Can We Design Concrete to Survive Nuclear Environments?

A summary of a panel discussion on the effects of nuclear environments on concrete and cementitious grouts

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Concrete is the most widely used man-made building material, engineered to withstand a wide variety of loads and environmental exposures. Concrete is the primary construction material for reactor containment and biological shielding structures, vital components of the fleet of nuclear reactors in service worldwide for power generation. Its use in nuclear applications, however, poses many challenges not found in most infrastructure applications.

Current Status of Concrete Nuclear Infrastructure

Per the International Atomic Energy Agency, as of February 2017, there are 449 operating nuclear reactors in 30 countries, and 60 new nuclear power plants are under construction in 15 countries.[1] Containment structures are designed to survive both short-term, high-intensity hazards (for example, earthquakes, explosions, impact), while also enduring years of exposure to elevated temperatures. Biological shielding structures are subject to long-term radiation exposure from the reactor within. Radiation and elevated temperature exposure are now known to alter the mechanical properties of the concrete and can cause a loss of crystallinity of aggregate-forming minerals; this can lead to a process called radiation-induced volumetric expansion (RIVE) and may cause otherwise nonreactive aggregates to become alkali-silica reactive.[2-5] Potential impacts from alkali-silica reaction (ASR), although not induced by temperature or radiation, also have come under exceptional scrutiny in the case of the Seabrook Nuclear Power Plant in Seabrook, NH, currently undergoing the license renewal process.

There are also existing challenges related to the storage and disposal of various radioactive waste materials from over 70 years of military, research, medical, mining, and power generation applications on every continent. Concrete and cementitious grouts are commonly used to provide shielding and encapsulation of these waste materials. Some waste isotopes and their decay products will pose a significant radiation hazard for up to hundreds of thousands of years, which demands extremely durable storage and disposal methods. Much more detailed proof of the long-term performance of concrete is needed on the timescales required for nuclear waste storage, which are longer than modern humans have been in existence. The potential effects of radiation exposure add to the technical challenges faced in designing concretes and grouts for these applications.

In the face of such challenges, it is only natural to ask: Can we design concrete to survive nuclear environments?

An international panel of experts debated this question during the 123 Forum session at The ACI Concrete Convention and Exposition – Fall 2016 in Philadelphia, PA, on October 24, 2016. Eric Giannini and Tengfei Fu organized and moderated the session. The panelists included Kim Kurtis, Georgia Institute of Technology; Yunping Xi, University of Colorado; Michał Glinicki, Institute of Fundamental Technological Research, Polish Academy of Sciences; and John Provis, University of Sheffield, UK. All four panelists and one of the moderators have been actively involved in research efforts to improve the understanding of the effects of nuclear environments on concrete and cementitious grouts, and the ability of engineers to monitor degradation of nuclear structures. The objective of this article is to disseminate the ideas presented and discussed by the panel to ACI members.

License Renewal of Nuclear Reactors—Scope and Challenges

The 99 operational reactors in the United States at the end of 2016 produced more than 30% of the world's nuclear-generated electricity and contributed about 20% of the total U.S. energy portfolio.[6,7] Reliance on nuclear power in the U.S. grew steadily from the 1970s until the late 1990s, when the nuclear energy output in the United States leveled off. The

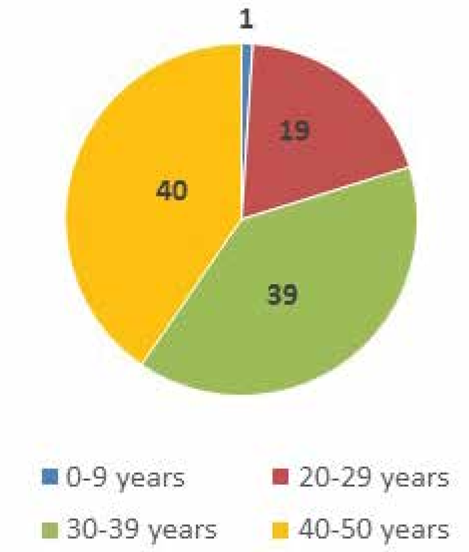

Fig. 1: Age of existing U.S. nuclear power reactors at the end of 2016. Data obtained from U.S. NRC[9] and adjusted for decommissioning of Fort Calhoun reactor

Fig. 2: Construction of full-scale mockup (left), previously constructed mockup (back, right) and specimen casting for materials testing during recent investigation of delamination of the concrete containment structure in Florida's Crystal River Nuclear Power Plant, led by Georgia Institute of Technology, with funding from the Electrical Power Research Institute (EPRI) *(photo courtesy of Kimberly E. Kurtis)*

2005 U.S. Energy Policy Act[8] was meant to reinvigorate the U.S. nuclear power sector, but concerns following 2011's Fukushima earthquake, in particular, have dampened those expectations. Today, with just two nuclear reactors under construction, much of the U.S. nuclear infrastructure is aging.

In fact, this can be viewed as a critical time in determining the future of the U.S. nuclear infrastructure. New facilities are typically granted an initial 40-year operating license and can apply for an additional 20-year license through an initial license renewal (LR) and potentially a second license renewal (SLR), toward a long-term operation (LTO) period of 80 years. As shown in Fig. 1, about 40% of existing nuclear reactors are more than 40 years old and 40% are between 30 and 39 years old—meaning that well over half of the active U.S. nuclear power infrastructure is at or beyond the LR stage. In fact, at least 84 plants are operating under renewed licenses, some of which were granted long before the expiration of the initial license.[9] Some plants will be deemed too uneconomical to operate beyond their initial license (for example, Fort Calhoun and Vermont Yankee[10,11]) and some, such as the decommissioned Crystal River, FL, facility (Fig. 2), may be too damaged to repair in anticipation of SLR.[12,13] However, for the majority of aging nuclear facilities, there is a growing need to understand the type, scope, and progression of aging-related degradation.

A recent review, published in a Georgia Tech doctoral thesis, identifies the predominant modes of degradation that have been reported in concrete nuclear structures.[12] These include many of the "usual suspects" associated with long-term aging and damage in more ordinary concrete applications[14-16]:

- Drying shrinkage as a common cause of concrete cracking;
- Limited instances of excessive stress relaxation in tendons in post-tensioned structures[15,17,18];
- Isolated instances of tendon corrosion and four cases of localized steel liner corrosion due to embedded foreign objects[15,19];
- Freezing-and-thawing damage, leading to spalling[20]; and
- ASR identified in at least one facility.

Other nuclear infrastructure durability issues are related to the specific service conditions and massive scale associated with these applications. This includes the effect of chronic high-temperature exposure—150 to 200°F (66 to 93°C)—which is often combined with stress and creep effects. In addition, because concrete structures are effective in shielding and containment, in the event of radiation exposure above certain levels, compressive and tensile strength and stiffness may be reduced.[3,4] Also, as previously noted, swelling and amorphization of siliceous aggregates have also been reported.[4] Finally, during routine maintenance operations, it may be necessary to untension and retension containment structures to gain interior access; the possibility of unbalanced loads during these operations also has the potential to influence concrete performance.

Several key issues emerge when considering concrete durability for SLR. First, given the criticality of the safety and containment function of concrete containments, there is no room for failure or even public perception of the possibility of failure in those structures. Thus, imperative needs exist for quantifiable and accurate measures of concrete condition assessment. But, obtaining this understanding is an important challenge. For example, it may not be possible to obtain cores or other samples through destructive means. Also, inspection and nondestructive evaluation may be limited by access to a single side of a massive element. Further, conditions under which measurements are to be made may be harsh. Yet, accurate data on damage state and rate of damage accumulation are vital for assessing the need and practicality of repair and, toward LTO, for modeling remaining service life. Finally, enhancements in service-life models that appropriately consider reliability and risk remain a priority for the U.S. Nuclear Regulatory Commission.[21]

Aging of Concrete in Nuclear Environments

As discussed earlier, there are many aging mechanisms of reinforced concrete structures, such as creep and shrinkage, freezing and thawing, ASR, and corrosion of reinforcing steel. These aging mechanisms are caused by variations of environmental parameters and mechanical loadings. For example, drying shrinkage is caused by the loss of moisture and thermal expansion is a result of temperature fluctuations. For concrete biological shielding structures used in nuclear power plants, neutron and gamma irradiation from nuclear reactors must be added to the list of influential environmental parameters. An important common feature of many aging mechanisms is volumetric mismatch among the components of concrete, which can cause internal damage in the material such as generation of voids and development of cracks. For example, neutron irradiation can cause expansion of commonly used aggregates, while the loss of moisture caused by the heat of irradiation can cause drying shrinkage of cement paste surrounding the aggregates. This volumetric mismatch generates cracks in cement paste that will cause long-term deterioration of concrete.[2-5]

The coupling effects among the environmental parameters and mechanical loading on the volumetric mismatch were not studied systematically prior to the construction of nearly all nuclear power plants in operation today. However, research on these topics has benefited not only our understanding of concrete for nuclear power plants but also the understanding of creep and durability properties of concrete in a more general structural engineering context. To develop durable concrete for long-term operation of nuclear power plants, these coupling effects must be taken into account.

The environmental parameters and mechanical loading include temperature T, moisture (humidity) H, nuclear irradiation N (neutron particles and gamma rays), and mechanical loading P. The effect of P is called basic creep, which is the long-term deformation of concrete under the condition of $\Delta T = 0$, $\Delta H = 0$, and P = constant; the effect of ΔH is drying shrinkage, which is the long-term deformation of concrete under the condition of $\Delta T = 0$, $\Delta H \neq 0$, and $P = 0$; and the effect of ΔT is thermal expansion, which is the deformation of concrete under the condition of $\Delta T \neq 0$, $\Delta H = 0$, and $P = 0$. Each of the long-term deformations has been studied extensively under the influence of a single parameter, but the long-term deformation under simultaneous actions of more than one parameter has not been understood very well. For example, the long-term deformation due to a simultaneous action of P and ΔH is drying creep, which is larger than the sum of the basic creep and the drying shrinkage. The extra deformation is caused by the coupling effect of P and ΔH, called stress-induced shrinkage, or the Pickett effect.[22] Similarly, the long-term deformation due to a simultaneous action of P and ΔT is thermal creep, which is larger than the sum of the basic creep and the thermal expansion. The extra deformation is caused by the coupling effect of ΔT and P, called stress-induced thermal expansion. The long-term deformation due to a simultaneous action of P, ΔT, and ΔH is called load-induced thermal strain (LITS), which has been studied by only a few researchers (Fig. 3).[23]

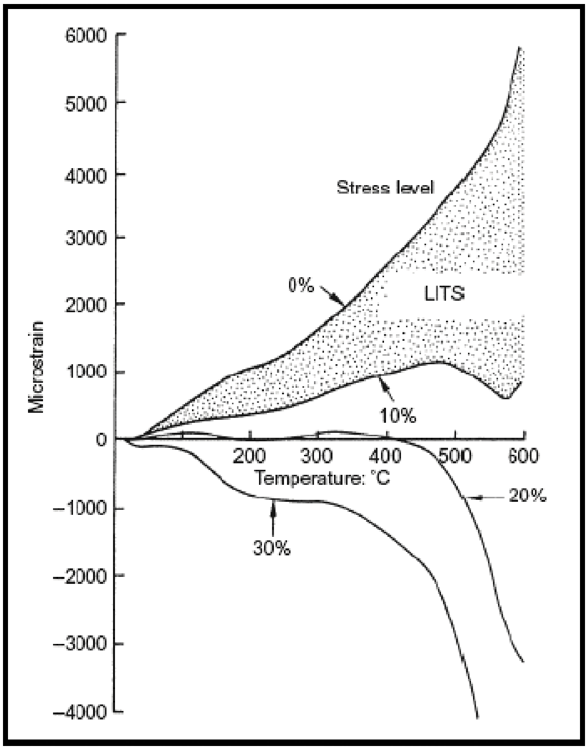

Fig. 3: Strains of unsealed basalt concrete under loading and heating (LITS = transient creep + basic creep + elastic strains. Transient creep is transitional thermal creep and drying creep in case of unsealed specimen)[23]

The effects of nuclear irradiation on properties of concrete in nuclear structures are more complicated because they are coupled with the thermal, mechanical, and moisture loads, which can all vary depending on the reactor design. These coupling effects are not well understood and thus need to be better studied experimentally. The outcomes from this research will need to be implemented in the form of improved design methods for nuclear concrete structures.[5,24]

Durability Design for Containment Structures

Nuclear containment structures are integrally designed with the various systems and components they support and protect to restrict the spread of radiation and radioactive contamination to the general public. The durability design is largely based on a suitable choice of concrete constituents that is guided by combined action of radiation, thermal and mechanical loadings, moisture, and chemical environmental

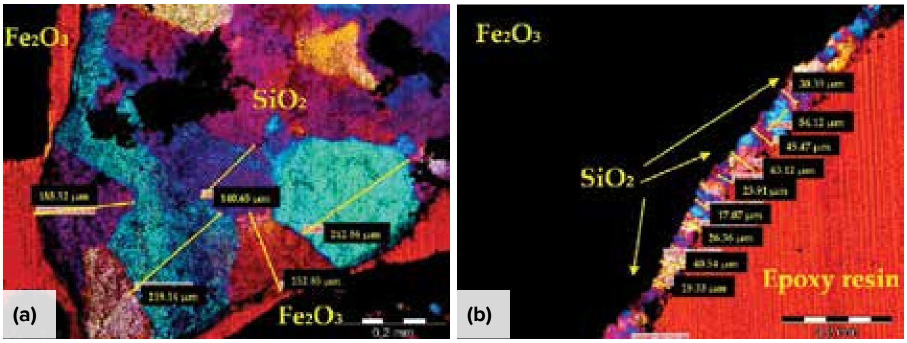

Fig. 4: Identification of quartz size in hematite aggregate on thin section in XPL with λ plate: (a) innocuous quartz, >130 μm; and (b) reactive quartz, 10 to 60 μm (from Reference 30)

factors. Concrete mixture designs need to be adjusted for the technology requirements arising from large-volume placements, pumping issues, and increased formwork pressure of heavyweight mixtures for radiation shielding.

For the long-term performance of containment structures, the protection of steel reinforcement against corrosion is of primary importance. The relevant long-term deterioration mechanisms of concrete include chemical attack by carbon dioxide (CO_2), aggressive sulfate exposure, chloride ions, or borated water as is the case in spent fuel pools.[25] Because most concrete containment structures are massive, limits are imposed on the temperature development in hardening concrete to prevent early-age cracking. The use of supplementary cementitious materials is proposed both for a decreased risk of thermal cracking during concrete hardening and for an increased chemical resistance of hardened concrete.[26] To counteract cracking that might occur as a result of microcrack accumulation and growth at later ages, deleterious expansive phenomena such as ASR and delayed ettringite formation (DEF) are carefully considered. Finally, the concrete mixture for the containment structure is also designed for very low permeability to both liquid and gas.

Considering the exceptional social significance of containment structures and the exceptional service demands, improved tools for materials selection are sought.[27-29] Relevant examples include a procedure for identification of effective size of quartz grains in heavyweight polymineral aggregates.[30] A recently developed digital image analysis technique on thin sections was found to be effective for evaluating the size of quartz grains that contrast with a black iron oxide phase in hematite aggregates (Fig. 4). ASR tests on such aggregates revealed large expansion of specimens correlated with a considerable content of reactive quartz phase within the hematite aggregate.

Aggregate gradation and grain shape have been found to have a significant influence on the air permeability of concrete, particularly in a case of special aggregates for radiation shielding that often come from soft rocks and contain flaky or irregular grains. However, to account for major effects of the internal concrete humidity on the air permeability, an improved tool for characterization of concrete at the designed relative humidity has recently been developed.[31] Enhanced prediction of the temperature field in hardening radiation-shielding concrete is facilitated with more sophisticated characterization of thermal properties of hardening concrete considered as functions of maturity. The numerical solution of the inverse heat transfer problem allows for determination of effective thermal conductivity, heat capacity, and the heat source function.[32] The tool developed has been used to support selection of low-heat portland and blended cements. A drawback of using very slow-hardening cements in large concrete placements is related to the increased formwork pressure of heavyweight concrete mixtures until substantial hardening takes place. Because the lateral pressure increases with increasing fluidity of mixture, the challenge is to control it while maintaining the proper filling capacity of densely reinforced elements.

New tools for critical evaluation of Hilsdorf data[3] on the effects of prolonged irradiation of concrete are under development, particularly within the works of the International Committee on Irradiated Concrete (ICIC).[5] These tools will be important for both evaluating license renewal applications and for more sophisticated materials selection for long-term performance of newly constructed nuclear power plants.

Advances in Dealing with Radioactive Waste—Important Processes and Improvements in Wasteform Grouts

In many parts of the world, low- and intermediate-level radioactive waste streams, which may be derived from the nuclear fuel cycle or from many other areas of society including medical processes, are treated through incorporation into cementitious grouts, mortars, or concretes.[33] The diverse types of waste that are generated in the nuclear fuel cycle necessitate the use of a wide variety of processes, wasteforms, and grout types to render them safe for storage (which may be for a period of up to 100 years) and/or eventual disposal (for example, in a deep geological repository).[34] However, in many nations, the grouts that are in common use involve addition of slag cement (ground-granulated blast-furnace slag [GGBFS]) to a portland-cement-based binder system to provide the necessary performance in terms of very high reproducibility of material performance, controllable setting, relatively low heat release during hydration, low permeability, and chemical (including redox) binding of key radionuclides. The radionuclides of interest and importance in low and intermediate level wastes are extremely diverse in concentration and half-life, but much attention is often paid to ^{137}Cs and ^{90}Sr as dose-limiting and potentially mobile species that must be effectively immobilized, at least in the medium term (decades to centuries). These nuclides each have half-lives of approximately 30 years, and are important fission products of uranium and plutonium.

There are many potential modes of evolution or damage that may influence the ability of cementitious wasteforms to retain immobilized radionuclides. Many of these are common to other applications of cement and concrete in civil and infrastructure applications, but with the additional factor of potential irradiation damage as the immobilized radionuclides decay. Wasteform materials used for disposal must also be designed to serve for a period of hundreds of thousands of years, far exceeding the timescales on which cement and concrete performance are considered in most engineering contexts. In wasteform cements, unlike in reactor environments as discussed earlier, neutron doses and radiogenic thermal effects tend to be relatively less damaging than gamma (and sometimes alpha) irradiation and autogenous heating due to cement hydration in the wasteforms themselves, or in the backfill that is placed around them after emplacement in a repository. The cements are usually designed so that autogenous heating does not drive the temperature up to the point where water could boil and cause cracking or other hazards such as radionuclide release, but radiolytic effects are significant in defining the design and use of wasteform grouts.

Gamma radiolysis within portland cement hydrates and the associated pore solution is a complex process, where radiolysis of the water present in the pores of the cement is a significant issue that must be addressed.[35] Alpha irradiation is less widely studied, but can also cause hydrogen generation through radiolytic pathways.[36] Radiolysis of water can yield hydrogen peroxide (H_2O_2) and gaseous hydrogen (H_2). The hydrogen, if not correctly controlled (for example, through venting of containers), can generate hazardous conditions. Hydrogen peroxide and other radical species can react with the cement by oxidizing some of the hydrous binder components—for example, with portlandite $Ca(OH)_2$ to produce calcium peroxide. The addition of GGBFS to the cement consumes some of this portlandite and generates a chemically reducing environment, thus controlling the oxidation reactions by providing sulfides that can be oxidized to sulfates. This can create additional ettringite within the cement,[37,38] but this has been observed to be beneficial in resisting cracking and enabling strength to be retained during irradiation with simultaneous heating. Radiolysis or chemical degradation of organic or carbonaceous material within cementitious wasteforms can also lead to gas generation (for example, release of CO_2 or methane), which raises the need for detailed control and understanding of organic inventories in wasteform design, processing, and handling.

However, due to the wide variety of important radionuclides that potentially are to be immobilized or disposed in cementitious matrices, as mentioned earlier, it is not always optimal (or desirable) to use portland cement as the basis of the binder matrix. Recent innovations in wasteform cement design include the use of geopolymer-based, alkali-activated magnesium phosphate (for example, "ceramicrete"), magnesium silicate hydrate, calcium aluminate, or calcium sulfoaluminate binders, which provide opportunities to effectively immobilize waste components such as reactive metals, contaminated oils, and radionuclides, which, for various reasons, cannot be effectively bound in portland cement-based matrixes. Each of these cement types has its advantages and disadvantages,[39] but some of these cements are now seeing full-scale use in the industry, particularly those based on geopolymer and alkali-activation technology.[40] There is both a clear need and an evident opportunity for innovation in cement science to lead to higher-performing, more reliable cements in these applications, which will remain important in service for many millennia to come.

Summary and Future Research Needs

Concrete is an indispensable material for the construction of structures for nuclear power generation, and the encapsulation of waste materials from nuclear power plants and many other applications. The aging global fleet of commercial nuclear power plants is facing significant challenges, highlighted by the U.S. experience with the license renewal process:

- Long-term aging effects on concrete nuclear structures have come into focus. In addition to sources of degradation commonly affecting long-serving concrete infrastructure, these structures must deal with coupled effects of mechanical loading, temperature, and moisture as a result of their service environment, known as LITS; the combined effects of these three loads are greater than the sum of the effects of each individual parameter and greatly in need of further study;
- The ability of engineers to assess the condition of concrete in nuclear power structures, and quantify distress and risk, is complicated by the limits on access to the structures and the inhospitable operating environment. These assessment data on damage state and state of progression are vital information for nuclear power plants undergoing SLR;
- Recent advances in materials characterization, selection, and mixture design for the specific application of impermeable and durable reactor containment structures will inform the design of concrete mixtures that are more resistant to the thermal and irradiation exposure expected in service; and
- The need to immobilize and dispose of radioactive wastes for extremely long time periods presents additional challenges, requiring cementitious wasteform materials capable of resisting the chemical degradation under long-term exposure to gamma and alpha radiation, and binding potentially mobile waste materials. Recent innovation in binder systems other than slag-modified portland cement systems may be able to deliver the performance required of wasteforms.

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Received and reviewed under Institute publication policies.

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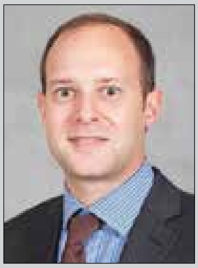

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