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Invited paper

Review of the Current State of Knowledge on the Effects of Radiation on Concrete[1)]

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Abstract

A review of the current state of knowledge on the effects of radiation on concrete in nuclear power production applications is presented. Emphasis is placed on the effects of radiation damage, as reflected by changes in engineering properties of concrete, in the evaluation of the long-term operation and for plant life or aging management of nuclear power plants (NPPs) in Japan, Spain, and the United States. National issues and concerns are described for Japan and the United States followed by a discussion of the fundamental understanding of the effects of radiation on concrete. Specifically, the effects of temperature, moisture content, and irradiation on ordinary Portland cement paste and the role of temperature and neutron energy spectra on radiation-induced volumetric expansion (RIVE) of aggregate-forming minerals are described. This is followed by a discussion of the bounding conditions for extended operation; the significance of accelerated irradiation conditions; the role of temperature and creep; and how these issues are being incorporated into numerical and meso-scale models. From these insights on radiation damage, analyses of these effects on concrete structures are reviewed, and the current status of work in Japan and the United States is described. Also discussed is the recent formation of a new international scientific and technical organization, the International Committee on Irradiated Concrete, to provide a forum for timely information exchanges among organizations pursuing the identification, quantification, and modeling of the effects of radiation on concrete in commercial nuclear applications. The paper concludes with a discussion of research gaps, including (1) interpreting test-reactor data, (2) evaluating service-irradiated concrete for aging management and to inform radiation damage models with the Zorita NPP (Spain) serving as the first comprehensive test case, (3) irradiated-assisted alkali-silica reactions, and (4) RIVE under constrained conditions.

1. Introduction

As light water reactors (LWRs) that became operational in the 1960s and 1970s have aged and operational efficiencies have improved, national regulators, nuclear power plant (NPP) owners, and researchers have focused on enhanced aging management (examination, inspection, maintenance, and testing of key components) and, for some countries, lifetime extension to meet future national energy needs while reducing greenhouse gas emissions. To achieve these goals, a critical evaluation of the knowledge gaps of materials that comprise NPP structures and components must be completed. Moreover, although much of the focus has been on the performance and possible degradation mechanisms of metals due to increased exposure time to temperature, stress, coolant, and radiation fields, other materials such as concrete and cables also are critical to plant life management (PLM) and long-term operation (LTO) (Rosseel *et al*. 2015b). This report focuses on research aimed at reducing the knowledge gap of the effects of radiation on concrete as identified by Graves *et al*. (2014).

Until recently, the data on the mechanical degradation of concrete due to radiation were thought to have been quite limited. Moreover, in 2010, Kontani *et al*. (2010) recognized that reference levels based on data reported in Hilsdorf *et al*. (1978) and used to establish reference

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levels for concrete integrity might not be suitable because data were not representative of concrete mixtures, temperatures, and radiation fields typically seen in LWRs. Furthermore, a comprehensive review and reanalysis of the literature by Field *et al*. (2015) has greatly expanded the database and confirmed the predominant role of radiation-induced volumetric expansion (RIVE) in the degradation of irradiated concrete..

In this report, a review of the current state of knowledge on the effects of radiation on concrete in nuclear applications is presented. Emphasis is placed on the effects of radiation damage as reflected by changes in engineering properties of concrete for PLM and the evaluation of the long-term operation of NPPs in Japan, Spain, and the United States.

Following a discussion of issues and concerns in Japan and the United States, this report focuses on the current status of the fundamental understanding of radiation effects on concrete. It begins with the effects of radiation on hardened cement paste (HCP), including drying and temperature effects and radiolysis, and RIVE of aggregate-forming minerals. This is followed by a discussion of the combined effects of temperature, drying, and radiation on concrete, including the significance of accelerated irradiation conditions, the role of temperature and creep, as well as variations in energy spectra and damage energy, and how these factors are being incorporated into numerical and mesoscale models of concrete structures.

From these insights on radiation damage and the bounding fluences at extended lifetimes, analyses of these effects on concrete structures are reviewed, and the current status of work in Japan and the United States is described. Furthermore, the recent formation of a new international scientific and technical organization [the International Committee on Irradiated Concrete (ICIC)] to provide a forum for timely information exchanges among organizations pursuing the identification, quantification, and modeling of the effects of radiation on concrete in commercial nuclear applications is described.

The paper concludes with a discussion of current research gaps, including (1) interpreting test-reactor data, (2) evaluating service-irradiated concrete for aging management and to inform radiation damage models with the Zorita NPP (Spain) serving as the first comprehensive test case, (3) irradiated-assisted alkali-silica reactions, and (4) RIVE under constrained conditions.

Japanese conditions, issues and concerns:

LWRs have been identified as an important component of electric power generation as established by extensive discussions following the Fukushima Daiichi nuclear accident (METI 2014). Ensuring the safe, long-term operation of NPPs is necessary for the public and for electric power suppliers, particularly because construction of new NPPs is difficult in Japan. For these reasons, aging management of concrete structures that cannot be replaced is crucial for long-term operation of NPPs.

Before the Fukushima Daiichi accident, the Japanese government's PLM program inspected NPPs to evaluate their integrity after an operation period of 30 years and every 10 years thereafter. This process required data on the integrity of structures under present and future conditions (Japan Nuclear Energy Safety Organization 2013; Architectural Institute Japan 2015). To evaluate concrete structures in PLM, two measures of deterioration were chosen: reduction in concrete strength and reduction in shielding performance. The following factors arising from the environmental conditions and materials used in reactor buildings adversely affect the concrete strength: elevated temperature, irradiation, carbonation, the alkali-silica reaction (ASR), and machine vibration.

After the Fukushima Daiichi nuclear accident, new nuclear reactor regulations came into effect on 8 July 2013, and an approval system for extending the operation period of NPPs was introduced by the Nuclear Regulatory Authority of Japan (NRA). Generally, a 40-year operation period is allowed for every NPP although this period can only be extended to a maximum of 20 years after the facility passes a special inspection.

In PLM and the inspection for extending the operation period, the integrity of concrete affected by radiation is evaluated with reference levels taken from data reported in Hilsdorf *et al*. (1978) and interpreted by Kontani *et al*. (2010): 1×10^{20} n/cm^2 for fast neutrons and 2×10^5 kGy for gamma rays. However, in a national project organized by the Japan Nuclear Energy Safety Organization (partly a former organization in the NRA), the Hilsdorf compiled data were found to be unsuitable for this purpose because they did not cover the type of concrete used in LWRs (Kontani *et al*. 2010). A national project has been launched to elucidate the effects of radiation on concrete properties and to develop a system for evaluating the integrity of concrete affected by radiation (Maruyama *et al*. 2012).

US conditions, issues, and concerns:

As described in Rosseel *et al*. (2014), extending the operating lifetimes of current NPPs beyond 60 years and making additional improvements in their productivity is essential to meeting future United States national energy needs while reducing greenhouse gas emissions. To meet these goals, a critical evaluation of the knowledge gaps of the effects of exposure time to temperature, stress, coolant, and radiation fields on materials that comprise the structures and components of an NPP is required. And although it is expected that the vast majority of concrete structures will continue to meet their functional and performance requirements during future licensing periods, there may be isolated examples where structures may not exhibit the desired durability, primarily due to environmental effects such as radiation.

To address these extended lifetimes effects, the Electric Power Research Institute (EPRI), through the Long-

Term Operations (LTO) Program, and Oak Ridge National Laboratory (ORNL), through the US Department of Energy (DOE) Light Water Reactor Sustainability (LWRS) Program, established a research plan to investigate the aging and degradation processes associated with NPP concrete. The basis for the plan arose in part from the Expanded Materials Degradation Analysis (EMDA) report on the aging of concrete, an effort jointly supported by DOE and the US Nuclear Regulatory Commission (NRC), and performed by expert panels from US national laboratories, industry, academia, and international organizations. A key finding of the EMDA was the urgent need to develop a consistent knowledge base on irradiation effects in concrete (Graves *et al*. 2014). Although not part of the EPRI/LWRS plan, the NRC has initiated confirmatory research for the developing the technical basis for the regulatory framework of subsequent license renewal (see for example Section 6.2).

The LWRS program is focusing on (1) assessing the radiation environment in concrete biological shields (CBSs), defining the upper bound of the neutron and gamma dose levels expected at extended operation, and estimating adsorbed dose; (2) harvesting and testing service-irradiated concrete to validate models; (3) irradiating prototypical concrete and its components under accelerated ion, neutron and gamma dose levels to establish upper bounds to inform radiation damage models; (4) developing improved models to enhance the understanding of the effects of radiation on concrete; and (5) establishing an international collaborative research and information exchange to leverage capabilities and knowledge including developing cooperative test programs to improve confidence in data obtained from various concretes and from accelerated irradiation experiments (Rosseel *et al*. 2015a). EPRI is now focusing on the structural analyses of select US NPPs with CBS that perform structural support and whose estimated fluence at ~ 80 years of operation approaches the bounding fluence of 6×10^{19} n/cm^2 at E >0.1 MeV (Section 4.1).

2. Fundamental understanding of radiation effects on concrete

2.1 Introduction

Concrete is by nature and constitution, a composite material made of HCP and aggregate. Water is present in the HCP under various forms (i.e., chemically bound, adsorbed and free water). Gamma and neutron irradiation interact differently with the concrete compounds [See **Table 3** in Kontani *et al*. (2010)]. In particular, the poorly crystallized structure of the calcium-silicate-hydrates (C-S-H) and the HCP porosity makes them less susceptible to neutron-induced lattice defect accumulation and distortion(Kontani *et al*. 2010) as opposed to well-crystallized aggregate-forming minerals. Concurrently, gamma-induced radiolysis occurs predominantly where water is present (i.e., in the HCP). Because irradiation results in energy-deposition-induced heating, temperature and moisture transport must be considered concurrently in the study of irradiation effects. The following sections provide a summary of the state of knowledge of the effects of temperature, moisture content and irradiation on ordinary Portland cement paste and aggregate.

2.2. Hardened cement hydrates

2.2.1 Drying and temperature effects:

The shrinkage and strength properties of HCP affect the physical properties of concrete. The aggregates restrain the shrinkage of cement paste during drying or heating (Carlson 1938; Hansen and Nielsen 1965; Hobbs 1974), and the large difference in volume change, due to environmental conditions, between the aggregates and the cement paste matrix produces cracks in the cement paste that act as pores. These cracks affect the strength and Young's modulus of the concrete (Bažant *et al*. 1982; Son and Hosoda 2010; Maruyama *et al*. 2014b; Lin *et al*. 2015). Therefore, the differential shrinkage is the main factor in the monotonic reduction of the Young's modulus of the concrete (Maruyama *et al*. 2014b).

The strength of the mortar and concrete decreases from high relative humidity (RH) to intermediate RH (40-50% RH at room temperature) and increases below the intermediate RH range under drying or heating conditions (Pihlajavaara 1974; Maruyama *et al*. 2014b). This behavior is attributed to the change in strength of the HCP, and the cracks due to drying are important in reducing the compressive strength above 60% RH through limiting the load bearing path in concrete (Maruyama *et al*. 2014b). This complex behavior of strength of HCP is explained by the colloidal nature of C-S-H (Jesser 1927; Powers and Brownyard 1946; Tomes *et al*. 1957; Thomas and Jennings 2006; Jennings 2008). During the initial drying process, the C-S-H particles forming colloidal C-S-H agglomerate, producing large pores in the cement paste. The increase in the volume of large pores reduces the paste strength. In contrast, drying strengthens the C-S-H by tightening the layered structure. The increase in the strength of the solid structure overcomes the reduction of strength caused by large pores, resulting in the strength increase of hardened cement paste at lower RHs (Maruyama *et al*. 2014a).

2.2.2 Radiolysis effects:

Gamma rays affect material through electronic excitations. Metastable calcium peroxide octahydrates can be produced in HCP with high water content (Bouniol and Aspart 1998; Vodák *et al*. 2005; Lowinska-Kluge and Piszora 2008), altering the strength and pore size distribution and, especially, resulting in subsequent carbonation. It is also suggested that gamma radiation-induced carbonation increases the strength of HCP (Vodák *et al*. 2005; Vodák *et al*. 2011). Underlying these phenomena is the radiolysis of water. Radiolysis is the physicochemical consequence of ionizations along the trajectory of radiation. Several groups, including Bouniol and Aspart (1998), Bouniol and Bjergbakke (2008), and Kontani *et al*. (2013), have studied the radiolysis of pore water in cement.

For example, Kontani *et al*. (2013) performed gamma-ray irradiation experiments on cement paste and collected all the gases from the samples by argon career gas. By analyzing time-dependent gas composition changes during gamma irradiation, they found the following phenomena of gas production in cement paste:

- The hydrogen production G value[2)] is linear with gamma-ray dose rate (1~10 kGy/h), and this relation is not affected by the sample temperature (20~60°C).
- The linearity between gamma-ray dose rate and hydrogen production G value implies a validation of accelerated gamma-ray irradiation experiments with gamma-ray dose rates less than10 KGy/h.
- Only a few percent of the chemically bound water decomposed even when subjected to the reference level of 2×10^5 kGy, since the G values were very low at around 0.03[3)]. This means that hydration products can maintain their integrity.
- The hydrogen generation rate decreased as the evaporable water content in the sample decreased.

Further scientific research is necessary to validate the integrity of concrete and find a possible degradation mechanism by radiolysis in concrete.

2.2.3 Interaction between creep and irradiation:

Creep of the cement paste is a key aspect of the mechanical behavior of concrete, especially considering the time scales involved in the LTO of NPPs. On the one hand, creep may affect the response of the structure itself, for example, by relaxing the pre-stress in rebars (e.g., Magura *et al*. 1963), and on the other hand, it strongly affects crack propagation in concrete (Rusch 1960; Bazant and Gettu 1992). Gray (1971) measured an increase in the creep and shrinkage of a cement grout specimen under neutron irradiation even though the various changes of hygro-thermal conditions during the experiments, and the lack of control specimen, make this specific experiment difficult to interpret. McDowall (1971) found that under gamma irradiation, creep of concrete was reduced, while autogeneous shrinkage was increased. This is consistent with creep experiments at low relative humidity (Whittmann 1970), and therefore might be a manifestation of the radiolysis in the cement paste. Understanding how creep is affected by radiation is critical because creep controls the long-term crack propagation by relaxation of the stresses in the cement paste [e.g., Altoubat and Lange (2001) for shrinkage cracking, Giorla, Scrivener *et al*. (2015) for ASR].

2.3. Aggregate-forming minerals

2.3.1 Radiation-induced volumetric expansion:

With about 70% of the volume fraction of concrete consisting of aggregate, understanding aggregate-forming mineral sensitivity to radiation is of the utmost importance. Alpha decay of certain radionuclides is known to produce a metamict state in rock-forming minerals (Ewing *et al*. 1988). Ion-beam irradiation of minerals generally results in crystalline-to-amorphization (CA) transition (Wang *et al*. 1991; Eby 1992; Douillard and Duraud 1996; Harbsmeier and Boise 1998) and, in some cases, in the precipitation of oxides (Meldrum *et al*. 1997) or the formation of cavities (Templier *et al*. 1997). Amorphization is predominantly observed in silicate-based minerals. For example, while quartz presents strong long-order radiation-induced disordering, irradiation of calcite causes primarily the rotation of the carbonate groups (Pignatelli *et al*. 2016). The critical CA dose in silicates depends on several factors: (1) the melting temperature; (2) a structural factor (e.g., degree of SiO_4 polymerization, elastic properties); and (3) the proportion of Si-O bonding (Eby *et al*. 1992). Amorphization results in changes in physical, optical and mechanical properties (e.g., Wittels 1957; Primak 1958; Mayer and Lecomte 1960; Wong 1974). Initial anisotropic crystalline elastic properties are gradually modified with the irradiation dose to converge toward an isotropic stiffness tensor (Mayer and Lecomte 1960; Zubov and Ivanov 1967). Amorphization also induces significant density change, especially in silicates. For example, the maximum volumetric expansion of quartz and feldspars has been shown to be as large as ~18% (Primak 1958) and ~8% (Krivokoneva 1976), respectively, while the change of density in calcite remains rather low (~0.3% according to Wong). Depending on the mineralogical content, considerable variations in aggregate RIVE have been observed as described in the comprehensive review by Field *et al*. (2015). Moreover, some observed post-irradiation expansions exceed what is considered as detrimental by ASR researchers (e.g., Fournier and Berube 2000; Rajabipour *et al*. 2015).

2.3.2 Role of temperature in radiation damage:

Increased irradiation temperature delays the onset of the critical dose for amorphization and reduces the expansion kinetics of quartz (Bykov *et al*. 1981) and silicates such as feldspars (Krivokoneva 1976) and pyroxenes, yet it does not seem to affect the total RIVE at the end of the amorphization process. The effect of temperature on quartz expansion is explained by the annealing of point defects during irradiation occurring at an early and intermediate stage of amorphization. However, no significant annealing or anti-annealing (Primak 1958, Yano *et al*. 2007) of RIVE is expected from temperature

[2)]The G value refers to the number of specified chemical events produced in an irradiated substance per 100 eV of energy absorbed.

[3)]This value is estimated on an assumption that only hydroxyl groups in the cement paste sample absorb the gamma-ray energy.

variations occurring in the test reactors and LWRs (Le Pape *et al*. 2016).

2.3.3. Energy spectra and damage energies:

As previously noted, a review of existing data on irradiated concrete reveals that neutron fluence is predominately used to characterize the radiation environment (Hilsdorf *et al*. 1978; Field *et al*. 2015), but there is little consistency in the neutron energy cutoff applied for the neutron fluence (Remec *et al*. 2016b). However, in the CBS of pressurized water reactors (PWRs), the neutron fluence for a cutoff energy of E >0.1 MeV is about 8-15 times higher than the neutron fluence at E >1 MeV (and the neutron fluence for E >0 eV can be 20-40 times higher) (Field *et al*. 2015). Using consistent and appropriate energy cutoff is therefore crucial for the assessment of concrete degradation.

RIVE is currently considered the dominant cause for degradation of irradiated concrete (Field *et al*. 2015). It is, therefore, of interest to investigate neutron-induced atom displacements in aggregates. Remec *et al*. (2016b) showed that, for the neutron spectra in the biological shield of two-loop and three-loop (the number of reactor cooling circuits) PWRs, neutrons with energies above 0.1 MeV contribute more than 95% of all atom displacements in several widespread aggregate minerals, while neutrons with energies above 1 MeV cause only about 20 to 25% of the total atom displacements, as shown in **Fig. 1** (Remec *et al*. 2016a). The conclusions were similar when, instead of atom displacements, neutron-induced absorbed dose in minerals was considered. These observations suggest that neutron fluence with a cutoff of 0.1 MeV is the preferred parameter for radiation field characterization in irradiation experiments and for accessing radiation-induced concrete degradation**.**

Work is in progress to develop a unified irradiation parameter that will account for neutron and gamma-ray contributions to concrete degradation. In the typical radiation field of the PWR biological shield, the contribution of gamma rays to the total absorbed dose is similar and may exceed the neutron-induced absorbed dose (Remec *et al*. 2013). However, preliminary results indicate that the gamma-ray contribution to the atom displacements does not exceed a few percent of the neutron-induced displacements and will therefore be insignificant in most cases. In accelerated irradiation experiments, the conditions may be considerably different, and contributions of gamma rays need to be carefully evaluated for each specific experiment.

3. Irradiated concrete

3.1 Test reactor data

The essential reference used to systematize the analysis on irradiated concrete structural properties is Hilsdorf *et al*. (1978). Later, Fujiwara *et al*. (2009) limited their analysis to ordinary Portland structural concrete and found no data subject to any significant effect of neutron irradiation on concrete for fluence less than 1 × 10^{19} n/cm^2. More recently, Field *et al*. (2015) revisited historical test reactor irradiation data and collected 307 compression strength data, 62 tensile strength data, 138 elastic modulus data and 114 linear expansion data. The data suggest that concrete compressive and tensile strengths as well as the Young's modulus decrease above fluences around 1 × 10^{19} n/cm^2. This *threshold fluence* value, however, is subject to variations with the concrete composition and, possibly, the exposure conditions. A strong correlation with the radiation-induced (and thermal expansion) volumetric expansion of aggregate and concrete is observed, suggesting that RIVE is a primary degradation factor (Seeberger and Hilsdorf 1982). However, Field *et al*. (2015) noted important uncertainties limiting the interpretation of the gathered data, particularly for the fluence levels of interest (i.e., above 1 × 10^{19} n/cm^2): (1) The absence of energy cutoff normalization (as described in Section 2.3.3); (2) the absence of comparable mechanical testing procedures

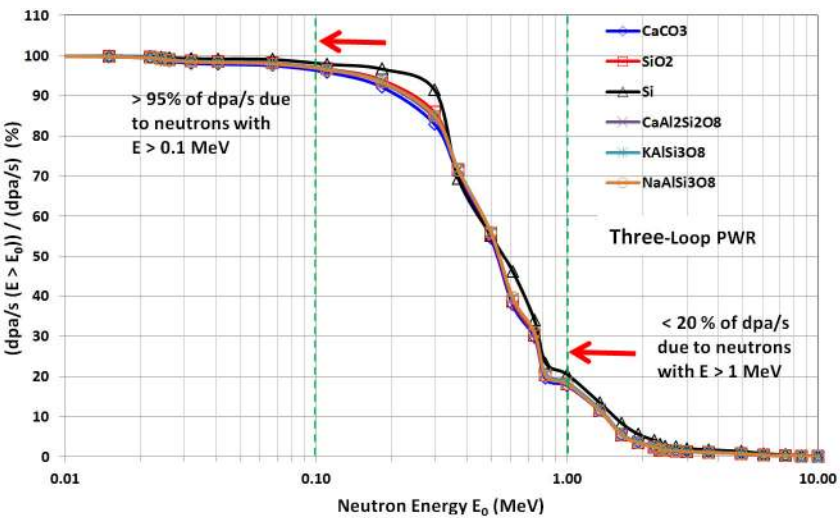


Fig. 1 Relative contribution of neutrons with energies E > E_0 to the dpa (displacements per atom) rate for the neutron spectrum in the cavity of a three-loop PWR, for several selected minerals.

(age, geometry and size of specimens); (3) the effects of conditioning (moisture content) before and during irradiation are often not reported; and (4) most data correspond to testing temperatures above 100°C leading to enhanced damage in the HPC and at the interface with aggregate. Finally, although reliable data on the effects of gamma irradiation on concrete mechanical properties are scarce (Alexander 1963; Kelly *et al.* 1969; Gray 1971), they seem to suggest limited impact. Further research is needed.

A correct interpretation of irradiation experiments requires a physics-based mechanistic understanding and modeling of the temperature, moisture content and irradiation effects in the concrete compounds. The next section provides some insights regarding the recent progress made using mesoscale models of irradiated concrete.

3.2 Numerical models

Mesoscale model:

The impact of the expansion of aggregates due to radiation and the resultant damage observed as changes in physical properties can be understood through numerical calculations. This approach has two objectives. The first is to find a mechanism that describes changes in physical properties of concrete components due to heat and drying, as well as radiation. This mechanism will contribute to the interpretation of experimental data and to formulating a universal conclusion. The second is to extrapolate the phenomena from a laboratory specimen size to a structural size that cannot be obtained experimentally.

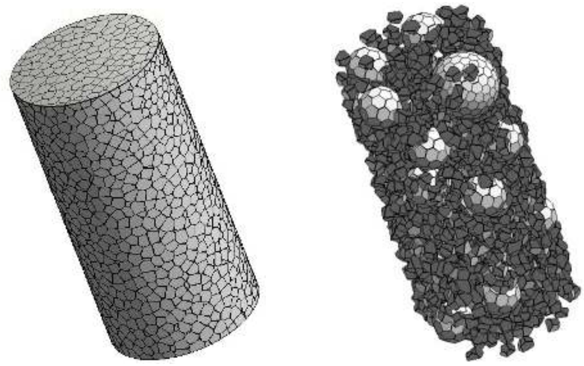
In the mesoscale, or aggregate-size scale, it is important to consider the role of aggregate explicitly in a numerical calculation. For this objective, the finite-element method (FEM) and rigid body spring model (RBSM) are applied to the evaluation of concrete under drying, heating and radiation.

Originally developed by Kawai (1978), RBSM has been applied extensively for structural analysis. RBSM deals with crack propagation of concrete directly since it represents a continuum material as an assembly of rigid particle elements interconnected by zero-size springs along their boundaries. Being nonlinear, these zero-size springs can simulate the cracking behavior of a continuum material. The nonlinearity and discrete behavior of the continuum material is emulated by cracks developing at the interfaces of the rigid particles. For this reason, crack patterns and the resultant nonlinear behavior of the target model are significantly affected when a mesh design is employed. To solve this problem, random geometry using Voronoi diagrams was applied (Bolander and Saito 1998).

Recently, RBSM has been applied to simulate volume change and compressive strength of concrete concurrently (Ogawa and Maruyama 2016). Coarse aggregate is explicitly modeled in the mesh (**Fig. 2**), and the interfacial transition zone (ITZ) role is considered according to the previous numerical calculation results of concrete shrinkage (Maruyama and Sugie 2014). Based on an input of mortar shrinkage, damage in concrete produced by the restraint of coarse aggregate was evaluated explicitly. Owing to the modeling of ITZ, shrinkage of concrete under different drying conditions can be reproduced by the proposed model. After drying, specimens were loaded and strength changes calculated. The results are shown as the change in the relative compressive strength (F_c/F_{co}) in **Fig. 3**. From the calculation, it is elucidated that compressive strength of cylinder concrete is determined by both damage in concrete due to the volume change difference between mortar and coarse aggregate and strength change of cement paste (in this calculation, it is strength mortar) due to colloidal alteration under heating and drying.

A preliminary simulation to evaluate the impact of aggregate expansion due to radiation was conducted. In the preliminary simulation, expansion of coarse aggregate in addition to the presented model discussed above cannot simulate the expansion of concrete and strength reduction due to damage in concrete. In the case of aggregate expansion, impact of damage in concrete is overestimated, and further study is required of the relationship of damage in concrete in large expansion to the physical properties of concrete.

A 2D mesoscale model for irradiated concrete (Giorla, Vaitová *et al.* 2015) was developed at ORNL based on



(a) Voronoi mesh (b) Aggregate mesh

Fig. 2. RBSM mesh (Ogawa and Maruyama 2016).

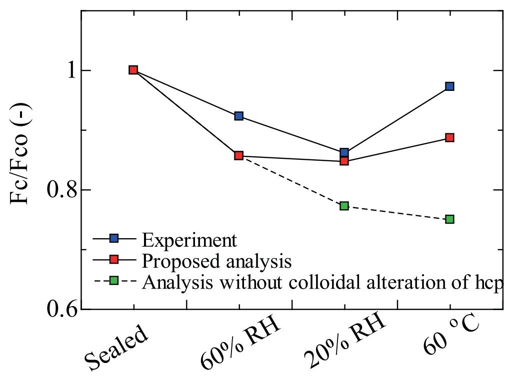


Fig. 3. Comparison of calculated F_c/F_{co} with experimental data (Ogawa and Maruyama 2016).

the AMIE finite element framework (Dunant and Scrivener 2010, Giorla *et al*. 2014). The model aim is to provide the concrete expansion and internal degradation as a function of the aggregate RIVE and the mechanical properties of the paste and the aggregates, notably accounting for the coupling between creep and damage in the cement paste (Giorla, Le Pape and Dunant 2016). The model was validated in an irradiation experiment with serpentine concrete (Elleuch *et al*. 1972) since it is one of the very few experiments in the literature in which the properties of the concrete, the aggregates, and the cement paste were independently characterized (see **Fig. 4**). Under variable temperature and flux, the model predicts a higher damage and expansion than under a constant temperature and flux (Le Pape *et al*. 2016). The model also shows an increase and orientation of internal cracking under external restraint, which is similar to the effect observed for ASR (Giorla, Le Pape, and Dunant 2016).

Even though these models (RBSM and FEM) are based on different representations of the material (notably in terms of material continuity and mechanical behavior), they both greatly overestimate the damage in the cement paste. This effect might be caused by the fact that aggregates are represented as homogeneous particles that swell uniformly, and could therefore be reduced with a more accurate description of the aggregate mineral microstructure. Accounting for different RIVE within the aggregate might cause damage to initiate within the aggregates rather than at their surface, as it is for example the case in ASR (Ben Haha *et al* 2007; Dunant and Scrivener 2010). Doing so requires an experimental investigation and characterization of the aggregates themselves, as well as a well-informed database of minerals and their properties.

4. Significance of irradiation for concrete structures in NPP

4.1 Bounding fluence at extended lifetimes and radiation transport in concrete

As previously discussed, (Remec *et al*. 2016b; Rosseel *et al*. 2014; Rosseel *et al*. 2015b), the onset of radiation-induced concrete degradation has been correlated to neutron fluence and gamma ray dose based on data assembled by Hilsdorf *et al*. (1978). Obtaining accurate estimates of the bounding fluence and dose values for the US NPP fleet at the projected extended service is clearly important. Due to variations in plant operations and differences in reactor designs, however, this effort is not simple. Fortunately, because operating NPPs are required to implement a reactor pressure vessel (RPV) surveillance program, a wealth of fluence information is available. In the United States, the information is available from the US NRC Agency-wide Documents Access and Management System (ADAMS) database (US NRC 2015).

The critical first step is the neutron and gamma ray transport calculations of the attenuation through the pressure vessel and CBS (Remec *et al*. 2013). An example of the attenuation for a Japanese PWR is shown in **Fig. 5**.

Using the transport calculations as guidance, a multistep process was implemented by Esselman and Bruck (2013) to determine the expected neutron fluence values on the CBS for the current US boiling water reactor (BWR) and PWR fleet for up to 80 years of operation assuming 92% operating efficiency. The results shown in **Fig. 6** highlight the importance of plant design and fuel loading scheme in determining the maximum expected fluence on the surface of the CBS at 80 years of operation. For example, 2- and 3-loop PWR reactors show significantly higher fluence values than the 4-loop plants. It should be noted that the BWR fluence at 80 years of operation is generally lower than 4-loop PWR fluence.

4.2 Structural analysis

Summary of concrete structural analysis literature:

The primary function of the CBS is to provide radiological protection from the radiation emitted from the nuclear steam supply system (NSSS), in particular, the RPV. In some PWR designs, the structural function of the CBS is to transfer the static or seismic load of the reactor to the foundation system and to mitigate the effects of accident scenarios. The CBS is generally made of conventional [i.e., normal weight aggregate (Esselman and Bruck 2013)] carbon steel reinforced concrete (Hookham 1991). Some designs also include a steel plate liner**.**

Limited irradiated concrete structural analyses have been published in the open literature using different models: analytical axisymmetric models (Andreev and Kapliy 2014; Le Pape 2015), finite-element membrane models (Mirhosseini *et al*. 2014), finite-element contin-

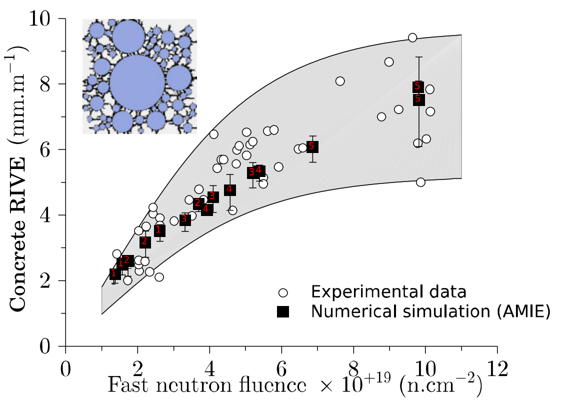


Fig. 4 Comparison of calculated concrete RIVE with experimental data from Elleuch *et al* (1972), and example of simulated damage pattern at a neutron fluence of 1.3×10^{19} n/cm^2). The gray area represents the scatter in the experimental data. Figure adapted from Giorla, Vaitova, *et al* (2015).

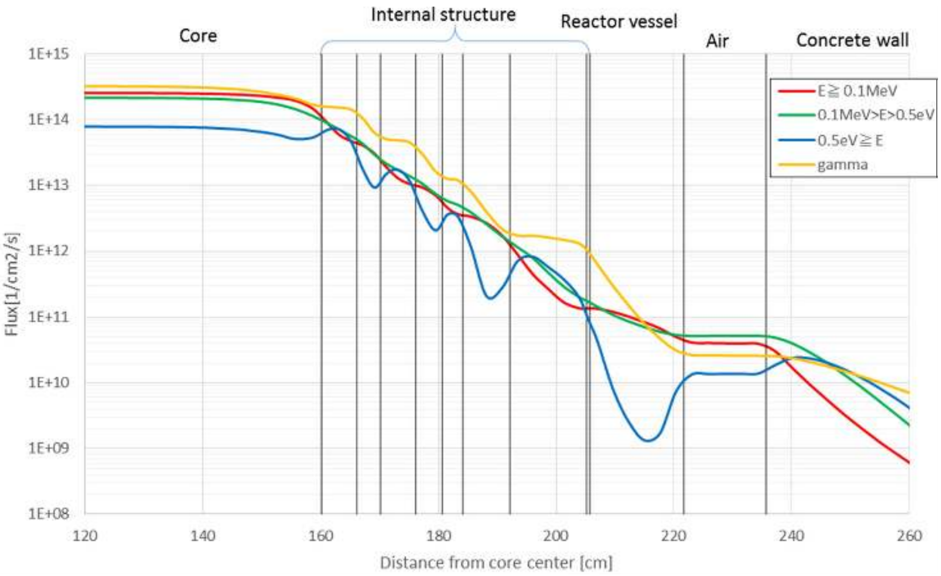


Fig. 5 Example of Japanese PWR radiation field developed from neutron transport calculations (Fukuya et al. 2002). Dr. O. Sato (Mitsubishi Research Institute Inc.) and Dr. T. Igari (MRI Research Associates Inc.) confirmed this calculation.

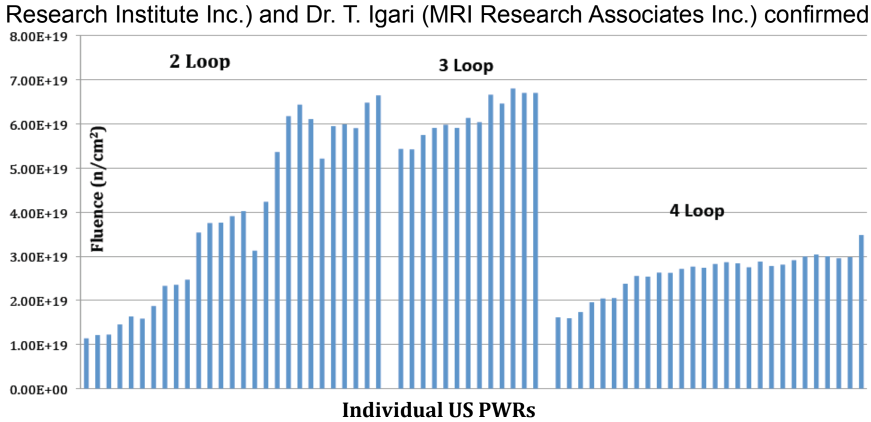


Fig. 6 Summary of US PWR fleet fluence at the outer diameter of the RPV extrapolated to 80 years (E >0.1 MeV) and grouped by design (heat transfer loops). Graph reinterpreted from Esselman et al. (2013).

uum models (no RIVE) (Pomaro *et al*. 2011), and finite-element mesoscale models (Salomoni *et al*. 2014; Giorla, Le Pape, and Huang 2016).

In the absence of RIVE (or swelling), radiation damage [i.e., induced loss of mechanical properties (Hilsdorf *et al*. 1978; Field *et al*. 2015)] has very limited effects on the CBS (Pomaro *et al*. 2011). Moreover, the radiation field's strong attenuation produces a high RIVE gradient (Le Pape 2015) causing high biaxial compressive elastic stresses in the vertical and hoop directions near the reactor cavity and important tensile hoop stresses toward the back of the CBS (Andreev and Kapliy 2014; Le Pape 2015). Simultaneously, the prolonged moderate temperature exposure (<65°C by design) and strong internal moisture content gradient (Oxfall 2013) affect the degree of hydration of concrete, and thus, its mechanical properties. In particular, it leads to the development of lower strengths toward the reactor cavity (Maruyama and Igarashi 2015). The dissipation of the RIVE-induced elastic stresses implies a potential relaxation in the cement paste, but, more significantly, the development of cracking (Giorla, Le Pape, and Dunant 2016). The extent of radiation-induced concrete damage appears to be limited to a depth of about <20 cm (Le Pape 2015; Giorla, Le Pape, and Huang 2016). The consequences of this damage to the structural performance under seismic conditions (e.g., impact on the RPV supports, or accident conditions; e.g., sudden increase of temperature in the cavity, or seismic events) still remain to be investigated.

Ongoing structural analysis work in the United States:

Structural analyses currently being performed by US industry are motivated by recent research and development gap identification and prioritization reviews for LWR concrete structures (Graves *et al*. 2014; Wall *et al*. 2012) and, more recently, the US NRC Draft Standard Review Plan for Review of Subsequent License Renewal Applications for Nuclear Power Plants (NRC 2015). The US industry approach, as described in Section 4.1, has been to first determine the predicted neutron fluence in the concrete biological shields for each NPP in the US fleet for neutrons of E >0.1 MeV extrapolated to 80 calendar years of operation (Esselman and Bruck 2013). This was done using reactor-specific surveillance data reported to the NRC, information that is publically available in the NRC ADAMS database (US NRC 2015). The bounding (maximum) neutron fluence was found to be ~6 × 10^{19} n/cm^2, which is higher than the threshold for damage in concrete, as described previously. To determine the effects of chronic neutron irradiation on the structural margin in biological shields that also perform a structural support function, a 3D wedge FEM is being developed. This model is based on the Westinghouse Type 1 PWR vessel support configuration (Lapay *et al*. 2000). A schematic of a typical PWR reactor vessel and biological shield is shown in **Fig. 7** for reference. The finite element mesh was developed from a set of design drawings obtained from a recently shut down 2-loop PWR. In this model, the biological shield in the vicinity of a hot or cold leg feed-through, under which the reactor vessel support sits, and the embedded steel rebar geometry are meshed separately and merged. The biological shield concrete elements were segregated into annular layers to allow variation of mechanical properties and variations in RIVE as a function of depth into the concrete (**Fig. 8**). The model will be run to failure under live and seismic loading conditions to estimate the loss of structural margin and changes in the internal stress morphology due to chronic in-service neutron irradiation.

Ongoing structural analysis work in Japan:

Attempts to evaluate the impact of radiation-induced degradation by numerical simulation are being made in Japan. As previously discussed in Section 2.3.3, there is a distribution of fluence in a real-scale member under irradiation conditions because neutrons are always attenuated by interaction with concrete. Consequently, the radiation impacts show a distribution corresponding to the fluence variation. To evaluate the time-dependent spatial distribution of concrete properly, DEVICE (Damage Evaluation for Irradiated Concrete) a numerical code, which takes into account the heat, moisture, and radiation transport coupled with cement hydration, was proposed by Maruyama *et al*. (2016). This code is composed of the established computational cement-based material model (CCBM) (Maruyama *et al*. 2015) and the one-dimensional deterministic transport SN code ANISN (Engle 1967). DEVICE predicts the property of concrete and creates the input for further structural analysis. The rigid-body spring network, which was originally proposed by Kawai (1978), was applied for the structural analysis, and the impact of expansion of the aggregate and the resultant change of seismic performance was investigated. The preliminary result, which is shown in **Fig. 9**, indicates that there is a risk of

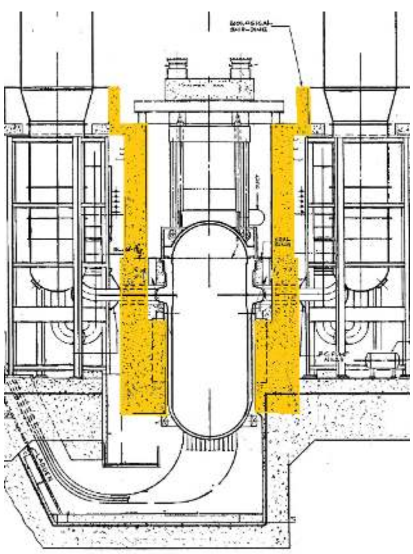


Fig. 7 A typical PWR biological shield configuration (shown in yellow) with the reactor vessel in the center.

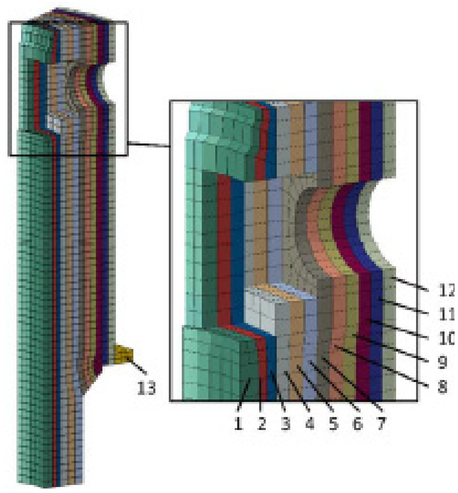


Fig. 8 A finite element mesh of the biological shield in the vicinity of a reactor vessel structural support showing element layers for varying concrete properties and swelling. The numbers represent annular layers of elements used to vary properties and swelling as a function of radial depth into the biological shield. For reference, the total radial thickness of the shield is approximately 6 ft (1.8 m).

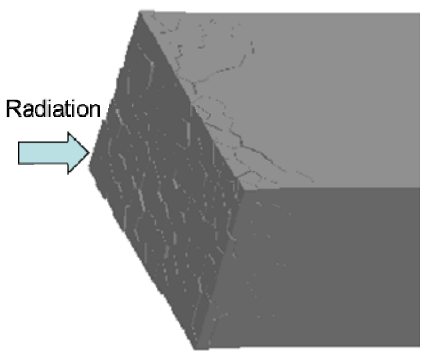


Fig. 9 The expected cracks in concrete under irradiation conditions. The fast neutron value (>0.1 MeV) at the surface of the member is 6.0 × 10^{19} n/cm^2.

Table 1 Exposure conditions in irradiation testing facilities and in-service PWRs. [(a)] (Remec *et al.* 2013), [(b)] (Maruyama *et al.* 2013), [(c)] (Field *et al.* 2015), [(d)] (Gray 1971), [(e)] (Elleuch *et al.* 1972), [(f)] (Dubrovskii 1967), [(g)] (Kontani *et al.* 2010), [(h)] (Seeberger and Hilsdorf 1982), [(i)] (McDowall 1971), [(j)] (Vodák *et al.* 2011), [(k)] (Łowińska-Kluge and Piszora 2008), [(l)] (Kelly *et al.* 1969), [(m)] (Kitsutaka and Matsuzawa 2010), [(n)] (Oxfall 2013).

	PWR	Test reactor/Gamma facility
Fast neutron flux (n/cm^2/s)	1 to 2 × 10^{10} (ID) [(a)]	5 × 10^{11} to 2 × 10^{14} [(b)]
Fast neutron fluence (n/cm^2)	<6 × 10^{19} (ID) at 80 yr [(c)]	<10^{20} [(d,e,f)]
Gamma flux (kGy/h)	5 to 20 [(a,g)]	0.02 to 200 [(h,i,j,k,l,m)]
Gamma dose (MGy)	50 to 200 at 80 yr	0.04 to 1.3
Temperature (°C)	<65 (design)	40 to >250 [(d,e)]
Relative Humidity (-)	Strong gradient ~0.5 at 50 mm, ~0.7 at 150 mm, >0.9 at 400 mm at 30 yr [(n)]	

spalling due to radiation-induced expansion of the aggregate.

5. International forum for collaborations: The International Committee on Irradiated Concrete (ICIC)

Understanding the effects of radiation on concrete is important for PLM and in determining long-term or extended operating performance of concrete structures in existing NPPs. Not surprisingly, this issue is being addressed by research organizations and utilities across the globe. In the last three years, researchers have been actively working to build international partnerships and collaborations in an effort to better define the issues, develop a sound approach to resolving the major questions, and maximize resources. The result is the creation of the ICIC (Rosseel *et al.* 2015b).

The purpose of the ICIC is four-fold. First, it provides a forum for timely information exchange among organizations pursuing the identification, quantification, and modeling of the effects of radiation on concrete in commercial nuclear applications (specifically, determination of the effects of radiation damage as reflected by changes in engineering properties). Second, it promotes the broad application of highly specialized or unique investigation techniques to relevant research materials of wide interest to develop improved understanding and predictions of the effects of radiation on concrete, cement composites, and constituents. Third, it facilitates the conception, planning, and guidance of new cooperative research programs, for example, facilitating the collection (harvesting) and distribution of pedigreed materials suitable for concrete irradiation damage investigations. And fourth, it promotes cross-institutional use of resources, where possible, that reflects mutual research interests or needs. Additional details may be found at the ICIC web page: http://web.ornl.gov/sci/psd/mst/ICICFGM/index.shtml.

6. Discussion of research gaps

6.1 Interpretation of test reactor data

Without an examination of service-irradiated LWR concrete, the current understanding of the effects of radiation on concrete is solely based on test reactor data (i.e., radiation under accelerated conditions). **Table 1** provides a detailed comparison of the differences in radiation flux and dose (E >0.1 MeV), temperature, and relative humidity. Accelerated testing raises questions of possible rate-effect mechanisms and, consequently, the significance of test reactor data. Interestingly, it is not clear that accelerated testing leads to higher damage for a given dose. For example, higher temperatures may increase the damage in the HCP and at its interface with an aggregate, while the RIVE rate is reduced, thereby potentially reducing the damage in the HCP.

Another example is the effect of low/fast-RIVE on the stress relaxation in polycrystalline minerals, aggregate and HCP, which has received little attention so far. In particular, the effects of gamma and neutron irradiation on the HCP creep properties are not clearly understood (McDowall 1971; Gray 1971).

Thorough fundamental understanding of both separated and combined effects of irradiation, temperature and internal moisture on aggregate and HCP is still needed and should be incorporated into physics-based modeling to derive irradiation consequences on LWRs.

6.2 Evaluating service irradiated concrete for PLM and to inform/Confirm radiation damage models: Harvesting zorita concrete

Harvesting of concrete cores from decommissioned NPPs may provide not only an opportunity to generate data from concrete that has experienced typical radiation fields, but it may also provide guidance to accelerated irradiation studies. Moreover, the coupling of accelerated or laboratory-irradiated concrete with harvested cores from NPP is expected to facilitate the effort to develop an understanding of the damage mechanisms in irradiated concrete, including understanding potential rate effects of accelerated irradiation studies. These results may also be used to confirm models of radiation-induced degradation at extended lifetime such as the ones proposed previously and for aging management programs.

The Jose Cabrera nuclear power station or "Zorita NPP" consists of a single-loop, Westinghouse PWR reactor with 160-MWe output. It was commissioned in 1968 and operated until 2006, accumulating 26.5 effective full power years of operation. In February 2010, Empresa Nacional de Residuos Radiactivos, S.A (ENRESA), the Spanish national company for nuclear waste, took responsibility for decommissioning the station. In 2009, the Consejo de Seguridad Nuclear (CSN), the Spanish Nuclear Safety Council, convened a meeting with national organizations to evaluate the feasibility of obtaining concrete from Zorita to research the effects of high radiation (neutron and gamma) and temperature on concrete structures. Similar to the Zorita Internals Research Project (ZIRP), Zorita's concrete structures could be of great value to developing an improved understanding of the effects of radiation on aging concrete because the neutron fluence on the Zorita CBS was approaching the onset for possible concrete deterioration as estimated by the Hilsdorf curve (Hilsdorf *et al.* 1978). In January 2010, the CEIDEN (Spanish Strategic Platform on Nuclear Research and Development) established a working group on Zorita's concrete cores. The current partners of the project are ENRESA, ENDESA (utility), IBERDROLA (utility), Gas Natural Fenosa (utility), licensee during operation of Zorita NPP, IETcc (Instituto Eduardo Torroja de Ciencias de la Construcción), CSN, and recently EPRI.

The structures being considered are the CBS (radiation and temperature effects), the spent fuel pool/transfer channel (boric acid degradation) and the containment building (as reference). The building also will be inspected through nondestructive testing of the steel liner under the concrete slab. The irradiated concrete tasks include: calculation of neutron and gamma fluence and temperature by numerical modeling; obtaining test specimens for chemical, mechanical, and microstructural characterization of irradiated and non-irradiated concrete. Specifically, core samples from the CBS will be drilled from areas of high irradiation, high temperature, and a combination of high irradiation and temperature. From each core sample, several test coupons will be obtained: the closest to the reactor vessel, one from the opposite side, and some intermediate coupons to verify the attenuation effects.

On behalf of the NRC, ORNL is reviewing information and reports provided by the Zorita Concrete Working Group and ENRESA to evaluate the concrete and conditions to determine whether it would serve as a suitable surrogate for US PWR NPPs. To determine the applicability to long-term aging degradation of US NPPs, three issues are being evaluated. The first issue is whether the concrete, and especially the aggregate, are the same or similar to concrete and aggregate found in the US NPPs. A careful study of concrete cores harvested from any decommissioned NPP would provide an opportunity to generate data from concrete that has experienced typical radiation fields and, therefore, would facilitate the effort to develop an understanding of the damage mechanisms in irradiated concrete. If the concrete and aggregate were not similar to concrete and aggregate found in some US NPPs, questions would remain as to its general applicability to extended operation of US NPPs. The second issue is whether the accumulated fluence is sufficient to create observable damage in the concrete and aggregate. Although the Zorita CBS has accumulated the highest radiation of any reactor currently scheduled for decommissioning, the fluence must reach the threshold level required to produce microstructural changes in the cement paste and the onset of RIVE in the aggregates. The third issue is the type of aggregate. Research has shown that siliceous aggregates exhibit higher swelling (damage) than calciferous aggregates (Field *et al.* 2015; Rosseel *et al.* 2015b). The greater the ratio of siliceous to limestone aggregates, the larger the observable effects of radiation on concrete. It is estimated that the evaluation will be completed in late 2016.

6.3 Irradiation-assisted alkali-silica reaction

Ion-implantation-induced disorder (amorphization) of the three-dimensional framework silicates, α-quartz (Ichikawa and Koizumi 2002; Pignatelli *et al.* 2016) and plagioclase (Ichikawa and Kimura 2007) results in an increase by several orders of magnitude of the dissolution rate in contact with a highly alkaline solution close to that of unirradiated amorphous silica (Pignatelli *et al.* 2016). Irradiated-amorphous silica also shows, to a lesser extent, a greater susceptibility to dissolution (Ichikawa and Koizumi 2002). Conversely, irradiation-induced density change in calcite is essentially caused by rotations and distortions of the carbonate groups with respect to the Ca atom positions leading to minor change of the dissolution rate (Pignatelli *et al.* 2016). The increasing reactivity of irradiated aggregate is one factor driving the formation of alkali-silica reaction gels. Two other aspects must be considered before any definitive conclusion can be made. ASR gels are highly hydrophilic but only internal moisture resulting from mix excess water is available for their formation, which could rapidly become a limiting factor. The stability of alkali-silica hydrates under gamma irradiation (radiolysis) is unknown and requires further research.

6.4 RIVE in restrained conditions

Although no literature data were found on the effect of mechanical restraint on the development of RIVE, it is likely that restrained RIVE is subject to the same effects as alkali-silica reaction (ASR) in concrete due to the similarity of both mechanisms (i.e., expansion of a specific phase contained in the aggregates). ASR expansion is reduced in the direction of the load and increased in the lateral directions (although the total volumetric expansion is not constant with the load level), while damage is oriented preferably in the direction of the load and increases with the load level (e.g., Larive 1997; Multon and Toulemonde 2006; Dunant and Scrivener 2012). Preliminary mesoscale simulations suggest that RIVE is indeed subject to such anisotropy of expansion and degradation when confined by external restraints (Giorla, Le Pape and Dunant 2016) although experimental verification is needed to conclusively determine this effect.

7. Conclusions

In most countries with operating NPPs, LWRs have been identified as an important component of electric power generation. For that reason, ensuring the safe, long-term operation of NPPs is critical to maintaining a reliable energy supply. Moreover, extending the operating lifetimes of current NPPs as allowed by national regulations, and making additional improvements in productivity using PLM or aging management is essential to meeting future national energy needs while reducing greenhouse gas emissions. To meet these goals, a critical evaluation of the knowledge gaps of the effects of exposure time to temperature, stress, coolant, and radiation fields on materials such as concrete is required. And although it is expected that the vast majority of concrete structures will continue to meet their functional and performance requirements during licensing periods, there may be isolated examples where structures may not exhibit the desired durability, primarily due to environmental effects such as radiation (Rosseel *et al*. 2014; Rosseel *et al*. 2015a).

In this paper, a review of the current state of knowledge on the effects of radiation on concrete in nuclear applications is presented. Emphasis is placed on the effects of radiation damage as reflected by changes in engineering properties of concrete in the evaluation of the LTO of NPPs and for PLM in Japan, Spain, and the United States.

Following a discussion of issues and concerns in Japan (post Fukushima Daiichi) and the United States (LTO and anticipated subsequent license renewal), this report focuses on the current status of the fundamental understanding of radiation effects on concrete and its components. Specifically, it addresses drying and temperature effects and radiolysis on HCP and RIVE of aggregate-forming minerals. This is followed by a discussion of the combined effects of temperature, drying, and radiation on concrete, including the significance of accelerated irradiation conditions, the role of temperature and creep, as well as variations in energy spectra and damage energy, and how these factors are being incorporated into numerical and mesoscale models of concrete structures. Using these insights, attempts to evaluate the impact of radiation-induced degradation by numerical simulations of concrete structures are reviewed, and the current status of work in Japan and the United States described.

The recent formation of a new international scientific and technical organization (the ICIC) to provide a forum for timely information exchanges among organizations pursuing the identification, quantification, and modeling of the effects of radiation on concrete in commercial nuclear applications is also described. The paper concludes with a discussion of current, high-priority research gaps including evaluating service-irradiated concrete from decommissioned NPPs, such as Zorita (Spain), to inform degradation models and aging management programs, evaluation of test reactor data, irradiated-assisted alkali-silica reactions, and RIVE under constrained conditions.

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