Recent Advances in Understanding Radiation Damage In Reactor Cavity Concrete

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ABSTRACT

License renewal up to 60 years and the possibility of subsequent license renewal to 80 years has resulted in a renewed focus on long-term aging of materials at nuclear power plants (NPPs) including concrete. Large irreplaceable sections of most nuclear generating stations include concrete. The Expanded Materials Degradation Analysis, jointly performed by the Department of Energy, the Nuclear Regulatory Commission and Nuclear Industry, identified the urgent need to develop a consistent knowledge base on irradiation effects in concrete (Graves et al., (2014)). Much of the historical mechanical performance data of irradiated concrete (Hilsdorf et al., (1978)) does not accurately reflect typical radiation conditions in NPPs or conditions out to 60 or 80 years of radiation exposure (Kontani et al., (2011)). To address these potential gaps in the knowledge base, the Electric Power Research Institute and Oak Ridge National Laboratory, are working to better understand radiation damage as a degradation mechanism. This paper outlines recent progress toward: 1) assessing the radiation environment in concrete biological shields, defining the upper bound of the neutron and gamma dose levels expected in the biological shield for extended operation, and estimating adsorbed dose, 2) evaluating opportunities to harvest and test irradiated concrete from international NPPs, 3) evaluating opportunities to irradiate prototypical concrete and its components under accelerated neutron and gamma dose levels to establish conservative bounds and inform 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 effort to leverage capabilities and knowledge including developing cooperative test programs to improve confidence in data obtained from various concretes and from accelerated irradiation experiments.

INTRODUCTION

As described in Rosseel et al. (2014), extending the operating lifetimes of current nuclear power plants (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 materials that comprise the structures and components of a NPP is required. Moreover, while 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 are also critical to long-term operation. For example, large sections of most NPPs have been constructed using concrete and the performance of reinforced concrete structures through the first 40 years of service has been very good. And although it is expected that the vast majority of these 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.

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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 support of the US Department of Energy (DOE), Light Water Reactor Sustainability (LWRS) Program, have 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 the DOE, 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 concrete expert panel was the urgent need to develop a consistent knowledge base on irradiation effects in concrete (Graves et al., (2014)).

This paper outlines recent progress on the joint EPRI / LWRS program to examine radiation as a degradation mechanism in NPP structural concrete. It focuses on: 1) assessing the radiation environment in concrete biological shields (CBS), defining the upper bound of the neutron and gamma dose levels expected at extended operation, and estimating absorbed dose, 2) evaluating opportunities to harvest and test service-irradiated concrete, 3) evaluating opportunities to irradiate prototypical concrete and its components under accelerated neutron and gamma dose levels to establish upper bounds and 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 effort to leverage capabilities and knowledge including developing cooperative test programs to improve confidence in data obtained from various concretes and from accelerated irradiation experiments.

RADIATION ENVIRONMENTS IN CONCRETE BIOLOGICAL SHIELDS

As discussed previously (Remec et al. (2014) and Rosseel et al. (2014)), radiation-induced concrete degradation historically has been correlated to neutron fluence and gamma-ray dose (Hilsdorf et al. (1978)). For that reason, estimates of the bounding fluence and dose values for the US NPP fleet at the projected extended service are vital. Moreover, due to the substantial differences in reactor designs and variations in plant operations, this effort is not trivial. Although routine monitoring of radiation fields in biological shields has not been established, every operating US NPP is required to implement a reactor pressure vessel (RPV) surveillance program so that a wealth of fluence information is publicly available from the US NRC ADAMS database (US NRC, (2015)). To capitalize on this information, coupled neutron and gamma ray transport calculations were performed at ORNL (Remec et al., (2014)) for a three-loop and a two-loop pressurized water reactor (PWR) to provide guidance on how to extrapolate the pressure vessel flux/fluence data to the flux/fluence in the CBS. An example of the attenuation of the neutron and gamma flux through the pressure vessel and biological shield is shown in Figure 1.

Figure 1: Radial variation of neutron flux at four energy ranges, gamma-ray flux, and heating rates for a three-loop pressurized water reactor. Graph reinterpreted from Remec et al. (2014).
Using the neutron transport calculations as guidance, a multi-step process was implemented by Bruck et al., (2013) to mine the ADAMS database to determine the expected neutron fluence values on the CBS for the current US boiling water reactor (BWR) and pressurized water reactor (PWR) fleet up to 80 years of operation. Specifically, RPV surveillance data reported to the NRC are extrapolated to an operating life of 80 years and attenuated to the vessel outside diameter wall. The results shown in Figure 2 highlight the importance of plant design, fuel loading scheme, and other operational procedures in determining the maximum expected fluence in the biological shield at 80 years of operation. For example, the four loop PWR reactors show significantly lower fluence values than the majority of both 2 and 3 loop plants. Note that BWR fluence is typically an order of magnitude lower than PWR fluence.

The ability to establish a more precise threshold fluence for the onset of radiation damage in concrete has been hampered by the large variation in residual compressive strength at any given neutron fluence value. As depicted in Hilsdorf et al., (1978), the scattering of the data, assembled from a large number of independent experiments, is caused by variations in numerous factors, such as different types of cement and aggregate, different cement-to-water ratios, different specimen sizes, variation in concrete curing and thermal treatment, and different specimen temperatures during irradiation. Of particular relevance is that the reported “neutron fluence” varies greatly between experiments. Some reports only provide qualitative descriptions such as “slow” or “fast” neutron fluence, others report total (E > 0 MeV) fluence or energies above 0.1 MeV or 1.0 MeV, while some experiments did not address the energy of the neutrons. Moreover, the neutron spectrum was actually provided only for a few experiments. For these reasons, it is not possible to “re-normalize” the data to a common denominator, such as fluence of neutrons with energies E > 0.1 MeV (Remec, 2014).

To address these concerns, Remec (2014) is evaluating the effects of radiation on concrete and its components by the number of times an atom of the material is displaced from its location in the lattice during the irradiation. This parameter, called displacements per atom (dpa), is well established as the preferred irradiation parameter for characterization of radiation effects on metals. Using the Specomp computer code to calculate dpa cross-sections for quartz (SiO$_2$) and calcite (CaCO$_3$), it was shown that these two widespread components of concrete aggregates have very similar variations with energy as silicon (Si), i.e., from a minimum at around 100 eV the cross sections rapidly increase with increasing neutron energy. The relative contributions to the dpa rate were calculated with the neutron fluence rate spectrum in the cavity of a two-loop PWR and three-loop PWR, obtained from previous analyses by Remec et al. (2014). As shown in Figure 3, neutrons with energies above 1 MeV cause only about 20 % to 25 % of the total atom displacements while neutrons with energies above 0.1 MeV contribute more than 95% of all atom displacements.
Figure 3: Relative contribution of neutrons with energies $E > E_0$ to the dpa rate for the neutron spectrum in the cavity of a two-loop PWR.

HARVESTING AND TESTING SERVICE IRRADIATED CONCRETE

Over the last 15 years, several NPPs in the United States and other countries have been decommissioned, are in the process of being decommissioned, or decommissioning is expected to commence in the near future. Examples include the Zion Nuclear Generating Station (NGS), Millstone 1, Indian Point Unit 1, Crystal River 3, Zorita (Spain), and Krummel (Germany). Harvesting concrete cores from decommissioned NPPs will provide an opportunity to obtain data from concrete that has experienced typical radiation fields while also providing guidance to accelerated irradiation studies. For example, the coupling of laboratory-irradiated concrete data with the characterization of harvested NPP cores may facilitate the effort to develop an enhanced understanding of the damage mechanisms in irradiated concrete, including understanding possible accelerated radiation rate effects. The acquisition of concrete cores will, however, need to be supplemented by extensive investigations into the operating history of the NPP as well as the concrete composition and performance history. Information such as the material test reports for cement, admixtures, and aggregates; concrete mix design; and aggregate characterization, including type, petrographic analyses, and gradation would be of high value. Other critical information might include concrete property test results, such as the modulus of elasticity, the reference 28-day compressive strength and concrete strength testing over the life of the plant (Rosseel et al, (2014)).

The LWRS Program has entered into discussions with Zion Solutions, a subsidiary of Energy Solutions, to harvest concrete cores from eight to ten locations (Figure 4) at the Zion Unit 2 NPP. These cores will be characterized for changes in mechanical properties (strength, modulus, hardness, and density) and microstructural features at the onset of possible aggregate swelling and cement paste cracking. The goal is to develop a better understanding of and ability to predict concrete degradation at extended lifetimes. Research will be focused on (1) validating predictive models based on accelerated aging studies with empirical data obtained from field-aged concrete in radiation and thermal environments, and (2) evaluating concrete radiation gradients in the CBS to investigate the changes in properties as a function of the level of radiation. With the addition of concrete from ambient or controlled environments, it may be possible to separate the effects of radiation and thermal environments (Rosseel, (2015)).

It is anticipated that Zion NPP concrete data will be integrated with data from other decommissioned reactors and possibly with data from accelerated irradiation testing of well-characterized NPP-like concrete. For example, the NRC has initiated discussions with the Zorita Consortium concerning options
Figure 4: Proposed Zion Unit 2 Concrete Core Harvest Locations.

IRRADIATION OF PROTOTYPICAL CONCRETE

Until recently, the data on the mechanical degradation of concrete due to long-term irradiation were thought to have been quite limited. Moreover, recent studies have suggested much of the data compiled in the Hilsdorf Curve (Hilsdorf et al., 1978) were not representative of concrete mixtures, temperatures, and radiation fields seen in LWRs (Kontani et al., 2010). Experiments by Fujiwara et al. (2009), however, appear to support Hilsdorf data for neutron irradiations while results from Vodak et al. (2005) show concrete degradation at much lower gamma-ray doses. The reasons for these seemingly contradictory interpretations are two-fold. For many of the older experiments, important information such as the neutron fluence energy, concrete composition, irradiation temperature, and gamma-ray dose is often limited or missing. Consequently, the applicability to NPP concrete may be uncertain. Furthermore, critical data may not have been in the public domain when Hilsdorf’s report was completed and fundamental studies following its publication had not been compiled (Rosseel and LePape, 2014).

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 radiation damage. The newly collected database contains about 300 (30) compressive strength data, 60 (30) tensile strength data, 140 (30) elastic modulus data and 110 dimensional change data. Note, the numbers in brackets indicate the approximate number of data points in Hilsdorf’s report. Although the new data also have limitations due to variations in irradiation temperature, neutron energy, and different aggregate types, which results in the scatter band observed in Figure 5, a decrease in compressive strength above a neutron dose of $2.0 \times 10^{19} \text{ n/cm}^2$ is suggested with an average loss of strength of about 50% of the initial strength at $1.0 \times 10^{20} \text{ n/cm}^2$. Similar trends are observed with tensile strength and elastic modulus data with average losses of, respectively, about 60% and 30% of the initial strength at $1.0 \times 10^{20} \text{ n/cm}^2$. Moreover, the development of a specifically targeted micromechanical model using the available data from literature confirmed the predominant role of aggregates on the macroscopic expansion of irradiated concrete and the development of damage in the cement paste. In particular, ORNL studies (LePape et al. (2015)) support the hypothesis formulated by Seeberger and Hilsdorf, (1982), concerning the role of silicate-bearing aggregates on radiation effects on concrete. Furthermore, Vanelstraete and Laermans, (1990), have shown that fast neutrons ($E > 0.1 \text{ MeV}$) cause displacement cascades in quartz resulting in disordered regions of the crystal. For sufficiently high doses, these regions overlap, reducing long-range ordering and resulting in amorphization of the SiO$_2$ phase. The loss of ordering is observed as a reduction
of density and increase in swelling of the quartz phase. Complete amorphization is thought to be reached at a neutron dose of $> 2.0 \times 10^{20}$ n/cm$^2$ (Rosseel and LePape, 2014).

![Figure 5: Relative compressive strength of concrete and mortar specimens versus neutron fluence. The irradiation temperature and reported neutron energy cut-off varies with each data point. Siliceous concrete is depicted with red symbols, calcareous with blue, and miscellaneous concretes with green. Graph reinterpreted from Field et al. (2015).](image)

Based on the re-evaluations of the accelerated irradiated concrete data found in the literature and the confirmation of the predominant role of aggregates on the macroscopic expansion of irradiated concrete due to the development of internal damage of the aggregates by Field et al. (2015), it was determined that well-defined experiments that reduce uncertainty were needed. Moreover, the effects of elevated temperatures and dose rate have not been fully evaluated for accelerated irradiation experiments. For these reasons, single variable experiments to investigate RIVE of aggregates and concomitant swelling and possible loss of structural integrity of NPP concrete at fluences approximating extended lifetime conditions have been initiated. Specifically, mineral analogues of concrete aggregates have been irradiated as a function of dose ($E > 0.1$ MeV) at two temperatures (50 °C and 250 °C). The fluences were selected based on estimates of the onset of swelling through amorphization or saturation. The two temperatures were selected based on service irradiation conditions ($< 65^\circ$C) and conditions used during the majority of accelerated irradiation studies reported in the literature (Rosseel and LePape, 2014).

As of the preparation of this report, data on post-irradiation changes in dimensions, mass, hardness, mechanical properties, such as stress, strain, elastic modulus, and microstructure are being collected and evaluated. If this experiment is successful, a larger scale experiment will be performed followed by a study of aggregate swelling.

**IMPROVED UNDERSTANDING OF THE EFFECTS OF RADIATION ON CONCRETE**

As noted above, the correlation between the loss of mechanical properties and the development of radiation-induced swelling is quite strong. Irradiated concrete damage is a convolution of several effects: thermal expansion of the concrete constituents, radiolysis-accelerated shrinkage of the cement paste, and radiation-induced aggregate expansion that results in cracking of the cement paste and at the aggregate-
paste interfaces. As shown in figure 6, the development of a micromechanical model using the available data from Kelly et al. (1969) and Elleuch et al. (1972) confirmed the predominant role of aggregate swelling on the development of internal damage and of the macroscopic expansion of irradiated concrete (LePape et al. (2015)).

![Figure 6: Post-irradiation length change. Left: Measured aggregate expansion. The solid lines correspond to the best fitting curve assuming Zubov and Ivanov's (1966) model. Right: Measured expansions and theoretical estimates derived by micromechanical modeling. Reinterpreted from LePape et al. (2015).](image)

A recent report by Giorla (2015) builds upon previous research results that were unable to simultaneously predict both radiation-induced swelling of aggregates and damage in concrete by using new tools to establish a deeper understanding of the underlying mechanisms. Using a finite element mesh that represents aggregates as inclusions within the cement paste allows a precise description of the localization and rate of propagation of damage in the microstructure, accounting for the influence of neutron fluence, temperature, humidity, and strain rate on its properties. The result is the development of a numerical model for concrete exposed to irradiation that accounts for cracking and creep in the cement paste and its coupling with the effects of damage, temperature and relative humidity. In future work, the complete model will be applied to the analysis of irradiation experiments that requires a careful examination of the experimental environmental conditions. Once the model has been validated, it could be applied to simulate concrete from nuclear power plants.

**INTERNATIONAL RESEARCH AND INFORMATION EXCHANGE**

Understanding the effects of radiation on concrete is important in determining long-term or extended operating performance of concrete structures in existing NPPs. As partially described in Rosseel et al., (2014), this issue is being addressed by research organizations and utilities across the globe. Moreover, in the last three years, the LWRS Program has 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 maximizing resources. As part of that effort, two international meetings, entitled, “International Irradiated Concrete Information Exchange Framework Meetings,” were proposed and organized by ORNL in Barcelona and Helsinki. The foundation of these meetings is that international cooperation will provide the best opportunities to share resources, acquire valuable specimens from decommissioned NPPs, and build a systematic database to provide a framework for decisions concerning extended operation of NPPs in a timely and efficient manner. The first meeting was hosted and held in cooperation with the Consejo Superior de Investigaciones Científicas - CSIC (Spanish National Research Council). Nineteen researchers from five countries attended the meeting, which was held at the Hotel Colon in Barcelona, Spain, on March 12-14, 2014. The second meeting was hosted and held in cooperation with Fortum, Oyj. Twenty-five attendees from 18 organizations and seven countries attended the second meeting, which was held at Fortum in Helsinki, Finland, on October 8 – 10, 2014.

The purpose of these meetings was three-fold. First, to provide a forum for discussing issues that advance the state of knowledge of the effects of radiation on concrete used in nuclear facilities. Secondly, to develop the framework for exchanging information on a broad set of topics related to the effects of radiation on concrete used in NPPs by those who are actively pursuing research, were active in the field, or wish to contribute to advancing the current state of knowledge. This included a discussion of the types of information that could be exchanged, the level of release, an organizational framework for cooperation including resource and data sharing, and the development of a charter based on the International Group on Radiation Damage Mechanisms (IGRDM) in reactor pressure vessels. Thirdly, if the attendees were in agreement, to launch a new international organization, the International Committee on Irradiated Concrete (ICIC), with a defined organizational structure, requirements for membership and regular future meetings.

At the conclusion of the Helsinki meeting, the participants approved the ICIC Charter with 19 of the participants and 16 organizations accepting membership in the duly constituted ICIC. The participants also selected four Technical Areas (TAs) and corresponding Technical Area Coordinators (TACs). The main function of the TAs and TACs is to facilitate and expedite progress by the membership in specified technical topics. The four TAs and TACs selected by vote of the members are as follows:

1. Structural Performance and Mechanistic Understanding of the Effects of Radiation on Concrete (Y. Le Pape – ORNL, USA)
2. Harvesting and Characterization of Service Irradiated Concrete (M. Ordonez, ENRESA, Spain)
3. Accelerated Irradiation Studies of Concrete & Components (M. Koleska, RC-Rez, Czech Repub.)
4. Characterization of Irradiated Concrete (C. Andrade, CSIC, Spain)

Finally, the participants elected T. M. Rosseel, ORNL, as Chairman, I. Maruyama, Nagoya University, as Vice Chairman, and M. Ferreira, VTT, as Secretary of the ICIC. It is anticipated that the first General Meeting of the ICIC will be held in the US in late October 2015.

CONCLUSION

This paper describes recent advances in understanding radiation damage in reactor cavity concrete by a collaborative effort between the EPRI LTO Program and ORNL, through the support of the US DOE, LWRS Program, based on the EMDA concrete expert panel’s (Graves et al., 2014) key finding of the urgent need to develop a consistent knowledge base on irradiation effects in concrete. A summary of the status and progress on this effort is listed below.

- Based on the guidance from neutron transport calculations, RPV surveillance data reported by utilities can be extrapolated into the CBS to an operating life of 80 years and some US PWRs will exceed levels of $2 \times 10^{19}$ n/cm$^2$. 
• Neutrons with energies $>0.1$ MeV contribute more than 95% of all atom displacements suggesting its use as the best cut-off energy for evaluating radiation damage.

• Harvesting concrete cores from decommissioned NPPs is expected to provide a better understanding of and ability to predict concrete degradation at extended lifetimes by focusing on validating predictive models based on accelerated aging studies with empirical data obtained from field-aged concrete in radiation and thermal environments.

• Accelerated irradiation experiments will require considerably higher neutron and gamma fluxes than those observed in the CBS. For that reason, the analysis of the temperature of the samples will be necessary and additional cooling of concrete samples may be required. Moreover, the effect of dose rate has not been fully evaluated.

• A comprehensive literature review and reanalysis by Field et al. (2015) has greatly expanded the Hilsdorf et al. (1978) database and confirmed the predominant role of RIVE. Although the new data have limitations, a decrease in compressive strength above $2.0 \times 10^{19}$ n/cm$^2$ is suggested with an average loss of strength of about 50% at $1.0 \times 10^{20}$ n/cm$^2$. Similar trends are observed with tensile strength and elastic modulus data.

• Single variable studies of mineral analogues and aggregates are expected to provide critical information for interpreting irradiated concrete results.

• The development of a micromechanical model using the available data from literature confirmed the predominant role of silicate-bearing aggregates on the development of internal damage and of the macroscopic expansion of irradiated concrete.

• New meso-scale models are being developed using a finite element mesh to represent aggregates as inclusions within the cement paste to incorporate geometrical interactions, that are making it possible to evaluate the influence of neutron fluence, temperature, humidity, and strain rate on concrete properties.

• A new international organization, the ICIC, has been formed to provide a forum for discussing issues that advance the state of knowledge of the effects of radiation on concrete. It is anticipated that this information exchange will leverage capabilities and knowledge, including developing cooperative test programs, and improve confidence in data obtained from service-irradiated and prototypical concrete experiments.

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