

Determining the Effects of Radiation on Aging Concrete Structures of Nuclear Reactors - 10243

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ABSTRACT

The U.S. Department of Energy Office of Environmental Management (DOE-EM) is responsible for the Decontamination and Decommissioning (D&D) of nuclear facilities throughout the DOE Complex. Some of these facilities will be completely dismantled, while others will be partially dismantled and the remaining structure will be stabilized with cementitious fill materials. The latter is a process known as In-Situ Decommissioning (ISD). The ISD decision process requires a detailed understanding of the existing facility conditions, and operational history. System information and material properties are needed for aged nuclear facilities. This literature review investigated the properties of aged concrete structures affected by radiation. In particular, this review addresses the Savannah River Site (SRS) isotope production nuclear reactors. The concrete in the reactors at SRS was not seriously damaged by the levels of radiation exposure. Loss of composite compressive strength was the most common effect of radiation induced damage documented at nuclear power plants.

INTRODUCTION

The U.S. Department of Energy (DOE) focuses on managing the nuclear waste generated at the different nuclear complexes that were used for the production of nuclear weapons; this involves the Decontamination and Decommissioning (D&D) of nuclear facilities (i.e., nuclear reactors, chemical separation processing plants, etc.). These nuclear facilities are no longer needed. Performing D&D is quite a complex task, as careful measures need to be taken for personnel protection due to the high levels of radiation in parts of the facilities. The D&D program involves several approaches such as decontamination and reuse, complete dismantlement, and partial dismantlement and entombment (e.g., In-situ Decommissioning [ISD]). Therefore, it is crucial to understand the properties of the structures after being exposed to many deteriorating factors (e.g., environmental, irradiation, chemical attack, corrosion of steel, etc.). This information will support current ISD operations and predictions of the performance of the ISD closure.

Nuclear facilities have proven to be one of the biggest challenges in terms of environmental cleanup for engineers at different DOE sites. Among these facilities, the highest priority targets for D&D are the nuclear reactors facilities and the chemical separation processing plants (e.g. canyons). Many studies have been conducted over the years on the deterioration of concrete in nuclear facilities; however, it is not very clear what the effects are on the concrete over time after being exposed to radiation. To address this data gap, a literature review was conducted to investigate the properties of aging concrete in nuclear facilities exposed to radiation. Results of this review provide initial information to compare the potential effects of

radiation exposure on concrete at the SRS nuclear reactor facilities. This literature review is intended to be the first action for facility specific structural evaluation needed to support ISD.

METHODOLOGY

The first part of this literature review documented the radiation levels and types of concrete used in the SRS reactor facilities. Comparisons with commercial nuclear reactor facilities were made in order to investigate and estimate the radiation levels experienced by the concrete as well as the components or types of concrete present in the facility. The second part of this study documents the reviews of radiation levels that damage concrete and the radiation levels experienced by the concrete in SRS reactor facilities.

SRS REACTOR FACILITIES

SRS Heavy Water Reactor

The Heavy Water Reactor at SRS were design to operate at less than 500 MW thermal, but they were later increased to 2500 MW from 1955 to 1965 in order to increase production [1]. The areas of highest radiation exposure at the SRS reactor facilities are the containment structure (biological shield (BS)) around the reactor vessel (RV) and the disassembly basins. At these locations, the contamination levels are high due to elevated radiation levels in the reactor vessel and contamination in the disassembly basin. A general depiction of the SRS Heavy Water Reactor is shown in Figure 1.

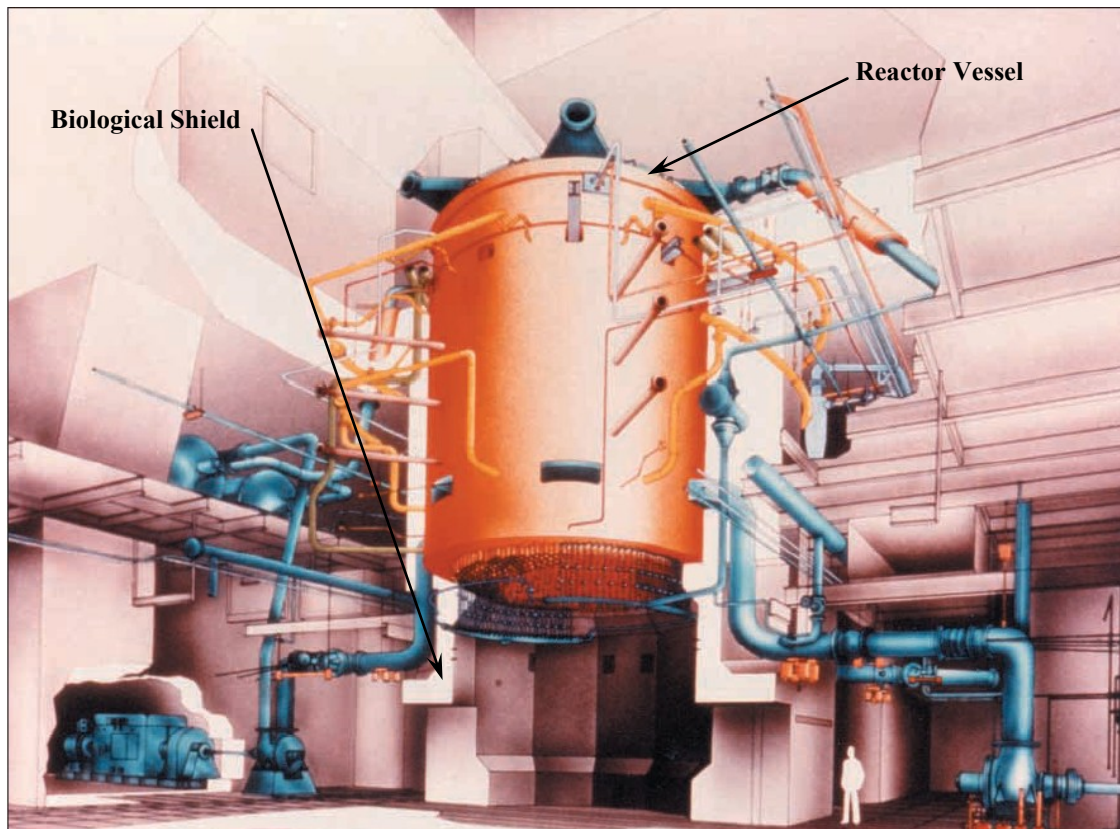


Figure 1. SRS Heavy Water Reactor [2].

Disassembly Basins

Contrasting the biological shield, the disassembly basins in the nuclear facility at SRS are contaminated with different levels of radiation. As mentioned before, the radiation levels of the basin are lower than those of the experienced by the biological shield. For instance, the R-Disassembly Basin at SRS contains radiation levels of $< 100\text{mR/hr}$ in the Vertical Tube Storage (VTS) with hot spots of 75 and 134 R/hr in the Dry Cave Section (DCS) and Machine Basin (MB) areas, respectively [3]. At the same time, there is water in the basin which is contaminated with ~ 1.8 curies of Cs-137 and Tritium concentrations of $38 \times 10^3 \text{ pCi/mL}$ [4]. These radiation values are not of the same magnitude of the ones the biological shield exposed to. The R-Disassembly Basin general configuration is shown in Figure 2.

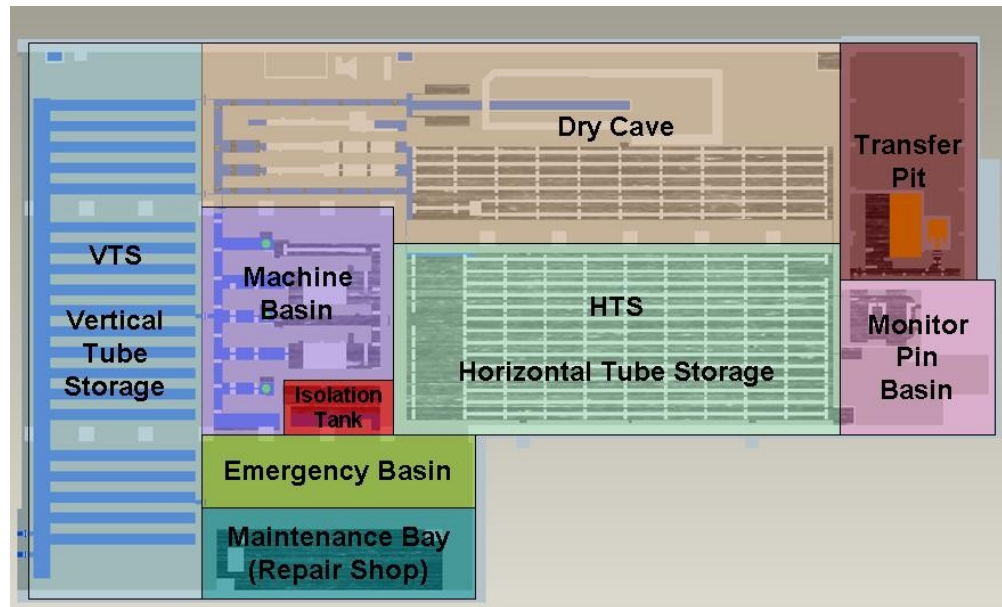


Figure 2. SRS R-Disassembly Basin [2]

CONCRETE COMPONENTS

Biological Shield

The biological shield of the reactor vessel is where the most contamination is found at a nuclear reactor facility. The biological shield consists of the primary reactor shield and the secondary reactor shield, where the primary reactor shield encloses the reactor vessel and the secondary reactor shield surrounds the latter including the main coolant loop [5]. The primary reactor shield was usually built of concrete with a thick secondary reactor shield varying between three feet to six feet [5, 6]. Meanwhile, the secondary reactor shield was four to five feet thick [5]. Figure 1 shows the general SRS Heavy Water Reactor configuration as described above.

According to studies conducted at the Oak Ridge National Laboratory Graphite Reactor, the primary reactor shield was built using barite (to increase the concrete density and lower wall thickness secondary reactor shield), haymite (to increase concrete mix water content), water, bituminous coatings, and Portland cement [7]. The graphite reactor was one of the first reactors built for the production of nuclear weapons; therefore, they served as a reference for the construction of the heavy water reactors found at SRS. High-density or heavyweight concrete for the primary reactor shield was a common practice due to

its high resistance to radiation. Based on U.S. Patent No. 2,726,339 [8], knowledge of the usage of high-density concrete as well as metal aggregates was known before the construction of the nuclear reactor facilities at SRS. Furthermore, the usage of Type II Portland cement was very common in the construction of the biological shield [5]. Admixtures (e.g., Plastiment and Intrusion Aid) were also used to enhance the flow of the dense concrete [9]. For the construction of the secondary reactor shield, ordinary concrete was used [7].

Based on the information above, it was assumed that the construction of the biological shield in nuclear reactor facilities at SRS involved some if not all of the components presented above. Davis, H.S. [5] noted, the construction of the primary reactor shield in the N-Reactor at Hanford (Richland, Washington) was made out of several concretes, mortars, and grouts. The construction of nuclear facilities was also based on the availability of local aggregates. Thus, one of the main components that the primary reactor shield in nuclear reactor facilities at SRS is more than likely to have is barite. Since SRS is located in South Carolina (SC), the closest sources of high-density aggregates were located in Tennessee and Georgia with barite [10]. At the same time, gravel is abundant in SC and could have been used for the construction of the secondary reactor shield in the heavy water reactors of the SRS.

Disassembly Basins

Unlike the biological shield, the disassembly basins in the nuclear reactor facilities at SRS were made of reinforced concrete with varying thicknesses of 2.5 to 7 feet [11]. Epoxy coatings were also used to protect the reinforced concrete in the basin. According to Duncan et al. [11], Amercoat 33 (vinyl paint) was one type of coating used to prevent its direct contact of the concrete with the disassembly basin water.

EFFECTS OF RADIATION ON CONCRETE

Several studies conducted explain that major deterioration of concrete occurs after being exposed to high levels of radiation. Granata and Montagnin [12], reported that at neutron fluxes of the order 10^9 neutrons/cm² (at temperatures of 130°C) had minimal effects on Portland 730 and limestone aggregate (standard mortar) and Portland 730 cement and barite aggregate (BHT mortar); the gamma dose equivalent used during the experiment was measured to be 10^{11} roentgens (1roentgen \sim 1 rad). However, upon increasing the temperatures to 280°C, (commonly found in heavy water reactors, and neutron integrated flux up to 10^{20} neutrons/cm²), the concrete samples were heavily damaged [12]. A similar experiment was conducted by Elleuch et al. [13], using serpentine granulate and aluminate cement paste while maintaining a temperature of 200°C. The density of the concrete was 2.51, which was lower than that used in other high density concretes, especially that of barite. The samples were exposed to doses of 1.2×10^{19} to 1.2×10^{20} n/cm² fast flux, resulting in dehydration of the cement paste and decreasing bending strength by 50% [13]. It was concluded that fluxes ranging from 2×10^{19} to 10^{20} neutrons/cm² damage concrete; nevertheless secondary reactor shield, the effects are noticed after irradiation doses of 3×10^{19} neutron/cm² with the expansion of the material, shrinking of the cement paste, and micro-cracking [13].

Hilsdorf et al. identified that critical neutron and gamma irradiation levels causing concrete deterioration are in the order of 1×10^{25} neutrons/m² (1×10^{21} neutrons/cm²) and 10^{10} rad, respectively, resulting in the cracking of concrete [14]. Similarly, Morinaga noted that fast neutrons of the order of $\sim 5 \times 10^{19}$ neutrons/cm² causes expansion of aggregates and shrinkage of cement paste [15]. These irradiation levels have been used as reference in many studies involving the irradiation of concrete but are nowhere near the

values endured by a NR. Nevertheless, discrepancies exist as to whether the concrete is really affected in nuclear reactor facilities by radiation at all [16].

One of the most common concerns in the study of the effects of radiation in concrete involves discerning between temperature- and radiation- related deterioration. To date, it has been difficult to determine whether the deterioration of biological shield in a nuclear reactor facility comes from the radiation itself or the high temperatures in the reactor vessel. Yevick noted that temperatures of 150°F (~66°C) are the threshold in which the hydrated-water molecule bond begins breaking [17]. Sakr and EL-Hakim [30] studied the effects of temperature on heavyweight concrete; temperature of 100°C starts decreasing the strength of the concrete. Furthermore, if the temperature increases beyond 500°C the strength drops sharply [18]. Sakr and EL-Hakim [18] also noted that magnetite and silica fume have high residual compressive strength after high temperature exposures. Based on the information presented above, the high temperatures are very detrimental to concrete. However, the concrete in the biological shield suffered from the breaking of the hydrated-water molecule and decrease in strength as depicted above. El-Sayed Abdo and Amin [17] conducted a study that dealt with the temperatures caused by slow (thermal) neutrons of the order of 10^9 neutrons/cm² and their effects on concrete and found that the heat generation by thermal neutrons occurs in the first layer of the concrete (10 – 50cm) with values of 0.2 mW/ cm³ (rise of 1.7°C) and 6.6E-3 mW/ cm³ (rise of 0.125 °C), respectively. As denoted by the latter, the heat generated by the thermal neutrons is quite insignificant in comparison to the temperature generated by the reactor vessel as a whole.

As mentioned above, telling the difference between the effects of temperature and radiation in concrete is not relatively easy to discern when both are acting at the same time. Most of the studies presented in this literature review have irradiated the concrete in the presence of temperatures ranging from 100°C to 280°C. In an effort to differentiate between the two, Lowinska-Kluge and Piszora [19] noted the effects of radiation were evaluated in different cement pastes without taking temperature into account. The Lowinska-Kluge and Piszora experiment involved four different types of cement: 1. ordinary portland cement; 2. portland cement containing fly ash; 3. portland cement containing silica fume; and 4. portland cement containing granulated aggregate, irradiated with a Co-60 source. The irradiation doses varied from 0 to 1409 MGy (1.409×10^{11} rads—values are converted to rads to compare to radiation levels found at nuclear reactor facilities). The results obtained by Lowinska-Kluge and Piszora [19] showed that portland cement samples were completely destroyed at radiation levels of 836 MGy (8.36×10^{10} rads). Cement samples containing fly ash, silica fume, and granulated aggregate showed progressive decomposition. The beginning signs of deterioration were at 130 MGy (1.3×10^{10} rads) in portland cement, 290 MGy (2.90×10^{10} rads) in silica fume and fly ash cements, and 466 MGy (4.66×10^{10} rads) in granulated aggregate cement; showing decomposition of the cement matrix, hydrates, clincker phase reflects separation of chemically bonded water, and primary reactor shield eudomorphoses and densification [19]. Most of the studies presented above, have irradiated the concrete with high levels of radiation. However, it needs to be understood that these values are at or above the publish values for concrete damage. Suggestions have been made to keep a limited flux of 1×10^{17} neutrons/cm² level during the life-time of the nuclear reactor facility to prevent the deterioration by radiation [14]. Sha and Hookhan [20] identified that these high radiation values are not experienced by the biological shield of the light water reactors; thus, having no detrimental effects.

Another important study conducted by Ichikawa and Kimura [15] evaluating the effects of radiation on concrete showed radiation-induced alkali-silica reactions (ASRs) do not take place during the lifetime of the nuclear reactor facility as long as ASR-tolerant aggregates are used; according to them, lower levels of radiation (e.g., 1×10^8 rads and 1×10^{16} neutrons/cm²) induce ASRs of aggregates containing silica-rich aggregates, such as plagioclase, as one of the major minerals. Nonetheless, the ASRs do not affect the soundness of the concrete structure [15]. Clifton [16] points out that it is unlikely that structures will collapse due to ASRs.

Most of the studies about the deterioration of concrete due to radiation assume full power phase of the reactor. According to Stacho et al. [21], radiation energies emitted by the reactor vessel are of up to 2 MeV for downtime phase and up to 10 MeV for full power phase. Thus, the irradiation on the concrete is five times less when the reactor is not operating. Fillmore [22] noted that the effects of low doses of radiation, i.e., $<10^{10}$ neutron/cm² or 10^{10} rad gamma dose, over a period of 50 years are negligible. Several studies have shown that the deterioration of concrete at nuclear reactor facilities involves chemical attack, leaching, radiation, temperature, cement-aggregate reactions, fatigue, freezing and thawing, abrasion and erosion, etc. [14, 23]. Taking this information into account, it can be concluded that overtime the major cause of deterioration for the concrete is not attributed to radiation alone. For instance, Basyigit, et al. [24] observed that freezing and thawing affects the attenuation properties of high-density concrete such as barite. This in turn makes the concrete vulnerable to radiation, and the radiation makes the concrete vulnerable to the other types of deterioration. In other words, radiation and the other types of deterioration factors help one another deteriorate concrete over time at nuclear reactor facilities, but with the effects of radiation being insignificant.

One important study relating to radiation of concrete was conducted at the Oak Ridge National Laboratory Graphite Reactor. Blosser et al. [7] investigated the properties of the primary reactor shield used for the graphite reactor after being exposed to radiation for 12 years. The shield at this reactor was made out typical aggregates, barites-haydite concrete, ordinary concrete, and bituminous coating [7]. Blosser's findings showed that radiation had no change on the chemical properties and density of the concrete used; nonetheless, the compressive strength was decreased by $\sim 40\%$ on the first layer (closest to the reactor vessel) made out of one foot of ordinary concrete. Similarly, another study conducted relating the Temelin Pressurized Water Reactor (Czech Republic) yield a 10% decrease in the compressive strength of the concrete, the porosity was reported to have decreased by one-half, along with calcite formation, and signs of brittleness [25]. In this study, the concrete was exposed to radiation levels of 5×10^5 Gy (5×10^7 rads)—57 years of normal operation; clearly, more than the operation of the SRS reactors facilities. SRS reactor facilities have operated less than 40 years [1]. The concrete components used were siliceous gravel and CEM I 42,5R Mokra [25].

DISCUSSION

The operation time of nuclear reactor facilities plays a major role in the deterioration of concrete by radiation. The longer the operation time, the more radiation the concrete is subjected to. This literature review shows that the levels of radiation within the lifetime of the SRS nuclear reactor facilities were lower than threshold radiation values depicted earlier. The operation time of the SRS nuclear reactor facilities did not reach the 40 year mark.

The biological shield of SRS nuclear reactor facilities were made of various radiation resistant concretes or high density concrete (e.g., barites, haymite, etc.); thus, providing better resistance to radiation deterioration than ordinary concrete as seen in the study conducted at the Oak Ridge National Laboratory Graphite Reactor. Most of the studies presented above show different kinds of concrete mixtures irradiated at or beyond threshold values, having significant impact on concrete, especially ordinary concrete. Nonetheless, these radiation level values were not present in the lifetime of the SRS nuclear reactor facilities.

The degradation of the existing reinforced concrete present at the disassembly basins of the SRS nuclear reactor facilities can be attributed to other deterioration factors other than radiation. According to J.B. Pickett [3], significant amounts of calcium and magnesium were found to be present in the disassembly basin water. Leaching of the concrete is a possible reason discussed by Pickett [3]. Water has been in the

basin for years and it seems to have had penetrated the coating of the concrete causing this leaching. Furthermore, various kinds of debris are present in the basin water; with time its presence seems to have damaged the concrete coating allowing direct contact of the concrete with the water. Duncan et al. [11] pointed out that the only visible signs of deterioration found at the L-Area Basins, are from cracks do to shrinkage of the cement paste and water migration through the wall—they concluded that the degradation is insignificant to the concrete. Meanwhile, there is no evidence that the concrete present at the basins have been affected by radiation. According to Vodak et al. [25], calcite is a by-product of irradiation in concrete; however, a high level of radiation (i.e., radiation levels of 5×10^7 rads) is needed for a significant reaction. Still, this is not the case at the disassembly basins since the radiation levels are low and therefore not significant enough to cause major damage on the concrete structure as mentioned earlier.

CONCLUSIONS

This literature review documents the effects of radiation in concrete similar to the nuclear reactor facilities found at SRS. This literature review supports the current In-Situ Decommissioning operational prediction of performance of the nuclear reactor facility closure.

The results of this study concluded that the effects of radiation in the concrete of the biological shield are not detrimental since the radiation level interacting are below the threshold values (i.e., orders of $\sim 10^{25}$ neutrons/cm² for neutrons and $\sim 10^{10}$ rads for gamma-rays). Based on the different studies presented, radiation values at or beyond these threshold values can damage the concrete severely, causing expansion of aggregates, shrinking of cement paste, brittleness, micro-cracking, dehydration of cement paste, and decrease in bending strength. However, the radiation levels found in many of these studies are beyond those found in nuclear reactor facilities.

One common sign of deterioration that was noticed to occur within the lifetime of a nuclear reactor facility involved the decrease of compressive strength of the concrete. As proven by Blosser studies [7], compressive strength of the primary reactor shield in the Oak Ridge National Laboratory-Graphite Reactor was decreased within the first layers (i.e., within one foot of ordinary concrete). Meanwhile, Basyigit et al. [24], a more recent study conducted concluding that the compressive strength of concrete is decreased by radiation.

The disassembly basins were also taken into account in this literature review, as there are significant levels of gamma-rays radiation in them, but not as strong as the levels found in the biological shield. Based on the information presented above, it was concluded that the levels of radiation present at the disassembly basins have insignificant effects on the existing reinforced concrete.

In conclusion, concrete found in the heavy water reactor facilities at SRS is not significantly affected by the radiation levels coming from the reactor vessel or the disassembly basins.

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ACKNOWLEDGEMENTS

This paper was prepared in conjunction with work accomplished at the Savannah River National Laboratory, Savannah River Nuclear Solutions, LLC under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy, and Florida International University-Applied Research Center Science & Technology Workforce Development Program under Contract No. DE-FG01-05EW07033 with the U.S. Department of Energy Office of Environmental Management.

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