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 established a renewed focus on long-term aging of nuclear generating stations materials, and recently, on concrete. Large irreplaceable sections of most nuclear generating stations include concrete. The Expanded Materials Degradation Analysis (EMDA), jointly performed by the Department of Energy, the Nuclear Regulatory Commission and Industry, identified the urgent need to develop a consistent knowledge base on irradiation effects in concrete [1]. Much of the historical mechanical performance data of irradiated concrete [2] does not accurately reflect typical radiation conditions in NPPs or conditions out to 60 or 80 years of radiation exposure [3]. To address these potential gaps in the knowledge base, The Electric Power Research Institute and Oak Ridge National Laboratory are working to disposition radiation damage as a degradation mechanism. This paper outlines the research program within this pathway including: (i) defining the upper bound of the neutron and gamma dose levels expected in the biological shield concrete for extended operation (80 years of operation and beyond), (ii) determining the effects of neutron and gamma irradiation as well as extended time at temperature on concrete, (iii) evaluating opportunities to irradiate prototypical concrete under accelerated neutron and gamma dose levels to establish a conservative bound and share data obtained from different flux, temperature, and fluence levels, (iv) evaluating opportunities to harvest and test irradiated concrete from international NPPs, (v) developing cooperative test programs to improve confidence in the results from the various concretes and research reactors, (vi) furthering the understanding of the effects of radiation on concrete (see companion paper) and (vii) establishing an international collaborative research and information exchange effort to leverage capabilities and knowledge.

INTRODUCTION:

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. In order to meet these goals, a critical evaluation of the knowledge gaps of materials that comprise the structures and components of a NPP must be addressed. 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 are also critical to long-term operation.

For example, large sections of most NPPs have been constructed using concrete and, in general, the performance of reinforced concrete structures through the first 40 years of service has been very good. Although it is expected that the vast majority of these structures will continue to meet their functional and performance requirements during the current and any future licensing periods, there may be isolated examples where structures may not exhibit the desired durability, primarily due to environmental effects.

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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 concrete used in NPPs. 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 [1].

This paper outlines the joint EPRI / LWRS program to examine radiation as a degradation mechanism in NPP structural concrete. It includes: (i) assessing the radiation environment in concrete biological shields and defining the upper bound of the neutron and gamma dose levels expected in the biological shield concrete for extended operation (80 years of operation and beyond), (ii) evaluating opportunities to harvest and test irradiated concrete from international NPPs, (iii) evaluating opportunities to irradiate prototypical concrete under accelerated neutron and gamma dose levels and to share data obtained from different flux, temperature, and fluence levels, (iv) furthering the understanding of the effects of radiation on concrete (see companion paper) and (v) establishing an international collaborative research and information exchange effort to leverage capabilities and knowledge including developing cooperative test programs to improve confidence in the results from the various concretes and research reactors.

RADIATION ENVIRONMENTS IN CONCRETE BIOLOGICAL SHIELDS

Radiation-induced degradation of concrete has been historically correlated to neutron fluence and gamma-ray dose [2]. Estimates of the bounding fluence and dose values for the US NPP fleet at the projected extended service are, therefore, clearly needed. Given the substantial design differences among manufacturers and variations in plant operations, this is a non-trivial task. Moreover, no routine monitoring of radiation fields in biological shields has been established. However, since every operating NPP in the USA is required to implement a reactor pressure vessel (RPV) surveillance program, a wealth of information from these programs is publicly available from the US NRC ADAMS database [4]. To capitalize on this information, a two-pronged approach was implemented: (a) coupled neutron and gamma-ray transport calculations performed on two selected pressurized water reactors (PWRs) and (b) a search of the ADAMS database to collect data from the RPV surveillance programs guided by the neutron and gamma-ray transport calculations.

Coupled neutron and gamma transport calculations using the Monte Carlo N-Particle (MCNP) Code for one, three-loop and one, two-loop pressurized water reactor (PWR) were conducted at ORNL [5]. Examples of the neutron and gamma flux variations through the pressure vessel and biological shield are shown in Figure 1. Neutron fluxes for all energies, except the thermal neutron flux, exhibit a monotonic decrease through the RPV, cavity, and inside the concrete shield. The rate of attenuation, however, varies depending on the composition of the material.

Several observations and conclusions from these radiation transport simulations of neutron and gamma fields are described as follows:

- Fast neutron ($E > 1$ MeV and $E > 0.1$ MeV) fluxes at the pressure vessel outer radius are 20 to 30% higher than the maximum fluxes in concrete. Therefore, the fluxes determined at the RPV outer diameter wall could be used as conservative estimates for fluxes in the concrete shield or for screening purposes.
- The total neutron and gamma flux at the pressure vessel wall agree within 10% of the maximum values in concrete. Heating rates calculated at the pressure vessel maximum flux location also are 10 to 20% higher than the highest rates in concrete.
- It is not practically possible to renormalize results from fast neutron fluence to total neutron fluence or vice versa without knowing the details of the irradiation environment.

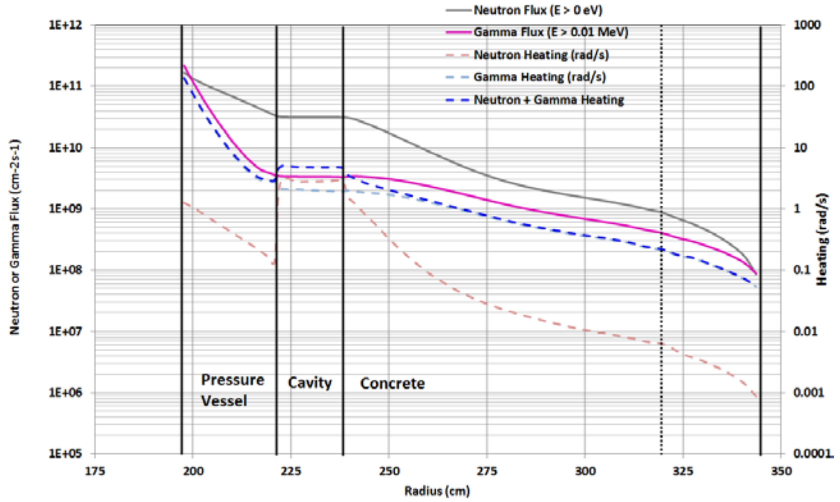

Figure 1: Radial variation of neutron and gamma-ray flux and heating rates for a three-loop pressurized water reactor. Graph reinterpreted from [5].

Based on the guidance from the neutron transport calculations, a multi-step process was implemented [6] to mine the ADAMS database additional information on the expected neutron fluences values in the biological shield for the current US PWR fleet up to 80 years of operation. RPV surveillance data reported by utilities to the NRC are extrapolated to an operating life of 80 years and attenuated to the vessel outside diameter (OD) wall. Furthermore, since the RPV surveillance data are typically reported for energies greater than 1.0 MeV while concrete current accelerated testing programs are reporting fluences with E > 0.1 MeV, an additional adjustment, which depends on the thickness of the vessel (as noted by the neutron transport calculations), was implemented. This value was defined as the maximum fluence that is expected to exist in the concrete.

The results shown in Figure 2 highlight the importance of considering the 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.

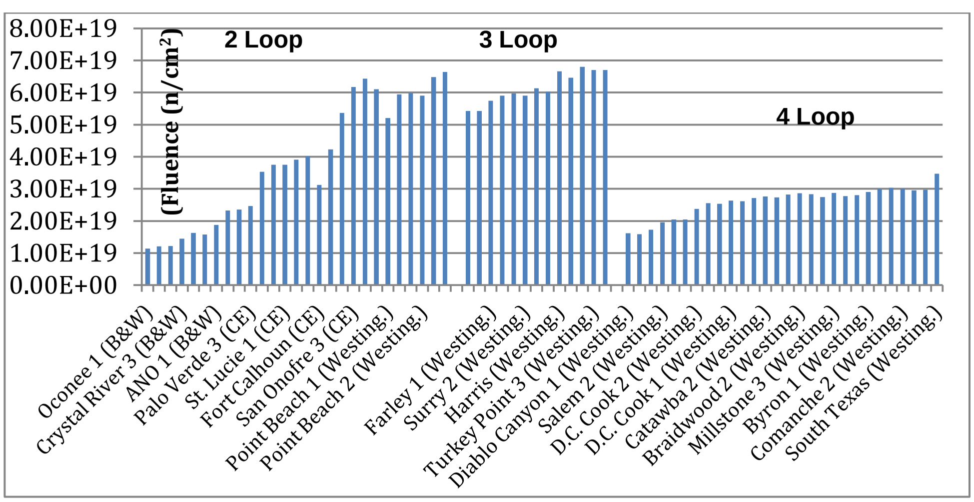

Figure 2: Summary of US PWR Fleet Fluence extrapolated to 80 years (E > 0.1 MeV). Graph reinterpreted from [6].

HARVESTING AND TESTING SERVICE IRRADIATED CONCRETE

During the last fifteen years, several NPPs in the United States and other countries have been decommissioned, are in the process of being decommissioned, or decommissioning will commence soon. Examples include Zion Nuclear Power Station Units 1 and 2, Millstone 1, Indian Point Unit 1, Crystal River 3, Zorita (Spain), Krummel (Germany), and Barseback (Sweden). Harvesting of concrete cores from decommissioned NPPs will provide an opportunity to generate data from concrete that has experienced typical radiation fields, while also providing guidance to accelerated irradiation studies. The coupling of accelerated or laboratory-irradiated concrete with harvested NPP cores is also expected to facilitate the effort to develop an understanding of the damage mechanisms in irradiated concrete, including understanding potential effects of accelerated testing.

In support of extended service of the U.S. nuclear reactor fleet, ORNL, through the LWRS Program, is coordinating and contracting with Zion Solutions, LLC (a subsidiary of Energy Solutions) the selective procurement of materials, structures, and components, including concrete from the decommissioned Zion reactors [7]. Physical acquisition of concrete cores will need, however, to be supplemented by extensive investigations into the operating history of the NPP, as well as the concrete composition and performance history. Collection of information such as the material test reports for cement, admixtures, and aggregate; concrete mix design; and aggregate characterization, including type, petrographic analyses, and gradation will be attempted. Other critical information might include concrete property test results such as the reference 28-day compressive strength and modulus of elasticity, results from any concrete strength testing over the life of the plant, and the American Society of Mechanical Engineering (ASME) inspection reports. 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 nuclear power plant-like concrete. For example, ORNL has initiated discussions with its CONSAFESYS (Concrete Containment Condition Status and Aging Examination System) partners concerning harvesting concrete cores from Barseback NPP Units 1 and 2. Proposed locations would focus on regions of elevated radiation, elevated temperature, elevated radiation and temperature, and ambient conditions.

The monotonic decrease of the neutron and gamma flux, as well as heating effects, as a function of distance from the reactor core may be highly beneficial in assessing the effects on concrete because different locations along a concrete core could provide specimens at different neutron fluences and gamma doses after serial sectioning. For example, from a single core, trend curves of mechanical properties versus fluence or dose could be generated. These results could be correlated with accelerated testing programs to extrapolate the effect of 80 years of operation on the mechanical performance of concrete.

IRRADIATION OF PROTOTYPICAL CONCRETE

Over the last decade, the data on the mechanical degradation of concrete due to long-term irradiation were thought to have been quite limited. For example, Fillmore [8] concluded, based mostly on the Hilsdorf et al. [2] compilation, that no effects of radiation on strength were found for neutron fluence less than 10^{19} n/cm^2 or gamma-ray doses less than 10^8 Gy for periods less than 50 years. Moreover, he noted that reductions in compressive and tensile strength and a marked increase in volume are reported for neutron fluences greater than about 10^{20} n/cm^2 or gamma-ray doses 2 x 10^8 Gy. However, data at intermediate fluences and doses were judged to be inconsistent. Kontani et al. [9] raised concerns on the classical interpretation of "Hilsdorf's curves" primarily because some data, in particular beyond 5.0×10^{19} n/cm^2, were not representative of concrete mixtures and radiation fields seen in LWRs. Experiments by Fujiwara et al. [10], however, appear to support the Hilsdorf data for neutron irradiations, while results presented by Vodak et al. [11] show degradation of concrete at much lower - 3 to 5 x 10^5 Gy - gamma-ray doses.

The reasons for these seemingly contradictory interpretations are two-fold. For many older experiments, important information such as the cutoff energy of the neutron fluence, the composition of concrete, irradiation temperature, and gamma-ray dose is often limited or missing. Consequently, the applicability to NPP concrete would appear to be uncertain. Furthermore, critical data may not have been in the public domain when Hilsdorf's report was completed and fundamental studies following the publication of the Hilsdorf paper had not been compiled into a single source. Recently, a comprehensive review of the literature by Field et al. [12] has greatly expanded the database with the goal of identifying first order effects in irradiated concrete. These issues will be discussed in greater detail in the section on understanding the effects of radiation on concrete.

More data under carefully controlled conditions would provide a firmer foundation for understanding the effects of radiation on existing NPPs. It is suggested that planning of experiments with accelerated irradiation of concrete should consider the following issues:

- It is desirable to obtain full neutron and gamma spectra in the samples for irradiation experiments since the relevant irradiation parameters for concrete have not been established.
- Irradiation experiments should be designed, if possible, so that the neutron and gamma spectra in the concrete samples will be similar to those observed in the biological shields of NPPs.
- Accelerated irradiation experiments will require considerably higher neutron and gamma fluxes than those observed in the shields on nuclear power plants. The acceleration factor and, therefore, fluxes are anticipated to be higher by a factor of 50 to 100. 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 evaluated.
- Studies of mineral analogues and aggregates may provide critical information for interpreting irradiated concrete results.

High-flux neutron irradiation facilities available at research reactors provide an opportunity for accelerated irradiation of concrete. These facilities potentially can meet the criteria for accelerated testing and, in fact, groups from Japan [13] and Finland have initiated experiments at the Institute for Energy Technology's JEEP-2 reactor facility at Kjeller, Norway, to address the question of reliable data. Other options include the LVR-15 reactor at Research Centre Rez, Czech Republic, the Advanced Test Reactor at the Idaho National Laboratory (INL), or the High-Flux Isotope Reactor (HFIR) at ORNL. Moreover, irradiations at research reactors could provide similar fluence/heating rate versus depth curves as observed in biological shields of NPPs, which would help develop testing techniques for harvested cores as discussed in the previous section.

IMPROVED UNDERSTANDING OF THE EFFECTS OF RADIATION ON CONCRETE

Acknowledging that Hilsdorf's review [2] and derivative works did not pay sufficient attention at extracting the data from the research works initially cited, and that important data may not have been in the public domain when the Hilsdorf report was completed, and expecting that more experiments were performed since that time, ORNL conducted an additional literature search with the objective to expand the database and to attempt to identify possible detrimental effects in irradiated concrete. Details are provided in [12]. 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; the numbers in brackets indicates the approximate number of data points in Hilsdorf's report. The new database is not immune from intrinsic limitations: (1) most experiments above $1.0\text{x}10^{19}$ n/cm^2 were conducted at temperatures higher than 100 ^{o}C while LWR concrete is limited to 65 ^{o}C by design; (2) the presented data could not be normalized consistently by a specific neutron energy cut-off creating some uncertainty on the actual neutron fluence; and (3) the normalizing concrete property varies with the authors (concrete age, curing, geometry). These unfortunately irreducible limitations added to the uncertainties created by different aggregate type and concrete mixtures collected, and are considered as a primary cause for the scatter band observed in Figure 3. Despite these limitations, a decrease in compressive strength above 2.0×10^{19} n/cm^2 is suggested with an average loss of strength (dashed line) of about 50% at 1.0×10^{20} n/cm^2. Similar trends are observed with tensile strength and elastic modulus data with average losses of, respectively, about 60% and 30% at 1.0×10^{20} n/cm^2.

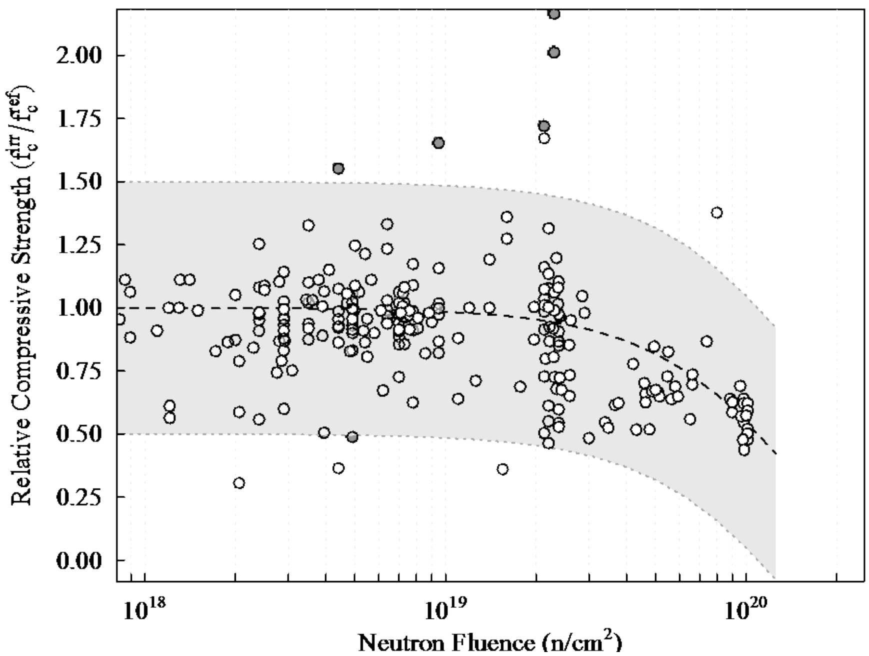

Figure 3: Relative compressive strength of concrete and mortar specimens versus neutron fluence limited to the range of 1×10^{18} n/cm^2 to 2×10^{20} n/cm^2. Note irradiation temperature and reported neutron energy cut-off varies depending from data point to data point. Graph reinterpreted from [12].

Among the hypothesized irradiation damage mechanisms in concrete, the role of radiation-induced volumetric expansion (RIVE) is suggested to be quite predominant [12,14]. Many authors have also reported the post-irradiation dimensional change (free swelling) of concrete. The correlation between the loss of mechanical properties and the development of radiation-induced swelling is particularly striking. Irradiated concrete expansion is a convolution of several effects: (possibly differential) thermal expansions of the concrete constituents; radiolysis accelerated drying shrinkage of the cement paste; aggregate radiation-induced expansion: cracking in the cement paste and at the aggregate-paste interfaces resulting from the previously listed dimensional changes. The development of a specifically targeted micromechanical model using the available data from literature (Figure 4) confirmed the predominant role of aggregate on the development of internal damage and of the macroscopic expansion of irradiated concrete [15]. In particular, ORNL studies supported the hypothesis formulated by Seeberger and Hilsdorf [14] about the role of silicate-bearing aggregates on irradiated concrete residual properties.

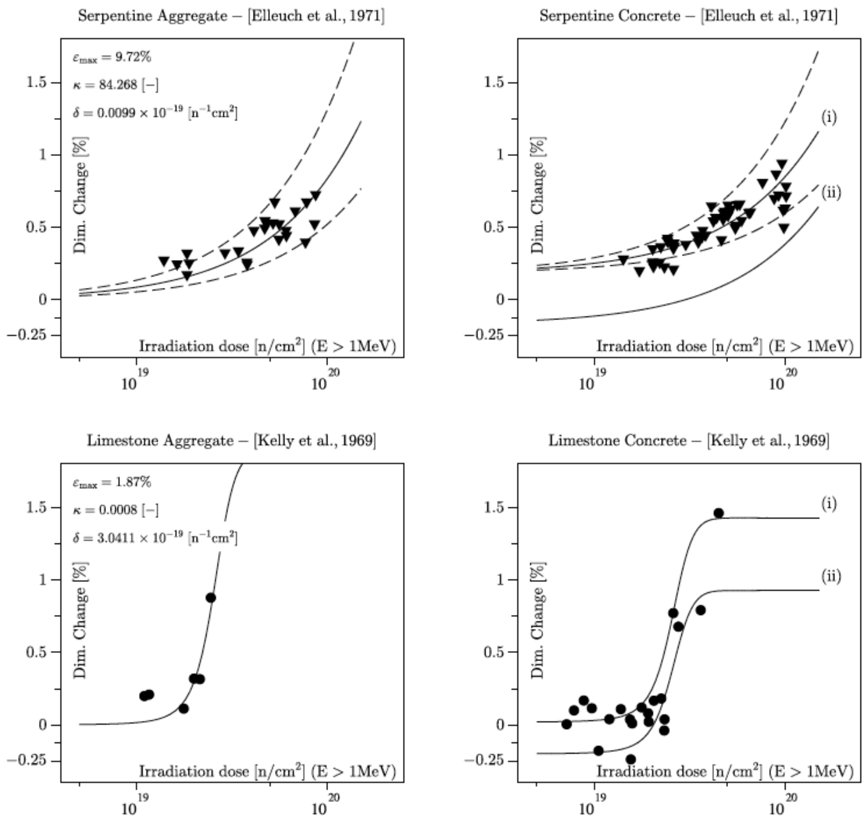

Figure 4: Post-irradiation length change. Left column: Measured aggregate expansion. The solid lines correspond to the best fitting curve assuming Zubov and Ivanov's model [16] (parameters given above the respective plots). Right column: Measured concrete expansions and theoretical estimates derived by micromechanical modeling. Graph reinterpreted from [15].

International Collaborative Research and Information Exchange Framework

Understanding the effects of radiation on concrete is important 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. Moreover, in the last two 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, an international meeting, entitled, "International Irradiated Concrete Information Exchange Framework Meeting," was proposed and organized by ORNL in cooperation with Professor Carmen Andrade, 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 foundation for this meeting is the understanding 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 nuclear power plants in a timely and efficient manner.

The purpose of this meeting was two-fold. First, to develop the framework for exchanging information on a broad set of topics related to the effects of irradiation on concrete used in NPPs by those who are actively pursuing research, were active in the field, or wish to contribute to the advancing the current state of knowledge. And second, to provide a forum for discussing issues that advance the state of knowledge of the effects of irradiation on structural concrete used in nuclear reactor facilities including storage sites.

The first portion of the meeting included presentations and discussions on past and current irradiated concrete research and / or issues related to irradiated concrete. The second portion of the meeting focused on establishing the framework for exchanging information. 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 RPVs.

The final portion of the meeting focused on making a decision or commitment by attendees to participate in the information exchange. At the conclusion of the meeting, the participants reached a consensus to move forward with the International Committee on Irradiated Concrete (ICIC) and that a follow-up meeting should be held within six months to finalize its charter and elect an executive committee. Moreover, the participants endorsed a plan to determine the frequency and location of future ICIC meetings. It is anticipated that future meetings will be held on a rotating schedule in Europe, the US, and Japan. The meeting concluded with the election of Dr. Thomas M. Rosseel, ONRL, Acting Chair, and Dr. Carlos Castelao, Consejo de Seguridad Nuclear, (CSN), Acting Vice Chair, to lead the interim process. Information on the ICIC, the draft charter, and meeting agenda and presentations can be found on the ICIC web site. [17]

CONCLUSIONS:

The EPRI LTO Program, and ORNL, through the support of the US DOE, LWRS Program, have established a collaborative research effort to investigate the effects of radiation on NPP concrete. This effort has focused on developing five parallel pathways: (i) assessing the radiation environment in concrete biological shields and defining the upper bound of the neutron doses in the biological shield concrete at extended operation, (ii) evaluating opportunities to harvest and test irradiated concrete from international NPPs, (iii) evaluating opportunities to irradiate prototypical concrete under accelerated neutron and gamma doses and to share data obtained at different flux, temperature, and fluence levels, (iv) furthering the understanding of the effects of radiation on concrete, and (v) establishing an international collaborative research and information exchange effort to leverage capabilities and knowledge including developing cooperative test programs to improve confidence in the results from the various concretes and research reactors. A summary of the progress and status of this work is listed below.

- Based on the guidance from neutron transport calculations, RPV surveillance data reported by utilities can be extrapolated into the biological shield to an operating life of 80 years.
- Projected neutron fluence values ($E > 0.1$ MeV) in the concrete biological shields of some US PWRs will exceed levels of 2×10^{19} n/cm^2.
- Harvesting concrete cores from decommissioned NPPs is feasible and will provide an opportunity to generate data from concrete that has experienced typical radiation fields while also providing guidance to accelerated irradiation studies. Physical 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.
- The coupling of accelerated or laboratory-irradiated concrete with harvested NPP cores is expected to facilitate the effort to develop an understanding of the damage mechanisms in irradiated concrete, including understanding potential effects of accelerated testing.
- It is desirable to obtain full neutron and gamma spectra for irradiation experiments since the relevant irradiation parameters for concrete have not been established.
- Irradiation experiments should be designed, if possible, so that the neutron and gamma spectra in the concrete samples will be similar to those observed in biological shields.
- Accelerated irradiation experiments will require considerably higher neutron and gamma fluxes than those observed in the shields on NPPs. The acceleration factor and, therefore, fluxes are anticipated to be higher by a factor of 50 to 100. 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 evaluated.

- Studies of mineral analogues and aggregates may provide critical information for interpreting irradiated concrete results.
- Previous interpretations of the Hilsdorf curves provided a valuable initial assessment of radiation effects but, more data, under carefully controlled conditions, would provide a firmer foundation for understanding the effects of radiation on existing NPPs.
- A comprehensive review of the literature by Field et al. [12] has greatly expanded the database and confirmed the predominant role of radiation-induced volumetric expansion (RIVE). Although the new data are not immune to limitations, a decrease in compressive strength above $2.0x10^{19}$ n/cm^2 is suggested with an average loss of strength (dashed line) of about 50 % at $1.0x10^{20}$ n/cm^2. Similar trends are observed with tensile strength and elastic modulus data with average losses of, respectively, about 60 % and 30 % at $1.0x10^{20}$ n/cm^2.
- The development of a specifically targeted micromechanical model using the available data from literature confirmed the predominant role of aggregates on the development of internal damage and of the macroscopic expansion of irradiated concrete. In particular, ORNL studies [15] supported the hypothesis formulated by Seeberger and Hilsdorf [14] concerning the role of silicate-bearing aggregates on irradiated concrete residual properties.
- A new international organization, the International Committee on Irradiated Concrete (ICIC), has been formed to provide the framework for exchanging information on a broad set of topics related to the effects of irradiation on concrete. The purpose is to provide a forum for discussing issues that advance the state of knowledge of the effects of irradiation on structural concrete used in nuclear reactor facilities including storage sites. It is anticipated that the information exchange will leverage capabilities and knowledge, including developing cooperative test programs, to improve confidence in the results from the various concretes and research reactors.

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