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Radiation damage evaluation on concrete within a facility for Selective Production of Exotic Species (SPES Project), Italy

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ABSTRACT

Concrete is commonly used as a biological shield against nuclear radiation. As long as, in the design of nuclear facilities, its load carrying capacity is required together with its shielding properties, changes in the mechanical properties due to nuclear radiation are of particular significance and may have to be taken into account in such circumstances. The study presented here allows for reaching first evidences on the behavior of concrete when exposed to nuclear radiation in order to evaluate the consequent effect on the mechanical field, by means of a proper definition of the radiation damage, strictly connected with the strength properties of the building material. Experimental evidences on the decay of the mechanical modulus of concrete have allowed for implementing the required damage law within a 3D F.E. research code which accounts for the coupling among moisture, heat transfer and the mechanical field in concrete treated as a fully coupled porous medium. The development of the damage front in a concrete shielding wall is analyzed under neutron radiation and results within the wall thickness are reported for long-term radiation spans and several concrete mixtures in order to discuss the resulting shielding properties.

1. Introduction

Irradiation in the form of either fast and thermal neutrons, primary gamma rays or gamma rays produced as a result of neutron capture can affect concrete. This is not the only mechanism known for degradation of concrete in a nuclear power plant, since also leaching is acknowledged as a source of damage on concrete, see e.g. [1,2]; particularly, a diffusion controlled transport mechanism can be responsible for the migration of radionuclides through the porous material. However leaching is conceived to involve mainly concrete repositories for low-up to high-activity wastes where the activated sources are essentially in liquid state, while in this work concrete is considered as a barrier for a fission facility, i.e. concrete shielding elements are subjected to irradiation alone.

If the effects of nuclear radiation on concrete are considered, changes in the properties of exposed concrete appear to depend primarily on the behavior of concrete aggregates that can undergo a volume change when exposed to radiation. Radiation damage in concrete aggregates is caused by changes in the lattice structure of the minerals in the aggregates. Fast neutrons are mainly responsible for the considerable growth, caused by atomic displacements, that has been measured in certain aggregates (e.g. flint). Quartz aggregates, made of crystals with covalent bonding, seem to be more affected by radiation than calcareous aggregates that contain a weaker ionic bonding. Neutron fluences of the order of 10^{19} n/cm^2 and gamma radiation doses of 10^{10} rad seem to become critical for concrete strength. It is here recalled that "particle fluence" or simply "fluence" refers to the number of particles (or photons, in case of gamma-rays), which enter a specified volume. This quantity is expressed as the number of particles per square length (e.g. [neutrons/cm^2]). The rate of change of fluence with time is referred to as the "particle fluence rate" or particle flux density" or simply "flux". This quantity is expressed as the number of particles per square length, per unit time (e.g. [neutrons/(cm^2 s)]).

Particularly, the study has been motivated by an ongoing research program in conjunction with the National Laboratories of Legnaro (Padua, Italy) within the SPES Project [3–7] and it represents a first step toward the assessment of radiation damage induced by nuclear radiation in concrete vessels; the facility to be built will be directed to the production of special radioactive heavy ion beams from a primary proton beam impinging on a target made of fissionable material, where fission reactions are expected to take place, thus representing ideally a point-source of neutrons in the specific problem. The phenomenon has been treated with a macroscopic approach under the assumptions made in Section 2 for the radiation field.

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An already available F.E. research code able to perform fully coupled hygro-thermo-mechanical analyses for cementitious materials has been upgraded to take into account the effects of nuclear radiation on the material via the introduction of a new damage variable; specifically, in Section 3 radiation damage is defined starting from experimental evidences and it is supposed to act in conjunction with the already implemented chemo-thermo-mechanical damage; numerical results which lead to quantify the decay in stiffness of a concrete wall under neutron radiation exposure are reported and discussed.

2. The mathematical model

By means of the 3D F.E. research code NEWCON3D [8–18] concrete is treated as a multiphase system where the micropores of the skeleton are partially filled with liquid water, both in the form of bound or absorbed water and free or capillary water, and partially filled with a gas mixture composed of dry air (non-condensable constituent) and water vapour (condensable), supposed to behave like an ideal gas [8–10].

When higher than standard temperatures are taken into account, several phenomena are considered: heat conduction, vapour diffusion and liquid water flow in the voids.

As regards the mechanical field, the model couples shrinkage, creep, damage and plasticity effects.

2.1. The radiation field

Neutron radiation is considered in the following, in line with the specificity of the problem treated within the SPES Project, which will be illustrated below.

Particularly, the effects of nuclear radiation on several types of concretes commonly used as shielding materials are here investigated, in order to evaluate for each of them not only their shielding capacity against neutrons as source of radiation but also the effects on the mechanical response of the material under such an extreme condition.

A macroscopic neutron behavior is considered to describe the attenuation of radiation in matter; two distinct approximate approaches of the more rigorous transport theory are preferred to the deterministic approach given by the solution of the transport equation by Boltzmann as well as to the stochastic approach by Monte Carlo techniques.

Specifically, the two approaches are: the diffusion theory, which applies to thermal neutrons, and the two-group theory, which allows for getting the fast group equation in a semi-infinite plane geometry [19]. Both of them are developed from the above-mentioned integro-differential equation by Boltzmann, obtained by considering the inflow and outflow balance of particles through the surface of an arbitrary closed volume in a steady state condition for the radiation field. These approaches renounce to solve the problems associated to primary neutrons attenuation, such as the production of photons during inelastic scattering, the capture of thermal neutrons leading to capture gamma photons and even production of secondary neutrons as a result of fission reactions; nevertheless they offer, in their simplified way, acceptable estimates of the expected flux density field [19–21].

The basic hypothesis of the diffusion theory is that the neutron current is isotropic and is given by

$$I = -D\nabla\Phi \tag{1}$$

thus leading to an equation of the type

$$-D\nabla^2\Phi + \Sigma_a\Phi - S = 0 \tag{2}$$

where Φ is the flux density (for instance in [n/(cm^2 s)], S is the source term, Σ_a is the absorption cross-section and D a diffusion coefficient, defined in a simple diffusion theory by

$$D = \frac{\lambda_s}{3} \tag{3}$$

λ_s being the scattering mean free path.

The solution of a plane source in an infinite medium can be derived from this approximated theory by imposing proper boundary conditions, e.g.

$$\Phi(x) = \frac{SL}{2D}e^{-(x/L)}, \quad L = \sqrt{\frac{D}{\Sigma_a}} \tag{4}$$

where L is known as the diffusion length.

This theory is valid deep in the medium, where the flux Φ is often given by multiplied scattered particles and hence it can be expected to be nearly equal in all directions, whereas near a free surface or a source the flux density is quite anisotropic and the diffusion approximation may be poor. Moreover the same hypothesis makes the theory suitable for thermal neutrons rather than fast ones [20].

The second approach becomes useful to describe a simplified behavior for fast neutrons: according to the two-group theory, neutrons in the shield are divided into neutrons at thermal energies and those above. The group diffusion equation (1) is applied to each group, the source term in each group being the neutrons slowed down in the immediately higher energy group. This leads to two equations of the type

$$\begin{aligned} &-D_f\nabla^2\Phi_f + \Sigma_f\Phi_f = 0 \\ &-D_t\nabla^2\Phi_t + \Sigma_t\Phi_t - \Sigma_f\Phi_f = 0 \end{aligned} \tag{5}$$

for the fast and the thermal groups respectively. The fast source term in the shield is zero, whereas the thermal source term equals the fast neutrons scattered into thermal energies ($\Sigma_f\Phi_f$) and, in a similar manner to the diffusion equation (see Eq. (4)), the solution of the fast group equation in a semi-infinite plane geometry is given by

$$\Phi_f(x) = \Phi_0 e^{-\Sigma_R x}, \quad \Sigma_R = \frac{1}{L_s} \tag{6}$$

where Σ_R is the macroscopic removal cross section, which is roughly equal to the reciprocal of the average relaxation length for fast neutrons in the shielding material, and Φ_0 is the fast neutron flux at $x = 0$.

The theory in this case is valid until the attenuation of fast neutrons is dominated by a removal process, i.e. if concrete contains sufficient moderating material (i.e. its hydrogen content) [21].

2.2. The coupled thermo-hygro-mechanical problem

The NEWCON3D F.E. model is based on a series of balance equations, i.e. a mass balance equation of water (both liquid and vapour, taking into account phase changes and hydration/dehydration processes), an enthalpy balance equation of the whole multiphase medium (considering the latent heat of phase change and hydration/dehydration), a linear momentum balance equation of the fluid phases (Darcy's equation) and a linear momentum balance equation of the whole medium.

Appropriate constitutive equations and some thermodynamic relationships are included as well. The field equations of the model are briefly recalled below; for additional details the reader is referred to [8–18].

The continuity equation for non-isothermal flow is expressed in terms of relative humidity as

$$\frac{\partial h}{\partial t} - \nabla^T\mathbf{C}\nabla h - \frac{\partial h_s}{\partial t} - k\frac{\partial T}{\partial t} + \chi m^T\frac{\partial \varepsilon}{\partial t} = 0 \tag{7}$$

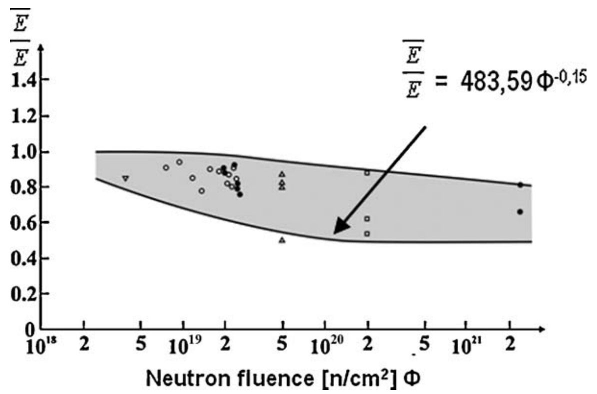

Fig. 1. Modulus of elasticity of concrete after neutron radiation $\bar{E}$ related to modulus of elasticity of untreated concrete E [22].

where h is the relative humidity, directly connected with the moisture content w, T is temperature, $k = \left(\frac{\partial h}{\partial T}\right)\big|_{w,\varepsilon}$ is the hygrothermic coefficient, i.e. the change in h due to one-degree change of T at constant w, ε and a fixed degree of saturation, dh_s is the self-desiccation, $\chi = \left(\frac{\partial h}{\partial \varepsilon_v}\right)\big|_{T,w}$ represents the change in h due to unit change of volumetric strain $\boldsymbol{\varepsilon}_v$ at constant w, T and a given degree of saturation, C is the (relative humidity) diffusivity diagonal matrix.

Heat balance requires

$$\rho C_q \frac{\partial T}{\partial t} - C_a \frac{\partial w}{\partial t} - C_w \mathbf{J} \cdot \nabla T = -div\mathbf{q} \tag{8}$$

where ρ is the mass density of concrete, C_q the isobaric heat capacity of concrete (per kilogram of concrete) including chemically bound water but excluding free water, C_a is the heat of sorption of free water (per kilogram of free water); C_w is the isobaric heat capacity of liquid water, $C_w \mathbf{J} \cdot \nabla T$ is the rate of heat supply due to convection by moving water, $\mathbf{J}$ is the flux of humidity, depending on the gradient of h by means of the permeability c

$$\mathbf{J} = -c\,\nabla h \tag{9}$$

$\mathbf{q}$ is the heat flux which can be due to temperature gradient or moisture concentration gradient. The constitutive law defining the heat flux comes from a Fourier's law (heat flux due to temperature gradient) and a Dufour's flux (heat flux due to moisture concentration gradient)

$$\mathbf{q} = -a_{Tw}\,\nabla w - a_{TT}\,\nabla T \tag{10}$$

where the coefficients a_{Tw} and a_{TT} depend on w and T.

Finally, the macroscopic linear momentum balance equation for the whole medium is to be accounted for

$$\mathbf{div}\quad \sigma + \rho_{\rm m} \mathrm{g} = 0 \tag{11}$$

where ρ_m is the density of the multiphase medium (concrete plus water species) and $\mathbf{g}$ an acceleration related to gravity.

As regards the mechanical field, the constitutive relationship for the solid skeleton in incremental form can be written as

$$d\boldsymbol{\sigma}' = (1-D)\mathbf{D}_T(d\varepsilon - d\varepsilon_{\rm T} - d\varepsilon_{\rm c} - d\varepsilon_{\rm lits} - d\varepsilon_{\rm p} - d\varepsilon_{\rm sh} - d\varepsilon_0) \tag{12}$$

where $\boldsymbol{\sigma}'$ is the effective stress tensor ($\boldsymbol{\sigma}' = \boldsymbol{\sigma} + p\mathbf{I}$, p being the mean pore pressure), directly responsible for deformations of the solid skeleton, D the upgraded scalar radio-chemo-thermo-mechanical damage (see below), $\mathbf{D}_{\rm T}$ the tangent stiffness matrix, $d\boldsymbol{\varepsilon}_{\rm T}$ the strain rate caused by thermo-elastic expansion, $d\boldsymbol{\varepsilon}_{\rm c}$ the strain rate accounting for creep, $d\boldsymbol{\varepsilon}_{\rm lits}$ the load induced thermal strain rate, $d\boldsymbol{\varepsilon}_{\rm p}$ the plastic strain rate, $d\boldsymbol{\varepsilon}_{\rm sh}$ is due to shrinkage and $d\boldsymbol{\varepsilon}_0$ represents the autogeneous strain increments, due, e.g. to chemical variations of the mixture and including the irreversible part of the strain rates not contained in the previous terms. Particularly, D is the damage variable following the scalar isotropic model by Mazars and Pijaudier-Cabot [14].

According to this theory, the classical effective stress concept [15] is modified to take into account damage, measuring a reduction in the resistant area due to cracks beginning and spreading

$$\overline{\boldsymbol{\sigma}'} = \boldsymbol{\sigma}' \frac{S}{\bar{S}} = \frac{\boldsymbol{\sigma}'}{1-D} \tag{13}$$

where S and $\bar{S}$ are the resistant area of the uncracked and cracked material, respectively.

Since the damaging mechanisms are different in uniaxial tension and compression experiments, the damage parameter D_m (the subscript stands for mechanical contributions only) is decomposed into two parts, d_t for tension and d_c for compression, which can be expressed in function of the equivalent strain $\tilde{\varepsilon}$

$$\tilde{\varepsilon} = \sqrt{\sum_{i=1}^{3} (\langle \varepsilon_i \rangle_+)^2} \quad \left(\langle \varepsilon_i \rangle_+ = \frac{|\varepsilon_i| + \varepsilon_i}{2} \right) \tag{14}$$

ε_i being the principal strains. Hence

$$D_m = \alpha_t d_t + \alpha_c d_c \tag{15}$$

where α_t and α_c are weighting coefficients defined in [14].

Chemo-mechanical damage has been introduced for the first time in [16]; thermo-chemical effects have been also taken into

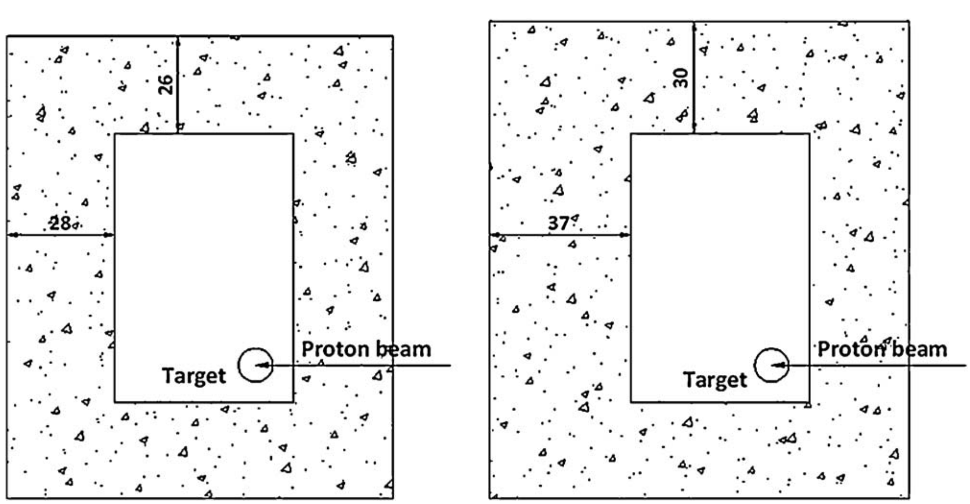

Fig. 2. Cross-section of the target cave with the required shielding thickness [dm] relative to two scenarios for the proton beam and localization of the modelled slice of wall.

Table 1
Elemental composition of the reference mixture for serpentine (a), barytes (b), limonite-steel (c), magnetite (d) concrete, in agreement with [27,29,30].

(a) Serpentine

Element	(wt.%)
MgO	33.4
SiO_2	37.59
$A1_2O_3$	3.42
Fe_2O_3	5.15
FeO	2.04
CaO	4.82
CO	1.65
Na_2O	1.13
H_2O	10.26

(b) Barytes

Element	(g/cm^3 of concrete)
H in water	0.0243
O in water	0.195
O in ore	0.872
O in cement	0.118
Mg in cement	0.00385
Al in cement	0.0137
Si in cement	0.0352
S	0.364
Ca in ore	0.0203
Ca in cement	0.147
Fe in ore	0.151
Fe in cement	0.0091
Ba	1.551

(c) Limonite-steel

Element	(g/cm^3 of concrete)
H	0.028
O	0.806
Mg Al	0.039
Si	0.078
Ca	0.25
Ti	–
Mn	–
Fe	3.03

(d) Magnetite

Element	(wt.%)
Fe	55.9
O	34.4
Ca	6.6
Si	1.6
H	0.7
Al	0.6
Mg	0.2

account in multiplicative way, as proposed by Gerard et al. [17] and Nechnech et al. [18]: a damage parameter D_{tc}, $0 \leq D_{tc} \leq 1$, describes thermo-chemical material degradation at elevated temperatures (mainly due to micro-cracking and cement dehydration) resulting in reduction of the material strength properties, so that Eq. (13) becomes:

$$\overline{\boldsymbol{\sigma}'} = \frac{\boldsymbol{\sigma}'}{(1 - D_m)(1 - D_{tc})} \tag{16}$$

As mentioned before, the total effect of the mechanical and thermo-chemical damages acting at the same time is multiplicative, i.e. the total damage D is defined by

$$D = 1 - (1 - D_m)(1 - D_{tc}) \tag{17}$$

The upgrade of the model has been developed by assuming that the nuclear radiation can activate a damage process combined with the mechanical and thermo-mechanical ones so that the above multiplicative relation is maintained and the total damage is redefined

$$D = 1 - (1 - D_m)(1 - D_{tc})(1 - D_r) \tag{18}$$

in which D_r accounts for radiation damage, whose evaluation is empirically based, as reported in the following section.

The application, within the numerical code NEWCON3D, of a standard finite element discretization in space of Eqs. (7), (8) and (11) results in

$$\begin{bmatrix} \mathbf{K} & \mathbf{HU} & \mathbf{TU} \\ \mathbf{L}^{\mathrm{T}} & \mathbf{I} & \mathbf{TP} \\ \mathbf{0} & \mathbf{TH} & \mathbf{TS} \end{bmatrix} \begin{Bmatrix} \dot{\bar{\mathbf{u}}} \\ \dot{\bar{\mathbf{h}}} \\ \dot{\bar{\mathbf{T}}} \end{Bmatrix} + \begin{bmatrix} \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{Q} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{TR} \end{bmatrix} \begin{Bmatrix} \bar{\mathbf{u}} \\ \bar{\mathbf{h}} \\ \bar{\mathbf{T}} \end{Bmatrix} = \begin{Bmatrix} \dot{\mathbf{f}} + \mathbf{c} \\ \dot{\mathbf{HG}} \\ \dot{\mathbf{TG}} \end{Bmatrix} \tag{19}$$

in which $\bar{\mathbf{u}}$, $\bar{\mathbf{h}}$ and $\bar{\mathbf{T}}$ are the nodal values of the basic variables: displacements, relative humidity and temperature, while the expressions in capital letters refer to vectorial/matricial notation [8,10,11]. **HU** and **TU** account for shrinkage and thermal dilation effects, respectively; $\mathbf{L}^{\mathrm{T}}$ and **TP** are the coupling matrices representing the influence of the mechanical and thermal field on the hygral one, respectively; **Q** is the diffusivity matrix accounting for sorption-desorption isotherms; **TH** the coupling matrix between the hygral and thermal fields in terms of capacity; **TS** the matrix of heat capacity; **TR** the matrix of thermal transmission including the convective term; **c** the matrix accounting for creep; **HG** includes humidity variation due to drying and **TG** accounts for heat fluxes.

For further explanations of the above terms the reader is referred to [8,10,11].

It has to be noticed that the outlined above assumptions on the radiation field enter the model in an indirect way via the damage variable which is dependent on radiation as explained in the next Section; such a simplified approach necessarily uncouples the radiation field form the hygro-thermal ones, being instead maintained the coupling with the mechanical field through the damage variable itself. However, at this stage any other assumption could be equally questionable in the absence of more complete experimental data relating the influence of radiation diffusion within concrete on e.g. concrete moisture content and temperature, and vice versa.

3. Radiation damage in concrete

Radiation on shielding materials is known to affect them in their chemical structure. For crystalline materials, radiation provides the displacements of atoms from their equilibrium lattice sites, causing lattice defects responsible for embrittlement of the material. For polymers the same result is given by the formation of additional cross-links due to the energy brought by radiation. For geomaterials, like concrete, nuclear radiation leads to the break down of atomic bounds, which is supposed to explain the decay in the mechanical properties.

Particularly, an increase in aggregates volume is envisaged, after radiation exposure. Quartz aggregates that contain crystals with covalent bonding appear to be more affected by radiation than calcareous aggregates containing crystals with ionic bonding. Neutron fluences of the order of 1×10^{19} n/cm^2 seem to become critical for concrete strength [21–23].

There is also evidence [23,24] that nuclear radiation significantly increases the reactivity of silica-rich aggregates to alkali; the decrease in resistance to nuclear radiation with increasing content of SiO_2 in aggregates strongly indicates that the deterioration is due to the acceleration of such a reaction in concrete and can partly explain the degradation of its mechanical properties.

The mechanism consists in a series of chemical reactions: OH^- ions of the alkaline solution in the micropores of concrete react with

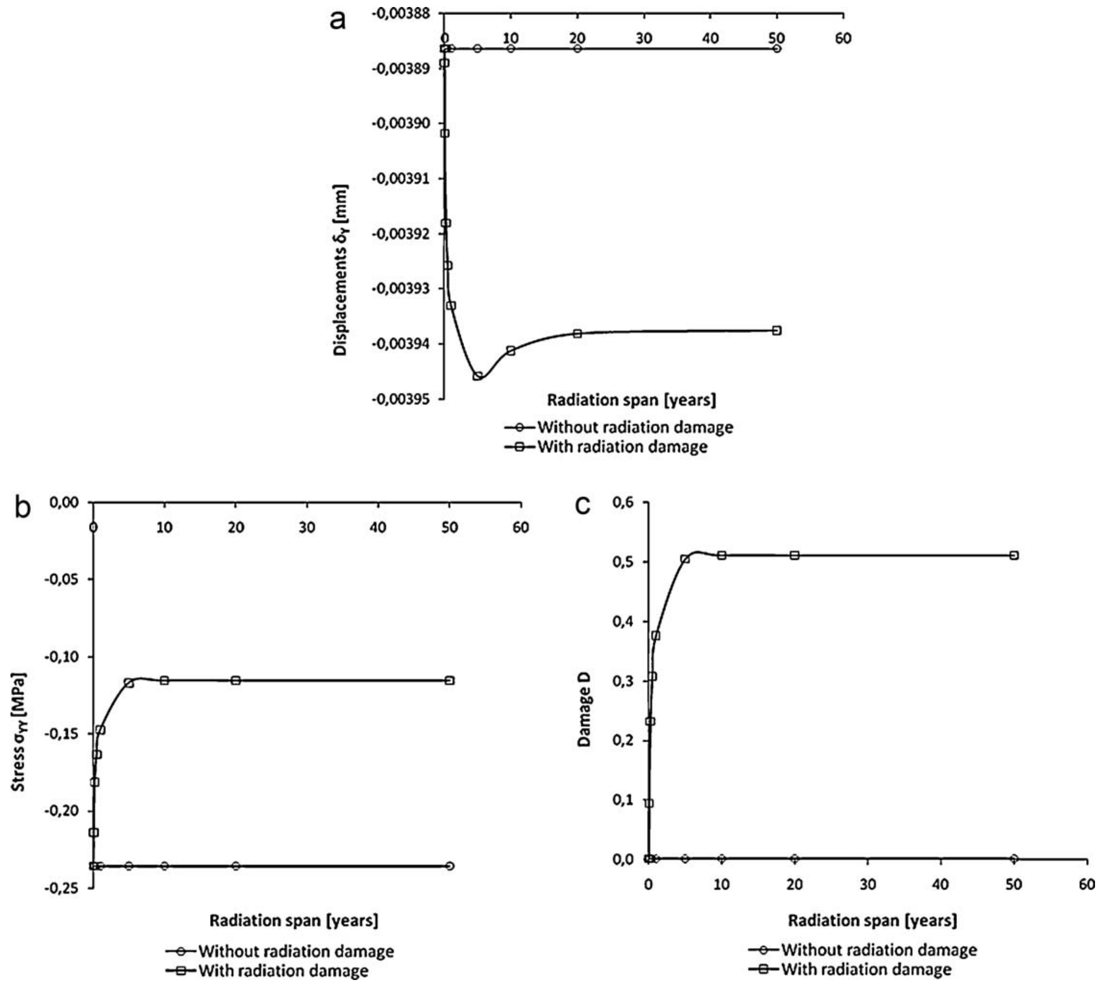

Fig. 3. Displacements δ_Y (a), principal stress σ_{YY} (b) and total damage D (c) as functions of the radiation span on OPC, very close to the directly exposed surface, both in the case that radiation damage is or not activated.

SiO_2 in aggregates, causing the scission of the Si–O bonding and the subsequent expansion of the aggregates by hydration of SiO_2. The consumption of OH^- ions, due to these scissions, leads to the dissolution of Ca^{2+} ions into the solution. The Ca^{2+} ions then react with hydrated SiO_2 gels to generate calcium silicate. Rigid calcium silicate shells are therefore formed on the surfaces of the aggregates by successive reactions with OH^- and Ca^{2+} ions and the alkaline solution is possible to penetrate into the aggregates through the calcium silicate shells and to dissolve SiO_2. Since the rigid shells prevent the aggregates deformation, the expansion pressure generated by the penetration of the solution is accumulated within them, finally leading to aggregates cracks and concrete expansion.

The alkali-silica reaction of the aggregates may be accelerated both by lattice defects in SiO_2 minerals from fast neutrons irradiation or small cracks pre-generated in aggregates.

In the present work radiation damage is given via phenomenological description, thanks to a collection of experimental results by Hilsdorf et al. [22] providing a decay law for the elastic modulus of concrete under irradiation. Fig. 1 shows the effect of neutron radiation on the modulus of elasticity of concrete; the modulus of irradiated concrete $\bar{E}$ is given as a fraction of the modulus of companion specimens E neither irradiated nor temperature exposed. A neutron fluence lower than 1×10^{19} n/cm^2 is shown to bring a slight decrease in the modulus of elasticity compared to the modulus of untreated companion specimens. With increasing neutron fluence the $\bar{E}/E$ ratio decreases, asymptotically reaching 50%.

The scattering emerging from the comparison of different test series is explained by the large variety of test conditions: concrete making materials, mix proportions of mortars, specimens size, cooling and drying conditions, impinging of fast or

Table 2
Assumed values for mechanical and shielding properties of the analyzed concretes, in agreement with [21,27,29,30].

Type of concrete	Density ρ [kg/m^3]	f_{ck} [MPa]	E [MPa]	Σ_R [cm^{-1}]	L [cm]
Ordinary	2.33	25	32.484	0.0787	8.0434
Serpentine	2.09	19.3	24.069	0.0836	2.11
Barytes	3.5	24.8	31.157	0.0819	5.4928
Ferro-phosphorus	4.65	30.4	28.200	0.119	2.2045
Limonite-steel	4.27	38.4	47.900	0.1158	2.1016
Magnetite	3.41	41.8	57.800	0.1061	3.0367

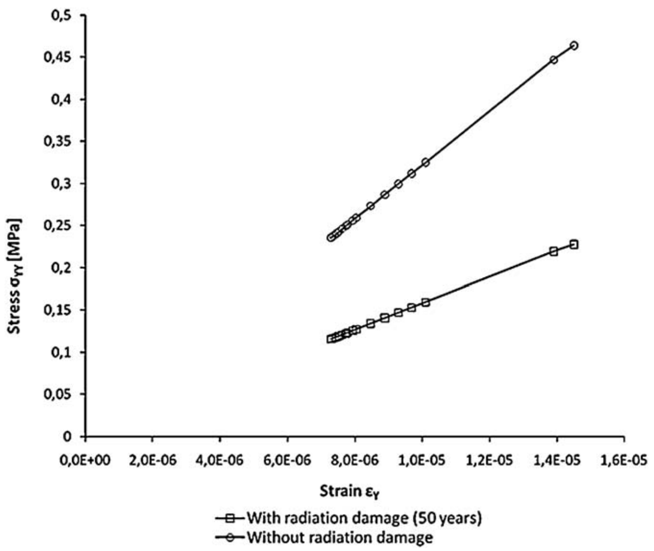

Fig. 4. Stress–strain relation along direction *Y*, loading direction, for OPC, near the most exposed surface, after a 50-years long radiation span.

slow neutrons, possible simultaneous temperature exposure of specimens.

The available data are not sufficient to separate the effects of radiation and of heating of the samples, which in many cases they undergo in nuclear vessels conditions, however it seems reasonable to observe that the strength loss is primarily due to neutron radiation, in analogy with similar graphs for the compressive and the tensile strength, for which a separation of the two effects was possible [22].

The lowest enveloping curve has been taken as reference (in favour of safety) to define the radiation damage parameter $(1-D_r)$ as the reported ratio between the two elastic moduli, in agreement with the effective stress theory.

Looking at the mean behavior and as stated above, it can be observed that a neutron fluence less than 10^{19} n/cm^2 leads to a slight decrease in the $\bar{E}/E$ ratio, then reaching 50% under increasing neutron fluxes. Hence it has been supposed that under neutron fluxes of 10^{18} n/cm^2 and below radiation damage is zero.

3.1. Numerical analyses

The case study takes its origin from the SPES Project that is currently being developed at the National Laboratories of Legnaro (Padua, Italy). The facility will be directed to the production of special radioactive heavy ion beams from a primary proton beam impinging on a target made of fissionable material, where fission reactions are expected to take place, thus ideally representing a point-source of neutrons for the specific problem.

The analyses have been performed to simulate the response of a concrete structure during about 50 years by assuming a neutron flux density of 10^{12} n/(cm^2 s), which is nearly twice the one expected for SPES and quite less than the radiation dose reached by nuclear cores (even of the order of 10^{19} n/(cm^2 s)). The radiation flux has been supposed to be directly transmitted, unaltered, to the inner surface of the shielding, being negligible the distance between the target and the concrete wall.

Two extreme situations have been considered for neutron energies: the case in which the whole flux is given by fast neutrons and the one in which it comes totally from thermal neutrons, in order to predict the minimum and maximum threshold for radiation damage under a realistic variegated neutron spectrum, which should represent an intermediate solution.

In our approach the attenuation process of neutrons in concrete is considered to be one-dimensional through the thickness of the shielding slab. This assumption is not restrictive if we consider the geometry of the problem, i.e. the diffusion of neutrons from a neutron source through a wall; the wall is modelled for a small portion, qualitatively situated within the bunker building, in agreement with the design drawings provided by INFN. The neutron fluence is supposed to proceed outwards from the room through the shielding medium.

The sample analyzed is a prism 3.5 m long, with a square cross-section of 1 m^2. The thickness of the model comes from radioprotection issues on the SPES facility involving the target cave that is to be kept under 0.25 mSv/h ambient dose equivalent [3].

The indicated thickness relative to two different work scenarios impinging on the UC_x target (radioactive ion beam of 40 MeV, 200 μA and of 70 MeV, 350 μA) are reported in Fig. 2, with the hypothetical position of the modelled portion of wall of the target cave.

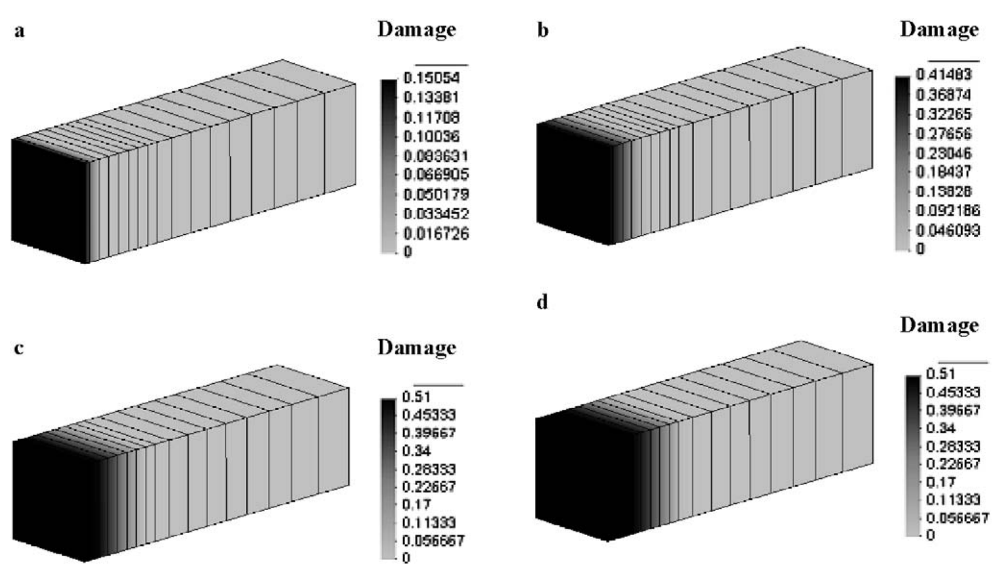

Fig. 5. Radiation damage progression with depth of the sample after a 1-month (a), 1-year (b), 10-years (c), 50-years (d) radiation fluence, from fast neutrons on OPC.

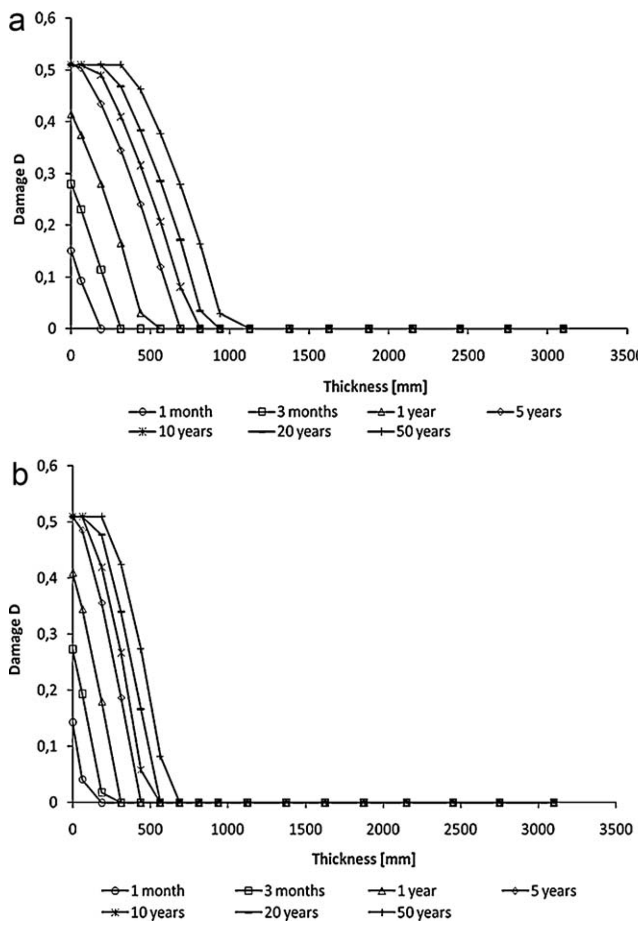

Fig. 6. Radiation damage progression with depth of the sample under radiation fluence up to 50 years, from fast neutrons (a) and thermal neutrons (b) on OPC.

The slice under analysis has been discretized by 20-nodes brick elements, with constraints on the base surface; self-weight has been accounted for as well.

The materials considered in the analysis are discussed below and their mechanical and shielding properties are summarized at the end of this section, with respect to a given mixture or average values from different mixtures of the same material, the latter being the case for ordinary concrete (OPC; see Tables 1 and 2).

We point out that the Young's modulus has been derived from the empirical relation proposed by the national standard [25,26] as a function of the compressive cylinder strength, when not provided in literature

$$E_{lcm} = 22,000\left(\frac{f_{lcm}}{10}\right)^{0,3}\eta_{E}, \quad \eta_E = \left(\frac{\rho}{2200}\right)^2 \tag{20}$$

where E_{lcm} is the average value of the secant elastic modulus after 28 days curing [MPa], f_{lcm} is the average compressive cylinder strength [MPa], ρ is the density of concrete [kg/cm^3]. Concrete is supposed to behave elastically under its self-weight.

As for OPC, the average values of the main parameters are taken from eight different mixtures reported by Kaplan [21]; the assumed values are listed in Table 2.

Serpentine is a hydrous magnesium iron phyllosilicate ((Mg, Fe)$_3$Si$_2$O$_5$(OH)$_4$) mineral. It is used as aggregate in concrete because, as a hydrous aggregate, it can retain most of its water of crystallisation at temperatures up to about 500 °C, thus keeping its efficiency against neutrons also in the extreme conditions of nuclear reactors. The reference values are taken from the serpentine concrete investigated by Ohgishi et al. [27].

Barytes (barium sulphate – $BaSO_4$) is used both as coarse and as fine aggregate for concrete. The mean volumetric weight of barytes concrete is about 50% higher than that of OPC and, therefore, it is more efficient than the latter against γ-radiation. Additionally this high specific weight aggregate has almost null reactivity with alkalis in the cement, which is understood to be one of the main causes of degradation of concrete during irradiation [23,24]. Moreover the high neutron capture cross-section of barium ensures good shielding properties even against neutrons, though also limonite or other hydrous aggregates can be used in conjunction to improve them [28]. The assumed data for barytes concrete are taken from Gallaher and Kitzes's report [29].

Ferro-phosphorus aggregates help reaching high densities on the mortars, thus ensuring good efficiency against photons. A volume increase at temperatures above 350 °C has been observed [21], which has been stated to be due to accelerated rates of oxidation of the aggregate. It has also been reported that ferro-phosphorus concretes may lead to the production of inflammable gases that could develop high pressures if confined and that they experience delay in setting and hardening. The quantities of interest, herein, refer to ferro-phosphorus concretes used for the N-Production Reactor at Hanford in the USA [30]; no chemical composition has been found for it.

Limonite (FeO(OH)·nH_2O) ores are hydrous iron ores used in concrete primarily because of their high content of chemically combined water (twice as high a concentration of water as an OPC may be achieved), thus improving the effectiveness of shielding against neutron flux. Limonite retains its water of crystallisation up to 200 °C. Fine limonite aggregate which gives sufficient plasticity to the concrete mix for casting is used for heavy concretes with the addition of metal scrap (steel punching, sheared bars, steel shots). The collected data are of a limonite-steel from Hanford [30].

Magnetite (Fe_3O_4) is used as heavy aggregate for special concrete. Its water content is not as high as in limonite and, consequently, it has lower shielding properties. The reference mortar is taken again from [30].

In Tables 1 and 2 the available chemical compositions of the investigated improved concrete mixtures and the mechanical and nuclear properties of the reference mortars have been reported, respectively.

3.2. Numerical results

The results have been collected mainly in terms of damage parameter vs. distance. Due to the negligible effect of compression from self-weight load, damage can be considered as totally due to radiation. The results are reported in Fig. 3 in terms of displacements δ_Y, principal stress σ_{YY} (with Y loading direction) and total damage D as functions of the irradiation time on OPC; the results refer to a node belonging to the directly impinged concrete surface. The radiation field is characterized by a neutron flux density of 10^{12} n/(cm^2 s), integrated on 50 years duration span.

Damage reaches its maximum (50%) after 5–10 years and correspondingly stresses reach their equilibrium value; differently, a delay in the occurrence of the asymptotic (steady) value for displacements is shown, accompanied by the occurrence of a moderate expansion after the peak value: such a behavior is referable to concrete creep. The stress state is strongly affected by radiation, generating the inability of concrete for sustaining even low compression values at the expense of larger deformations. Longer radiation exposure leads to more pronounced trends.

In agreement with the empirical curves of Fig. 1, the obtained stress–strain curve is shown in Fig. 4, where a 50% decay in the elastic modulus of concrete is evidenced (as expected) due to radiation damage.

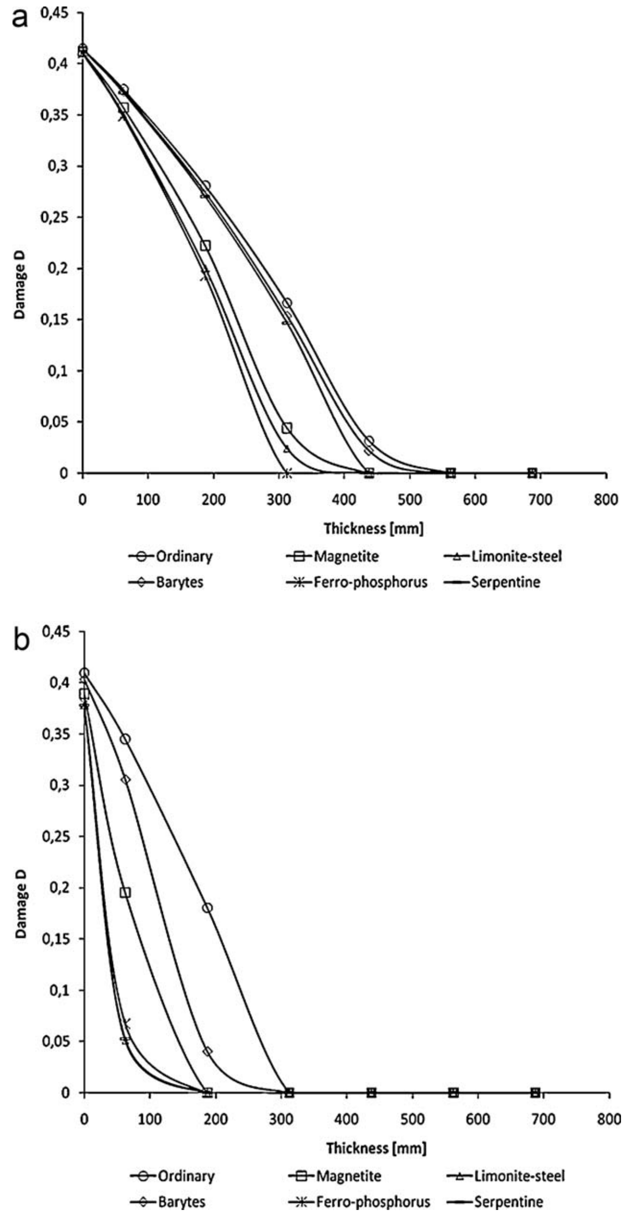

Fig. 7. Radiation damage progression with depth of the sample under the radiation fluence of 1 year, from fast neutrons (a) and thermal neutrons (b) on ordinary and special shielding concretes.

Following the diffusion theory for thermal neutrons and the two-group theory for fast neutrons, the damage behavior as function of depth for OPC has been investigated. Contour maps of damage relative to 1 month, 1 year, 10 years and 50 years radiation (fast neutrons) are reported in Fig. 5, whereas Fig. 6 shows the influence of thermal and fast neutrons on the development of damage vs. distance from the exposed surface: when fast neutrons are impinging, the damaged thickness is more pronounced and, specifically, the amount of the damaged thickness induced by fast neutrons exceeds the one induced by thermal neutrons of 80% of the latter, at 5 years radiation and of 70% of it, at 50 years radiation.

Special concretes (i.e. improved mixtures) have been additionally accounted for in the following, for a fast and a thermal neutron flux of 10^{12} n/(cm^2 s) integrated in 1 year exposure, respectively (Fig. 7): all the mixtures show better shielding properties than OPC, particularly magnetite, limonite-steel and ferro-phosphorus for fast neutrons and serpentine, limonite-steel and ferro-phosphorus for thermal neutrons.

4. Conclusions

Nuclear radiation both in the form of fast and thermal neutrons is known to affect concrete in its mechanical behavior over peculiar threshold quantities of radiation fluence. To our purposes, radiation in the form of neutron particles has been of particular interest, since the estimate of durability performances of the shielding walls for SPES target room, where the fissionable target represents the neutron source, is the expected utility of this study.

A collection of the most relevant experimental results on neutron irradiated concrete has been exploited to derive a formulation for radiation damage in the context of damage mechanics for concrete materials. An upgraded parameter has been introduced in a F.E. research code assessing the coupled hygro-thermo-mechanical behavior of concrete. Total damage is the result of a multiplicative relationship, accounting for radiation, as well as thermo-chemo-mechanical damage, the latter being already implemented in the constitutive law of the material, so that the effects are independent on each other, when not present simultaneously.

Results in terms of damage have been achieved, also taking into account special concrete commonly used for reactor shielding, which allow for defining up to a 50-years long radiation exposure, when a neutron flux density of 10^{12} n/(cm^2 s) is supposed to impinge the biological shielding under the assumptions of the approximated diffusion theory and two-groups theory for thermal and fast neutron fluences, respectively.

The numerical code in its upgraded form, taking into account the effect of radiation damage, is thus expected to describe the mechanical response of concrete when irradiated; in particular, under the considered irradiation scenario, OPC has shown a decrease in its elastic modulus of nearly 50%, thus providing a low strength capacity to external loads; when considering improved mixtures, under equal fluences, strength is less affected by damage so that special concretes are preferable not only for shielding requirements but also from a purely mechanical point of view.

Further steps in the research will focus on the coupling between radiation and hygro-thermal fields to check the coupling effects of prolonged irradiation exposures on concrete behavior.

Finally, as an alternative approach, Monte Carlo stochastic techniques will be considered in future to catch the complexity of the interaction of radiation with concrete, also in terms of collateral reactions to its attenuation on the shielding material.

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