Radiation damage in the target area shielding of a facility for Selective Production of Exotic Species (SPES Project)

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SUMMARY. The advancement of radioactive ion beam (RIB) science since its advent in the last few decades calls for the development of so-called next generation facilities, able to provide beam intensities several orders of magnitude higher than presently available. Two of them are planned by the European roadmap and are representative of the two main methods by means of which RIBs are produced nowadays: projectile fragmentation and isotope separation on-line (ISOL); the latter, based on the ISOL technique, is the EURISOL project: the plan encompasses the construction of three intermediate facilities, one of which is expected to be built at Legnaro (Padua) INFN Laboratories: the SPES Project (Selective Production of Exotic Species).

1 INTRODUCTION

SPES is an accelerator based facility intended to provide intense production of neutron-rich nuclei far from the beta stability, in the mass range 80≤A≤160amu, which represents a vast and still unexplored territory for nuclear physics experiments, and to perform forefront research in nuclear structure, reaction dynamics and interdisciplinary fields like medical, biological, astrophysics and science of materials.

In the proposed configuration a primary proton beam from a cyclotron as proton driver accelerator (15-70MeV, 0,75mA) directly impinges on uranium carbide disks; the exotic nuclei are produced as a result of high energy fissions at rate 10^{13}f/s induced on ^{238}U compounds of the target in a dedicated area located below ground level. The isotopes are then extracted from the target and ionized with a ion source connected to the production target; they are consequently charge breeded and mass separated to fit the required charge state and ion velocity for injection into the system: pre-accelerator PIAVE and further acceleration by the ALPI linac, from which the secondary beam is delivered to the experimental areas [1].

The shielding for the cyclotron cave and the direct target bunkers depends on the required minimum dose equivalent of 1mSv per year in the surrounding environment.

In the present study we investigate the effects of radiation on concrete shielding in order to enter with a properly defined variable of damage a numerical code modeling the coupled hygro-thermo-mechanical response of such a structural material.

2 THE MATHEMATICAL MODEL

Within the code NEWCON3D (at the Department of Structural and Transportation Engineering, University of Padua) [2, 14-17] concrete is modeled 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 (uncondensable constituent) and water vapour (condensable), supposed to behave like an ideal gas.

When higher than standard temperatures are taken into account several phenomena are to be considered: heat conduction, vapour diffusion and liquid water flow due to pressure gradients or capillary effects caused by the induced meniscus curvature in the voids, index of an altered equilibrium between liquid water and the partial pressure of water vapour. As for the mechanical field the model couples shrinkage, creep, damage and plasticity effects.

The model consists of the following balance equations: balance equation of dry air, mass balance equation of water (both liquid and vapour taking phase change and hydration/dehydration process into account), an enthalpy balance equation of the whole multiphase medium (considering the latent heat of phase change and the hydration/dehydration process), the linear momentum balance equation of the fluid phases (Darcy's equation), the linear momentum balance equation of the whole medium.

Several appropriate constitutive equations and some thermodynamic relationships are needed. The basic model results in some simplifications: the capillary effects are neglected, so that the two gas and water pressures coincide and the mass balance equations can be reduced into one; the latent heat of evaporation is neglected, so that the energy balance equation is simplified; the linear momentum balance equations of the fluids are substituted by a constitutive equation for fluences. Obviously enhanced and enriched versions of the model, where capillary effects, etc. are recovered, are also available.

The model is thus expected to solve the coupled thermo-hygro-mechanical problem for the porous multiphase material under medium and high temperatures.

2.1 The mechanical field

Dealing with porous media we refer to the effective stresses, $\boldsymbol{\sigma}'$, directly responsible for all deformations of the solid skeleton; the constitutive relationship for the solid skeleton in incremental form in terms of the new entity, when damage of the material is included, becomes:

$$\mathrm{d}\boldsymbol{\sigma}' = (1\text{-D})\, \mathbf{D}_\mathrm{T}\, (\mathrm{d}\boldsymbol{\varepsilon} - \mathrm{d}\boldsymbol{\varepsilon}_\mathrm{T} - \mathrm{d}\boldsymbol{\varepsilon}_\mathrm{c} - \mathrm{d}\boldsymbol{\varepsilon}_\mathrm{lits} - \mathrm{d}\boldsymbol{\varepsilon}_\mathrm{p} - \mathrm{d}\boldsymbol{\varepsilon}_\mathrm{sh} - \mathrm{d}\boldsymbol{\varepsilon}_0), \tag{4}$$

where D represents the total damage that will be defined later on, $\boldsymbol{D}_T$ is the tangent stiffness matrix, $\mathrm{d}\boldsymbol{\varepsilon}_\mathrm{T}$ is the strain rate caused by thermo-elastic expansion, $\mathrm{d}\boldsymbol{\varepsilon}_c$, the strain rate accounting for creep, $\mathrm{d}\boldsymbol{\varepsilon}_{lits}$ the load induced thermal strain rate, $\mathrm{d}\boldsymbol{\varepsilon}_p$ the plastic strain rate, $\mathrm{d}\boldsymbol{\varepsilon}_{sh}$ is due to shrinkage and $\mathrm{d}\boldsymbol{\varepsilon}_0$ represents the autogeneous strain increments (e.g. due to chemical variations) and the irreversible part of the strain rates not contained in the previous terms. In particular we would like to focus on the implemented parameter of damage in the model to extend its applicability for irradiated concretes, according to what has been shown from most relevant experimental tests since now.

2.1.1 Chemo-thermo-mechanical and radiation damage

Mechanical damage of concrete is considered following the scalar isotropic model by Mazars. In this model, the damaged material at given temperature T is supposed to behave elastically and to remain isotropic. Its Young's modulus at such temperature $E(T)$ can be obtained from that of the mechanically undamaged material at the same temperature $E_0(T)$ and a mechanical damage parameter d, $0 \leq d \leq 1$, being a measure of cracks' volume density within the material:

$$E(T) = (1 - d)E_0(T). \quad (5)$$

According to this theory, the classical effective stress concept is modified to take into account damage, measuring a reduction in the resistant area due to cracking as follows:

$$\tilde{\boldsymbol{\sigma}} = \boldsymbol{\sigma}' \frac{S}{\tilde{S}} = \frac{\boldsymbol{\sigma}'}{1-d}, \quad (6)$$

where *S* and $\tilde{S}$ are the resistant areas of the uncracked and cracked material, respectively.

Since the damaging mechanisms are different in uniaxial tension and compression experiments, the expression for the stress becomes:

$$\sigma_i = \left\{ \frac{(1 - A_i)K_0}{\tilde{\varepsilon}} + \frac{A_i}{\exp\left[B_i\left(\tilde{\varepsilon} - K_0\right)\right]} \right\} \varepsilon_i \qquad (i = t, c), \quad (7)$$

in which the constants A_t, A_c, B_t, B_c are characteristics of the material and can be identified through compression and tension tests on cylinders or bending tests on beams; K_0 is the initial value of the hardening-softening parameter *K(D).*

The damage parameter *d* results this way in a decomposition into two parts, d_t for tension and d_c for compression, which are function of the equivalent strain:

$$\tilde{\varepsilon} = \sqrt{\sum \left(\langle \varepsilon_i \rangle_+\right)^2} \qquad \left(\langle x \rangle_+ = \frac{|x| + x}{2}\right) \quad (8)$$

ε_i being the principal strains. Hence for damage parameter:

$$d = \alpha_t d_t + \alpha_c d_c, \quad (9)$$

where α_t and α_c are weighting coefficients.

Thermo-chemical effects have been taken into account in multiplicative way, as already proposed by Gerard and Nechnech: 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.

Radiation damage d_r is supposed to enter the model in a multiplicative way as well, affecting the unirradiated quantities: $\boldsymbol{\sigma}$ and *E* as suggested by scientific literature about influence of radiation with matter.

Hence, eq. (6) becomes:

$$\tilde{\boldsymbol{\sigma}} = \frac{\boldsymbol{\sigma}'}{(1-d)(1-d_{tc})(1-d_r)}, \quad (10)$$

thus leading to the definition of total damage *D* reported below:

$$D = 1 - (1 - d)(1 - d_{tc})(1 - d_r). \quad (11)$$

Damage enters also permeability, because due to the complex physical and chemical processes taking place especially at high temperature, a change of the inner structure appears accompanied by microcracks development and porosity increase. Hence, permeability increases since it is supposed to be governed by a joint effect of temperature, gas pressure and material damaging.

3 RADIATION DAMAGE

Nuclear radiation may influence structural and mechanical properties of materials significantly; Hilsdorf *et al.* [4] have collected published experimental data on the effect of nuclear radiation on the properties of concrete. It stands out that up to integrated neutron fluences of the order of $1x10^{19}n/cm^2$ the effects of the irradiation are relatively small [4-7], while higher fluences may have detrimental effects on concrete strength and modulus of elasticity. Thermal expansion coefficient, thermal conductivity and shielding properties are less affected by radiation.

Radiation damage is mainly caused by lattice defects in the aggregates which cause a volume increase of aggregates and concrete, thus leading to fractures.

Different aggregates show different radiation resistance so that the selection of suitable aggregates is the most important parameter in the definition of a radiation resistant concrete.

3.1 Effect of neutron radiation on concrete

3.1.1 Concrete compressive strength

In figure 1a the compressive strength of concrete samples f_{cu} from various test series as a fraction of the compressive strength of companion specimens f_{cuo}, which were neither irradiated nor temperature exposed is reported, while in figure 1b the same strength values are presented but with the concrete compressive strength related to the strength of companion specimens, f_{cuT}, which were not irradiated but temperature exposed. From this collection of published experimental results it may be concluded that some concretes can resist neutron radiation of more than $5x10^{19}n/cm^2$ without a strength loss while others exhibit a strength loss at a considerably smaller radiation dose. As an average a neutron fluence of more than $1x10^{19}n/cm^2$ leads to a marked decrease of the compressive strength of concrete, even though also for a neutron fluence of less than the above mentioned one the strength ratios may be less than one.

A comparison of the two figures indicates that the observed strength loss is primarily due to neutron radiation though some detrimental effect of the temperature increase during radiation is apparent. The experimental data vary over a wide range even for a given neutron fluence. For a neutron fluence of $5x10^{19}n/cm^2$ the strength ratios range from 0,72 to 1,05 and from 0,65 to 1,05 for f_{cu}/f_{cuo} and f_{cu}/f_{cuT} respectively. Fast neutrons seem to induce the greater decreases.

There is also evidence that concrete hardens under irradiation [6, 8] in fact the interference of penetrating ionizing radiation with the process of setting of cement paste has been found in the form of increased compressive and tensile strength, especially at the beginning of the hardening period.

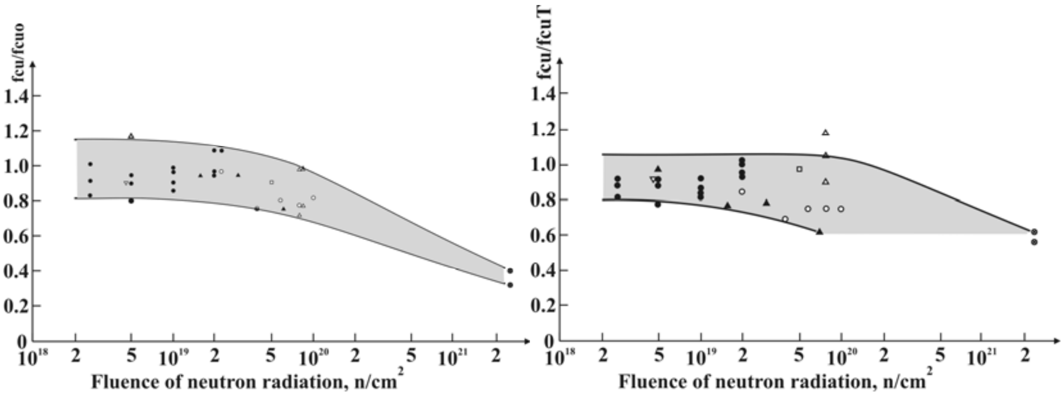

Figure 1: a) Compressive strength of concrete exposed to neutron radiation f_{cu} related to strength of untreated concrete f_{cuo}; b) Compressive strength of concrete exposed to neutron radiation f_{cu} related to strength of temperature exposed concrete f_{cuT} [4].

3.1.2 Concrete tensile strength

The effect of neutron radiation on the tensile strength f_{ru} of concrete samples is shown in figures 2a and 2b. The first gives the tensile strength of concrete samples after neutron radiation as a fraction of the tensile strength of companion specimens f_{ruo}, which were neither irradiated nor temperature exposed, whereas in 2b the tensile strength is related to the strength of non irradiated but temperature exposed specimens, f_{ruT}.

According to figure 2a neutron radiation with a fluence of more than $1\text{x}10^{19}$n/cm^2 may lead to a marked decrease of concrete tensile strength (reductions greater than that for compressive strength, at the same neutron fluence), while comparing the two figures it stands out that temperature exposure is not uniquely responsible for the strength loss but rather neutron radiation has caused a considerable part of the observed strength reduction.

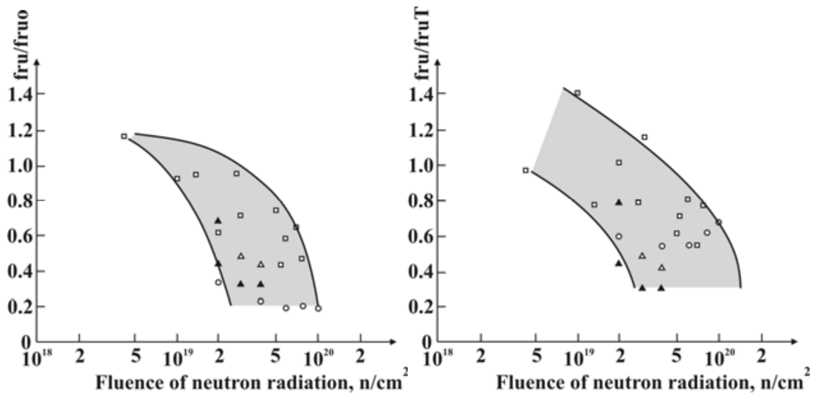

Figure 2: a) Tensile strength of concrete exposed to neutron radiation fru related to strength of untreated concrete fruo; b) Tensile strength of concrete exposed to neutron radiation fru related to strength of temperature exposed concrete fruT [4].

Also for the tensile strength the individual strength values vary over a wide range: for a neutron fluence of $5x10^{19}n/cm^2$ the observed strength ratios range between 0,2 and 0,82, 0,33 and 0,98 for f_{ru}/f_{ruo} and f_{ru}/f_{ruT} respectively.

In both cases the large scatter observed in the experimental data reported so far can be attributed most likely to differences in the composition of the samples and concrete making materials, apart from test procedures.

3.1.3 Modulus of elasticity

Figure 3 shows the effect of neutron radiation on the modulus of elasticity of concrete; the modulus of irradiated concrete E_c is given as a fraction of the modulus of companion specimens E_{co} neither irradiated nor temperature exposed.

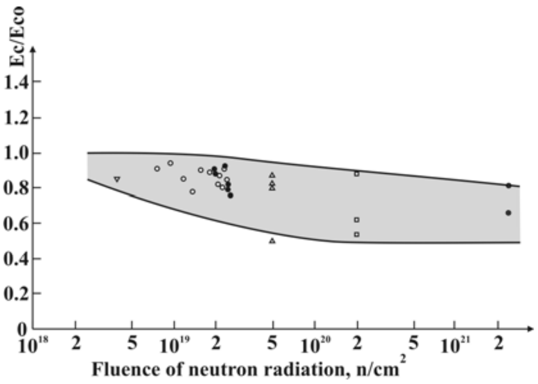

Figure 3: Modulus of elasticity of concrete after neutron radiation E_c related to modulus of elasticity of untreated concrete E_{co} [4].

A neutron fluence less than $1x10^{19}n/cm^2$ again leads to a slight decrease of the modulus of elasticity compared to the modulus of untreated companion specimens. With increasing neutron fluence the modulus of the concrete decreases.

Discerning between fast or slow neutrons action, Gray [9] found that for fast neutron fluence between $7x10^{18}$ and $3x10^{19}n/cm^2$ the modulus of irradiated concrete was between 10 and 20 percent less than that of unirradiated unheated concrete. Alexander [10] reported similar reductions in values of E for slow neutron fluences of about $2x10^{19}n/cm^2$.

It seems likely that for neutron fluences exceeding $10^{19}n/cm^2$ the modulus of elasticity of concrete is less than that of concrete which has not been exposed to nuclear radiation nor subjected to heat treatment.

3.1.4 Thermal conductivity and thermal expansion

The results from [9] are shown in figure 4 for flint aggregate (circles), limestone aggregate (squares), light weight aggregate (crosses), where the thermal conductivity K_c of irradiated samples is given as a fraction of the thermal conductivity of non irradiated companion specimens K_{co} is given as a function of neutron fluence to which the concrete was exposed.

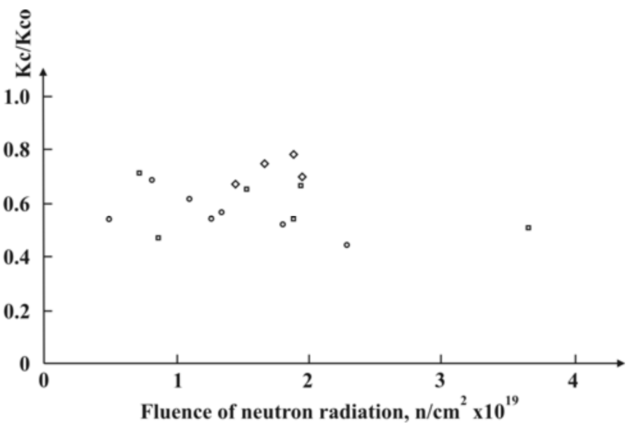

Figure 4: Thermal conductivity of concrete after neutron radiation K_c related to thermal conductivity of untreated concrete K_{co} [4].

According to the figure thermal conductivity decreases between 20 and 50 percent than that of companion unheated unirradiated specimens, as a consequence of the prevailing exposure conditions.

As for the coefficient of thermal expansion Hilsdorf *et al.* and Granata and Montagnini [4, 6] indicate that for neutron fluence less than $5x10^{19}$n/cm^2 there is no significant difference between the coefficient of thermal expansion of neutron-irradiated concrete samples and the coefficient of unirradiated samples which have been exposed to temperatures.

3.2 Parameters affecting the resistance of concrete against nuclear radiation

First of all it has been suggested that the energy spectrum of the employed neutron radiation pays an important role in defining its consequences on the properties of the material. In fact radiation damage in concrete aggregates is caused by changes in the lattice structure of the minerals in the aggregate. Fast neutrons with an energy of about 1MeV are generally required to bring about a sufficiently large number of lattice defects in crystalline solids to result in changes in physical and mechanical properties [5].

Figures 1, 2, 3, report the experimental data referred to as results from slow or fast neutrons exposure: filled symbols are used for slow neutrons, empty symbols for fast and half-filled symbols where non separation between fast and slow neutrons was possible.

Though generally expected that fast neutron radiation would lead to a more pronounced radiation damage than slow neutrons, this is not always confirmed on the basis of the data; nevertheless there is evidence that, all other parameters being equal, fast neutron are indeed more detrimental .

Moreover many investigators [9, 10] conclude that different type of aggregates lead to concrete with different resistance against neutron radiation.

In [9] it has been reported that the resistance of a particular type of aggregate against neutron radiation can be related to volume changes of the aggregate and concrete during radiation.

In figure 5 the relation between volume change and neutron fluences for concrete made of limestone aggregate and of flint aggregate is shown:

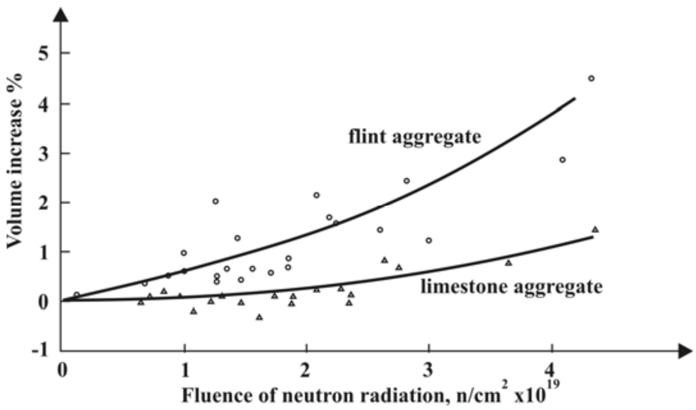

Figure 5: Volume change of concrete specimens exposed to fast neutrons [20, 24].

Concrete made with flint aggregate shows considerably larger volume changes than concrete made with limestone aggregates; moreover there is evidence [5, 7] that quartz aggregates are more affected by radiation than calcareous ones because of the presence of weaker covalent bonding between atoms, instead of ionic ones.

Gray [9] reports that aggregate of the same type with similar chemical composition may show different volume changes because of differences in their microstructure.

With the volume growth tensile strength too vary (on average decaying of the 62 and 47 per cent respectively for flint and limestone aggregates in the range of $2x10^{19}$-$4x10^{19}n/cm^2$); this let us conclude that concrete strength is the lower the larger the volume increase during neutron radiation; however concrete specimens which were only temperature exposed and not irradiated do not show such a volume increase but rather the expected shrinkage.

According to [6] a neutron fluence of less than $1x10^{19}n/cm^2$ does not lead to a volume increase of the irradiated samples: in fact in this range the volume change of irradiated samples is approximately equal to the shrinkage of temperature exposed specimens.

Further there is evidence [7, 11] that nuclear radiation significantly increases the reactivity of silica-rich aggregates to alkali; the decrease of the resistance to nuclear radiation with increasing the content of SiO_2 in aggregates strongly indicates that the deterioration is due to the acceleration of this kind of reaction in concrete and can explain in part the degradation of the mechanical properties of the shielding material.

The mechanism has been explained as follows [11]: OH^- ions of the alkaline solution in the micropores of concrete react with SiO_2 in aggregates to cause the scission of the Si–O bonding and the next expansion of the aggregates by hydration of SiO_2. The consumption of OH^- ions by the scission 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 deformation of the aggregates, the expansion pressure generated by the penetration of the solution is accumulated in the aggregates, thus leading to cracks in the cement paste and expansion of concrete.

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.

3.3 Effect of gamma-radiation on concrete strength

During the tests in which the effect of neutron radiation was studied the concrete samples were also exposed to primary and secondary gamma radiation.

Not many tests have been reported on the effects of gamma radiation alone without the simultaneous exposure to neutron radiation. Alexander [10] reports that for gamma radiation doses of about 10^{10}rad there is no reduction in the compressive strength of concrete when compared with the strength of companion specimens which have neither been irradiated nor heated; there is, however, evidence of reductions of between 25 and 60 per cent in compressive strength for doses exceeding about 10^{11}rad [12].

In this last test the specimens were immersed in demineralised water in order to shield them against neutrons.

Demineralised water may cause deterioration in concrete so that the obtained results must be evaluated with caution; in fact after several years of exposure the surface of the irradiated samples were partially destroyed whereas the non-irradiated samples did not show such damage, thus allowing to conclude that the simultaneous effect of demineralised water and gamma radiation leads to a more pronounced deterioration of concrete than only exposure to demineralised water.

Experimental data [13] on concrete under gamma irradiation also seem to suggest that the interaction with the shielding material leads to lowering both its strength and its porosity. The mechanism is explained to happen with a succession of chemical reactions in the material, starting from the radiolysis of water and ending with the formation of calcite, crystals of which grow into pores, thus resulting in a decreased size of pores, and destroy tobermorite gel by crystallization pressure.

4 CONCLUSIONS

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. A collection of the most interesting experimental results on the topic is presented here.

Changes in the properties of concrete appear to depend primarily on the behavior of concrete aggregates that can undergo a volume change when exposed to radiation. Changes or 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 aggregate (e.g. flint). Quartz aggregates that contain crystals with covalent bonding should be more affected by radiation than calcareous aggregates that contain crystals with ionic bonding. Neutron fluences of the order of $1x10^{19}n/cm^2$ and gamma radiation doses of 10^{10}rad seem to become critical for concrete strength.

The achievements in terms of experimental results on irradiated samples are supposed to lead us to the definition of a multiplicative variable of radiation damage for concrete modeled as a multiphase porous material in order to match the requirement of quantifying the effects of the operative conditions of the SPES Facility for the production of exotic species at Legnaro National Laboratories INFN.

The above information will be introduced in the F.E.M. code NEWCON3D [2, 14-17].

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