9)}$
- $\epsilon = \text{second-order strain tensor, deriving from } \mathbf{u}$

- 579 ϵ_{max} = amplitude of the RIVE (9)
- $\epsilon_t = \text{limit strain at the peak of the stress-strain curve (8)}$
- $\epsilon_{y,t} = \text{limit strain at the end of the softening curve (8)}$
- 582 $\eta = \text{uni-axial viscosity}$
- $\theta =$ thermal expansion coefficient
- $\kappa = \text{shape parameter for the Zubov and Ivanov curve (9)}$
- 585 $\nu = \text{Poisson ratio}$
- $\sigma = \text{second-order stress tensor}$
- 587 $au_c = ext{characteristic time of the logarithmic creep}$
- $\phi = \text{neutron fluence } (9)$

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Figure 1: Flowchart of the algorithm with two space-time finite elements (with 1 space dimension). Red indicates values that have been calculated at the corresponding step of the algorithm. Arrows show at which instant corresponds each displacements \mathbf{x} .



Figure 2: Damage pattern after failure obtained for a uniform plate subject to uni-axial tension with three different mesh sizes. Grey level indicates the value of the scalar damage variable (white = 1). The strength of one element in the center of the plate was reduced by 0.1 % to enforce the location of the damage band.



Figure 3: Stress-strain curves for a single element loaded in displacement (constant strain rate) with different time steps. The post-peak behavior is recovered by the algorithm even when the element is loaded beyond its peak.



Figure 4: Microstructure and zoom-in on the details of the finite element mesh.



Figure 5: Spring-dashpot model for the cement paste.



Figure 6: Volumetric expansion as a function of the neutron fluence for the restrained case.



Neutron Fluence [10¹⁹ n/cm²]

Figure 7: Damage in the cement paste as a function of the fluence for the free boundary condition. Trends for the restrained case are indicated in grey. The microstructures correspond to the free, long-term, elastic simulation (left) and free, long-term, visco-elastic simulation (right) for a neutron fluence of 0.56.



Figure 8: Damage in the cement paste as a function of the fluence for the restrained boundary condition. Trends for the free case are indicated in grey. The microstructures correspond to the restrained, long-term, elastic simulation (left) and restrained, long-term, visco-elastic simulation (right) for the fluence at which the macro-crack percolates.



Figure 9: Volumetric expansion as a function of the damage for the restrained case. The microstructures correspond to the restrained, long-term, visco-elastic simulation at three stages of the damage process: initiation and propagation of a compressive macro-crack ($\phi = 0.39$), initiation of diffuse micro-cracks ($\phi = 0.98$) and further opening of the micro-cracks ($\phi = 4$).



Figure 10: Measured shrinkage (\circ) and creep (\Box) strain of Portland cement grout. (\Box) correspond to creep under irradiation. Reconstructed from Gray's data. Numbers in brackets indicate the creep kinetics parameter, a, given in $\mu m m^{-1} d^{-1}$, assuming a logarithmic fit $\varepsilon = \varepsilon_0 + a \log(1 + t - t_0)$. Solids line: creep kinetics.