Radiation-Induced Degradation of Concrete in NPPs

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ABSTRACT

Nuclear power plant life extensions to 60 and potentially 80 years of operation have renewed interest in long-term material degradation. One material being considered is concrete. Swelling of aggregates driven by radiation induced displacements of atoms is currently considered to be the most probable leading contributor to the radiation induced degradation of concrete mechanical properties. In the biological shields of nuclear plants atom displacements are dominated by neutron contributions while gamma-ray contributions are negligible. For several minerals, which are common constituents of aggregates in concrete, it was shown that ~95% of the dpa is generated by neutrons with energies above 0.1 MeV. Neutrons with energies above 1 MeV contribute only ~20 to 25% to the dpa. Therefore, if neutron fluence is used as correlation parameter for the concrete degradation, the 0.1 MeV neutron energy cut-off should be used for the fluence. Based on the projected neutron fluence values (E > 0.1 MeV) in the concrete biological shields of the US pressurized water reactor fleet and the available data on radiation effects on concrete, some decrease in mechanical properties of concrete cannot be ruled out during extended operation beyond 60 years.

KEYWORDS: Concrete, degradation, atom displacements, neutron fluence, gamma-rays, rive

Introduction

In the USA more than 80 nuclear power plants (NPPs) were approved to extend their licenses to operate up to 60 years. Further extensions, to 80 years and beyond, are being considered to meet future national energy needs while reducing greenhouse gas emissions. NPPs lifetime extensions have revived interest in long-term aging of materials in general and recently in concrete. Large structures of NPPs are built from concrete, including safety-related structures such as the biological shields and containment buildings. Recent reviews of existing data on irradiated concrete revealed considerable gaps in knowledge base [1]. A research program on concrete aging and degradation processes in NPPs was established within the Light Water Reactor Sustainability (LWRS) program under the US Department of Energy (DOE) and is being

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performed in consultation with the Electric Power Research Institute's Long-Term Operations (LTO) Program and the US Nuclear Regulatory Commission (NRC). In addition, an International Committee on Irradiated Concrete (ICIC) has been created to facilitate information exchange and research coordination at an international level.

We reported on the initial results of the current studies at the 15th ISRD [2]. This paper will outline progress in the irradiated concrete research program with the focus on the assessment of the radiation environment in concrete biological shields in NPPs.

Previous work

Radiation-induced degradation of concrete has been historically correlated to neutron fluence and gamma-ray dose. Most of the legacy data on mechanical performance of irradiated concrete were analyzed by Hilsdorf et al. [3], (1978) who concluded that, for some concretes, neutron fluence of 1.0×10^{19} n/cm^2 may cause a reduction in compressive and tensile strength and a marked increase in volume. Our recent review of the data on irradiated concrete supports Hilsdorf's conclusion [4]; however, it was also observed that the lack of detailed information on several important parameters such as energy cut-off of the reported neutron fluences, temperature of concrete during irradiation, and many variables associated with the aggregates and concrete mix and preparation prevents clear separation of effects and makes reliable predictions of concrete degradation quite challenging.

Radiation transport calculations were performed for two selected pressurized water reactors (PWRs) to assess the radiation fields in the concrete biological shields (CBS). It was found that both plants remain below the 1.0×10^{19} n/cm^2 fluence value for the 1 MeV cut-off even for the 80 years of operation. However, both plants exceed the 1.0×10^{19} n/cm^2 fluence level during 40 years of operation for the 0.1 MeV cut-off, and during first 20 years if the integral fluence ($E > 0$ MeV) is used. Using the US Nuclear Regulatory Commission ADAMS database [5] and guidance obtained from the above-mentioned transport calculations, Esselmann and coworkers estimated the neutron fluence values in the CBS, at 80 years of operation, for the current US NPP fleet. It was found that the PWRs have higher expected neutron fluence values than the boiling water reactors. The effects of the PWR plant design were also clearly observed: four loop PWR reactors show significantly lower fluence values than the majority of both two- and three- loop plants. All PWRs are expected to exceed the 1.0×10^{19} n/cm^2 ($E > 0.1$ MeV) threshold before 80 years of operation, and the highest expected fluences will be in the 6-7 $\times10^{19}$ n/cm^2 ($E > 0.1$MeV) range [6].

Currently, radiation induced volumetric expansion (RIVE) of aggregates is considered to be the most probable leading contributor to the degradation of concrete mechanical properties, including compressive and tensile strength [7]. Swelling of aggregates, driven by radiation-induced displacements of atoms, and consequent loss of long-range order and amorphization, induces strains between the cement paste and aggregates. The strains cause micro cracking of cement paste resulting in damage and loss of mechanical properties of concrete [4,8]. For this reason, we investigated radiation induced atom displacements in aggregates.

Neutron induced atom displacements in aggregates

The displacement per atom (*dpa)* cross-sections for neutrons for a few materials that are widespread rock-forming minerals in concrete aggregates were generated with Specomp computer code, which is available as RSICC PSR-263 along with the Specter code and its database [9]. Selected minerals were quartz (SiO_2), calcite ($CaCO_3$, dimorphous form of aragonite), feldspars: albite (sodium aluminum silicate, $NaAlSi_3O_8$), anorthite (calcium aluminum silicate, $CaAl_2Si_2O_8$), and microcline (potassium aluminum silicate, $KAlSi_3O_8$, dimorphous form of orthoclase), almandine ($Fe_3Al_2(SiO_4)_3$, from garnet mineral group), and fayalite (Fe_2SiO_4, from olivine mineral group). The atom displacement threshold energies (E_d) used were 31 eV for carbon, 30 eV for oxygen, 25 eV for sodium, 27 eV for aluminium, 25 eV for silicon, and 40 eV for potassium, calcium, and iron, which are the values that are usually used for these elements [9]. Using the same E_d for the elements in binary and more complex compounds is a common assumption because more detailed information is typically not available. The primary recoil atom energy distributions used were those available in the Specter database and were originally generated from ENDF/B-V neutron cross-sections data.

The neutron *dpa* cross-sections for the materials mentioned above are shown in Fig. 1. For comparison, the *dpa* cross-section for silicon, which was obtained from displacement kerma factors for silicon provided in ASTM E 722 [10], is also shown. The *dpa* cross-sections for all materials exhibit very similar variations with energy: from a minimum at around 100 eV the cross sections rapidly increase with increasing neutron energy. The increase is in part due to the increasing neutron energy and in part due to opening of additional reaction channels. At low neutron energies, the cross sections increase with increasing energy reflecting the typical *1/v* increase of the neutron absorption cross sections (*v* is neutron velocity and is proportional to $E^{1/2}$).

The total displacement per atom rate is obtained as:

$$\frac{dpa}{s} = \int_0^{E_{max}} \sigma_{dpa}(E) \times \phi(E) dE \tag{1}$$

The E_{max} denotes the maximum energy of the neutron spectrum, $\sigma_{dpa}(E)$ is the energy-dependant *dpa* cross-section, and $\phi(E)$ is the neutron fluence rate spectrum.

To evaluate the relative contributions of neutrons from different energies to the total *dpa* rate it is useful to calculate the percentage of atom displacements that are induced by the neutrons with energy above E_0, for a given neutron fluence rate spectrum:

$$\frac{\frac{dpa}{s}(E > E_0)}{\frac{dpa}{s}} = \frac{\int_{E_0}^{E_{max}} \sigma_{dpa}(E) \times \phi(E) dE}{\int_0^{E_{max}} \sigma_{dpa}(E) \times \phi(E) dE} \times 100 \tag{2}$$

The relative contributions to the *dpa* rate as defined with Eq. 2 were calculated with the neutron fluence rate spectrum in the cavity of a two-loop PWR and three-loop PWR, obtained from previous analyses [2]. The results are shown in Fig. 2 and Fig. 3. The curves of the relative contributions to the *dpa* rate are remarkably similar for all materials considered, and are also very similar for the two-loop and three-loop PWR. The results are also shown for silicon, a material not directly relevant for concrete, but are very similar to those materials found in concrete aggregates. Figures 2 and 3 show that approximately 90% of the atom displacements are caused by the neutrons with energies between 0.1 MeV and 2 MeV while neutrons with energies above 1 MeV cause only about 20 % to 25 % of the total atom displacements. However, neutrons with energies above 0.1 MeV contribute more than 95% of all atom displacements. These statements hold for all materials considered and for the neutron spectrum in the cavity of a two-loop and three-loop PWR. This clearly indicates that, from the perspective of creation of atom displacements in aggregates, neutron fluence with $E > 0.1$ MeV is a more relevant correlation parameter than the fluence with $E > 1$MeV.

The dpa rates for the selected material are listed in Table 1, together with the projected dpa for 80 years of operation. It can be seen that there is relatively little variation – only about 25% – in dpa rate between the minerals that were considered. Another observation is that the dpa rates for the two-loop plant are about 2.6 times higher compared to the three-loop plant.

TABLE 1— Dpa rates and dpa for selected materials, for the neutron spectrum in the cavity of a two-loop and three-loop PWR.

PWR Type	**Aggregate**	***Dpa* Rate (s^{-1})**	***Dpa****
2 Loop	Quartz (SiO_2)	2.39E-11	0.055
	Calcite ($CaCO_3$)	1.90E-11	0.044
	Silicon (Si)	2.40E-11	0.056
	Anorthite ($CaAl_2Si_2O_8$)	2.22E-11	0.052
	Microcline ($KAlSi_3O_8$)	2.22E-11	0.052
	Albite ($NaAlSi_3O_8$)	2.40E-11	0.056
	Almandine ($Fe_3Al_2(SiO_4)_3$)	2.10E-11	0.049
	Fayalite (Fe_2SiO_4)	1.88E-11	0.044
3 Loop	Quartz (SiO_2)	9.15E-12	0.021
	Calcite ($CaCO_3$)	7.29E-12	0.017
	Silicon (Si)	9.04E-12	0.021
	Anorthite ($CaAl_2Si_2O_8$)	8.51E-12	0.020
	Microcline ($KAlSi_3O_8$)	8.51E-12	0.020
	Albite ($NaAlSi_3O_8$)	9.20E-12	0.021
	Almandine ($Fe_3Al_2(SiO_4)_3$)	8.03E-12	0.019
	Fayalite (Fe_2SiO_4)	7.15E-12	0.017

**Dpa* is calculated for 80 years of operation with 92% capacity factor.

Gamma-ray induced atom displacements

Gamma-rays do not cause atom displacements directly; they first interact with electrons via Compton scattering, photoelectric effect, or pair production; the energetic electrons then displace atoms via electron-nucleus collisions. The dpa cross sections for gamma-rays are not as well developed as the dpa cross section for neutrons. No cross sections for compounds (minerals) were available and in fact Kwon's paper was the only readily available source of the dpa cross-sections for gamma-rays for several elements [11]. Kwon's paper provides cross-sections for C, Si, Cr, Fe, Ni, Mo, Ag, Au, and U. For this work C, Si, and Fe were selected because they are components of many minerals, while other heavier elements were not considered because they are typically not present in the aggregates in concrete. The gamma-ray dpa cross-sections for C, Si, and Fe are shown in Fig 1 along with the neutron dpa cross sections for minerals. The gamma-ray dpa cross sections have a threshold of ~0.2 MeV, increase with increasing energy, and are several orders of magnitude smaller than the neutron dpa cross sections at the same energy. The gamma-ray dpa cross sections also show the expected tendency to increase from the elements with lower atomic numbers to heavier elements.

For the quantitative comparison of the neutron and gamma-ray induced dpa rates in the actual radiation environment in the NPP biological shield, the cross sections were folded with the neutron and gamma-ray fluxes obtained in a previous analysis of a two-loop and three-loop PWR [2]. The results are presented in Table 2. Neutron induced dpa rates are from ~ 600 times to about 6,000 times higher than gamma-ray induced dpa rates.

It should be noted that the comparison of the neutron- and gamma-ray – induced dpa rates was done only for three elements C, Si, and Fe and was not extended to the minerals; therefore, it is far from comprehensive. However, other elements which are common in minerals should have similar cross-sections [11], and the neutron to gamma-ray dpa rate ratios for the minerals are expected to be similar to the values listed in Table 2.

TABLE 2 —***Calculated neutron and gamma-ray induced dpa rates for selected elements calculated with the neutron and gamma-ray fluxes in the cavity of a two-loop and three-loop PWR.***

		***Dpa* Rate**		**Ratio**
PWR type	**Material**	**Neutron**	**Gamma-ray**	**Neutron/Gamma**
		(s^{-1})	**(s^{-1})**	
2-loop	**C**	1.80E-11	2.88E-15	6261
	Si	2.40E-11	6.65E-15	3609
	Fe	9.20E-12	1.19E-14	773
3-loop	**C**	6.90E-12	1.37E-15	5036
	Si	9.04E-12	3.24E-15	2790
	Fe	3.36E-12	5.29E-15	634

Another aspect that needs to be considered is that the neutron and gamma-ray fields change significantly within the pressure vessel, cavity, and the biological shield of the PWR, both in the spectrum and in relative intensity of neutron and gamma-ray fluxes. Therefore, it is necessary to investigate the variation of neutron and gamma-ray induced dpa rates as a function of location. An example is shown in Fig. 4, which depicts neutron and gamma-ray induced dpa rates in silicon versus radial distance from the core vertical axis. Regardless of variations, the neutron induced dpa rate is at least two orders of magnitude higher than the gamma-ray induced dpa rate at all locations considered. Therefore, it appears reasonable to conclude that in radiation fields in the biological shields of PWR the gamma-ray contribution to the dpa in the aggregates in concrete is not significant.

Absorbed dose

As mentioned before, radiation induced displacements of atoms in aggregates is currently considered to be the most probable leading contributor to the degradation of concrete. Therefore, we will just briefly discuss the neutron and gamma ray contributions to absorbed dose. The gamma-ray and neutron induced absorbed dose rates listed in Table 3 were calculated for the selected minerals and homogenized concrete, with the gamma-ray and neutron fluence rate spectra in the cavity of a two-loop PWR and three-loop PWR [2].

TABLE 3— ***Calculated gamma-ray and neutron induced absorbed dose rates and absorbed doses for selected materials in the cavity of a two-loop and three-loop PWR.***

		Gamma-rays		Neutrons		Absorbed Dose Rate Ratio
		Absorbed Dose Rate	Absorbed Dose*	Absorbed Dose Rate	Absorbed Dose*	
PWR Type	Material	Gy/s	MGy	Gy/s	MGy	Gamma-rays /Neutrons
2 Loop	Quartz (SiO_2)	4.48E-02	104.05	1.96E-02	45.4	2.29
	Calcite ($CaCO_3$)	4.65E-02	108.03	2.12E-02	49.1	2.19
	Anorthite ($CaAl_2Si_2O_8$)	4.54E-02	105.45	1.78E-02	41.3	2.55
	Microcline ($KAlSi_3O_8$)	4.53E-02	105.15	1.77E-02	41.1	2.56
	Albite ($NaAlSi_3O_8$)	4.44E-02	103.11	1.92E-02	44.6	2.31
	Concrete	4.55E-02	105.69	3.77E-02	87.5	1.21
3 Loop	Quartz (SiO_2)	2.11E-02	48.91	7.31E-03	17.0	2.89
	Calcite ($CaCO_3$)	2.18E-02	50.62	7.82E-03	18.1	2.79
	Anorthite ($CaAl_2Si_2O_8$)	2.13E-02	49.53	6.63E-03	15.4	3.21
	Microcline ($KAlSi_3O_8$)	2.13E-02	49.41	6.57E-03	15.2	3.24
	Albite ($NaAlSi_3O_8$)	2.09E-02	48.47	7.18E-03	16.7	2.91
	Concrete	2.14E-02	49.61	1.41E-02	32.7	1.52

*Absorbed dose is calculated for 80 years of operation with 92% capacity factor.

Compared with neutron-induced absorbed dose rates in the minerals, the gamma-ray-induced absorbed dose rates are higher: 2.2 to 2.6 times higher for the two-loop plant and 2.9 to 3.2 times higher for the three-loop plant. For concrete the gamma-ray dose rates are higher than the neutron-induced dose rates by ~ 20% for the two-loop plant and 50% for the three-loop plant. Investigation of spatial distribution reveals that the gamma-ray induced absorbed dose is higher than the neutron-induced absorbed dose throughout the biological shield and the difference increases from the inner wall towards the outer wall of the concrete shield. In the light of experimental evidence that irradiation with gamma-rays does not seem to cause degradation of concrete and minerals, this is likely not important for assessing concrete degradation directly. However, gamma rays also cause radiolysis of water contained in concrete, which may instigate drying and further changes in mechanical properties (such as creep). Contribution of gamma-rays to the heating also needs to be considered in the calculations of temperature profile through the biological shield.

Conclusions

Swelling of aggregates driven by radiation-induced displacements of atoms is currently considered to be the most probable leading contributor to the radiation induced degradation of concrete mechanical properties. In the biological shields of nuclear plants atom displacements are dominated by neutron contributions while gamma-ray contributions are negligible. For several minerals, which are common constituents of the aggregates in concrete, it was shown that ~95% of the dpa are generated by neutrons with energies above 0.1 MeV. Neutrons with energies above 1 MeV contribute only ~20 to 25% to the dpa. Therefore, if neutron fluence is used as a correlation parameter for concrete degradation, the 0.1 MeV energy cut-off should be used for the neutron fluence determination. A leading candidate for the unified irradiation parameter that could be used to correlate concrete degradation resulting from irradiation with neutrons, gamma-rays (and possibly electrons and ions; although this was not addressed in this paper) appears to be atom displacements in aggregates.

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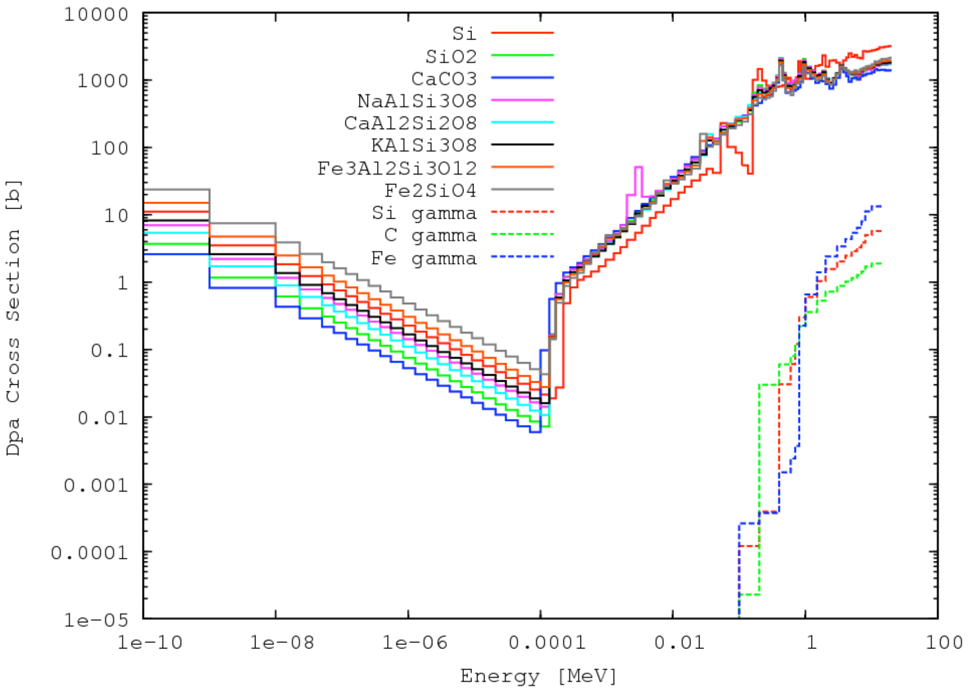
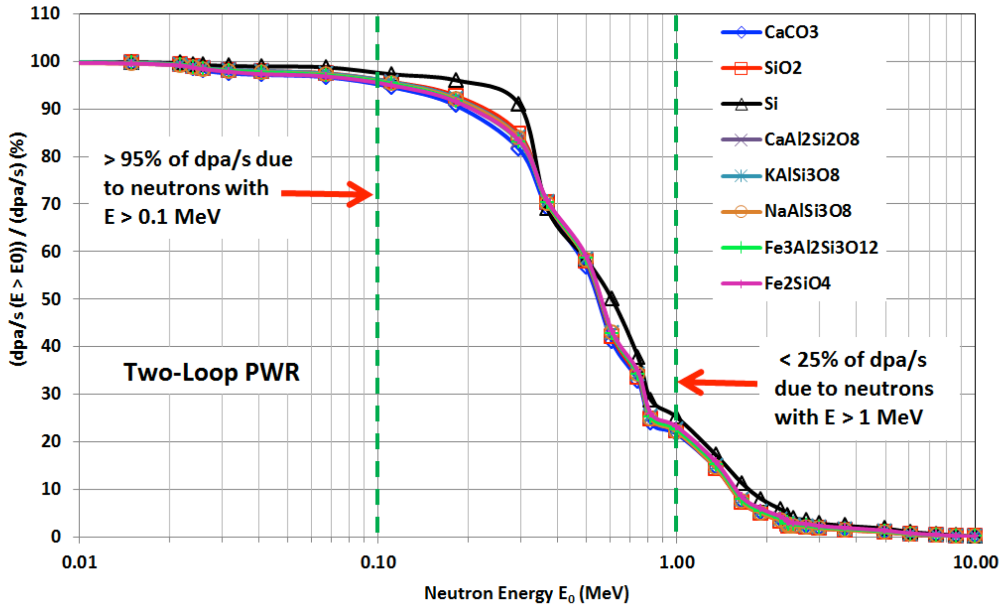


FIG. 1— *Dpa cross-sections for neutrons for selected minerals and gamma-ray*

dpa cross sections for silicon, carbon and iron.

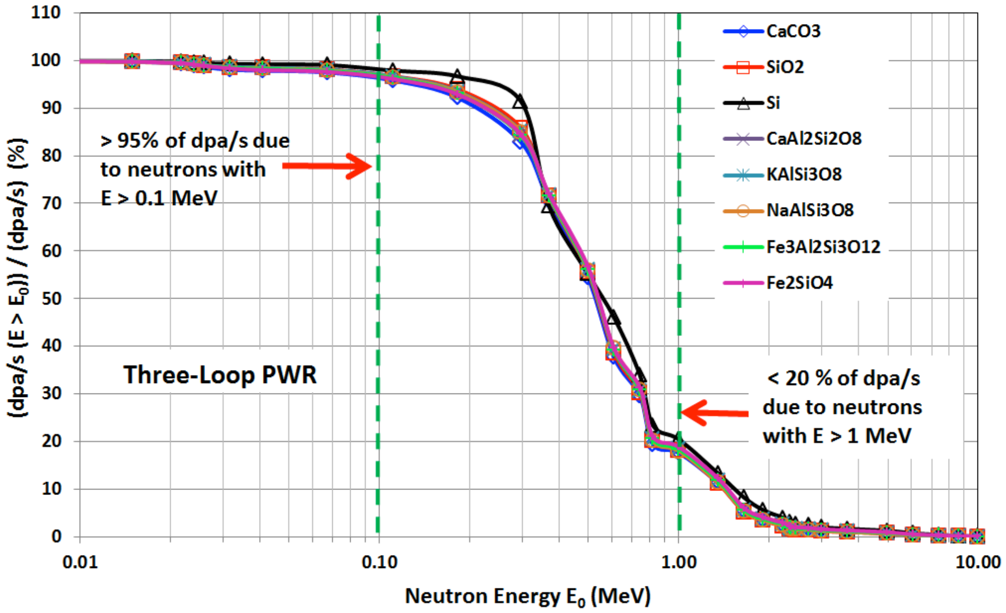


> 95% of dpa/s due to neutrons with E > 0.1 MeV

Two-Loop PWR

< 25% of dpa/s due to neutrons with E > 1 MeV

FIG. 2— 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.



> 95% of dpa/s due to neutrons with E > 0.1 MeV

Three-Loop PWR

< 20 % of dpa/s due to neutrons with E > 1 MeV

FIG. 3— *Relative contribution of neutrons with energies $E > E_0$ to the dpa rate for the neutron spectrum in the cavity of a three-loop PWR.*

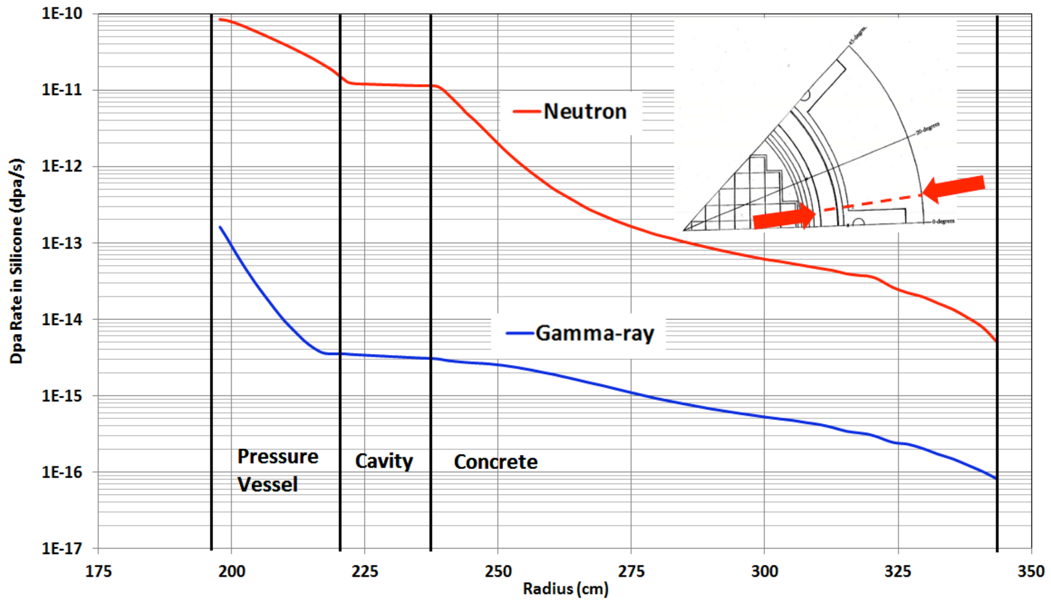


FIG. 4 *—Neutron and gamma-ray-induced dpa rates in silicon versus radial distance from the core vertical axis. The abscissa of the plot is the radial distance along the line marked with the arrows in the insert on the top right side and is in the reactor core mid-plane.*

References

[1] Graves, H., Le Pape, Y., Naus, D., Rashid, J., Saouma, V., Sheikh, A., and Wall, J., Expanded Material Degradation Assessment (EMDA), Vol. 4: Aging of Concrete, NUREG/CR-7153, ORNL/TM-2011/545. U.S. NRC, 2014.

[2] Remec, I., et al., Characterization of Radiation Fields in Biological Shields of NPPs for Assessing Concrete Degradation, EPJ Web of Conferences 106, 2016, 02002, http://dx.doi.org/10.1051/epjconf/201610602002

[3] Hilsdorf, H., Kropp, J., Koch, H., The effects of nuclear radiation on the mechanical properties of concrete, Special Publication of the American Concrete Institute 55, pp. 223–254, 1978.

[4] Field, K. G., Remec, I., Le Pape, Y., Radiation effects in concrete for nuclear power plants – Part I: Quantification of radiation exposure and radiation effects, *Nuclear Engineering and Design*, **Vol. 282**, pp. 126-143, ISSN 0029-5493, 2015.

[5] U.S. NRC, Agency-wide Documents Access and Management System (ADAMS), http://www.nrc.gov/reading-rm/adams.html, 2015.

[6] Esselman, T., et al., Expected Condition of Concrete at Age 80 Years of Reactor Operation, Technical Report, A13276-R-001, Lucius Pitkin, Inc., 2013.

[7] Rosseel, T. M., Maruyama, I., Le Pape, Y., Kontani, O., Giorla, A. B., Remec, I., Wall, J. J., Sircar, M., Andrade, C. & Ordonez, M. (2016). Review of the Current State of Knowledge on the Effects of Radiation on Concrete. *Journal of Advanced Concrete Technology*, *14*(7), 368-383.

[8] Giorla, A., Vaitová, M., Le Pape, Y., & Štemberk, P. (2015). Meso-scale modeling of irradiated concrete in test reactor. *Nuclear Engineering and Design*, *295*, 59-73.

[9] Greenwood, L. R., and Smither, R. K., "SPECTER: Neutron Damage Calculations for Materials Irradiations," ANL/FPP/TM-197 (January 1985); L. R. Greenwood, "Compound Displacement Damage

Calculations with SPECOMP," Informal Notes, Argonne National Laboratory (February 1988), SPECTER-ANL, PSR-263, Radiation Safety Information Computational Center.

[10] E722 Standard Practice for Characterizing Neutron Fluence Spectra in Terms of an Equivalent Monoenergetic Neutron Fluence for Radiation-Hardness Testing of Electronics, Annual Book of ASTM Standards, Volume 12.02.

[11] Kwon, J., and Motta, A. T., Gamma displacement cross-sections in various materials, *Annals of Nuclear Energy,* Vol. 27, 2000, pp. 1627–1642.