

Light Water Reactor Sustainability Program

Analysis of simultaneous thermal/gamma radiation aging of cross-linked polyethylene (XLPE) insulation—interim status report



June 2017

U.S. Department of Energy

Office of Nuclear Energy

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Leonard S. Fifield and Miguel Correa

June 2017

**Prepared for the
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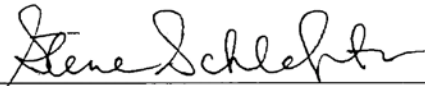
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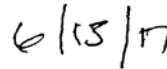
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Steve Schlahta
Nuclear Science Project Management Office Director



Date

Executive Summary

Electrical cables used for power, instrumentation and control functions within the containment building of a nuclear power plant can experience both elevated temperatures and gamma irradiation while in service. Since these cables may have several decades of service life, it is important to understand the aging and degradation of cable polymer insulation and jacketing materials to inform condition monitoring strategies and to guide cable repair and replacement decisions. Most historical studies of cable material longevity, including testing to environmentally qualify nuclear cables, investigated effects on material integrity of thermal stress followed by irradiation stress to simulate in plant service. However, in actual service cables may be exposed to both stresses simultaneously. This has led to questions as to how well observed effects from sequential exposure can predict effects from simultaneous exposure. That is, are the degradation mechanisms for cable polymer materials different in the two different scenarios?

This report describes results from our ongoing study of the effects of simultaneous thermal and gamma radiation exposure to cable materials. We focus here on cross-linked polyethylene (XLPE) insulation, the class of cable insulation most commonly found within containment in U.S. nuclear power plants. We exposed XLPE insulation samples from commercial nuclear cable to gamma total doses up to 32 kGy (32 Mrad) at dose rates from 116 to 540 Gy/h. These radiation exposures were performed while simultaneously heating the samples at 60, 90, or 115 °C. Cross-linking and scission of the polymer molecular chains in the XLPE insulation material with exposure was tracked using a solvent swelling method and the 'percent gel' and 'uptake factor' metrics. In this work, percent gel of the aged XLPE samples was seen to increase with exposure temperature and uptake factor was seen to decrease with exposure temperature. Evidence for inverse temperature effects, in which radiation-induced damage is higher at lower temperature, was not observed for the materials and conditions considered here. Characterization of the effects of similar exposure of XLPE samples at 25 °C that is currently underway will provide additional indication of evidence to support the understanding of the importance of inverse temperature effects for this material. This work directly addresses identified knowledge gaps in the aging and degradation of cable materials and cable systems related to simultaneous thermal and radiation aging including inverse temperature effects and dose rate effects.

Acknowledgements

The crosslinked polyethylene aging experiment described was planned in collaboration with the research group of Professor Nicola Bowler at the Iowa State University under the “Advanced Models for Nondestructive Evaluation of Aging Nuclear Power Plant Cables” Nuclear Energy University Programs (NEUP) project (PICS:NE Work Package # NU-14-IA-ISU_-0401-01).

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Acronyms

CSPE	chlorosulfonated polyethylene
EAB	elongation at break
EPR	ethylene-propylene rubber
HEF	High Exposure Facility
IEEE	Institute of Electrical and Electronics Engineers
LOCA	loss of coolant accident
PNNL	Pacific Northwest National Laboratory
QR	qualification report
REM	Radiological Exposures & Metrology Laboratory
SCRAPS	Sandia's Cable Repository of Aged Polymer Samples
XLPE	cross-linked polyethylene
XLPO	cross-linked polyolefin

Introduction

Nuclear power plants contain hundreds to thousands of miles of electrical cables. Many of these cables may have service lives of several decades. Therefore it is important to understand the natural degradation processes of polymeric cable insulation and jacketing materials to plan for condition monitoring and cable replacement or repair as needed. Elevated temperature is one of the most important stress factors to consider with regard to cable material aging and degradation. Nuclear plants also have a unique aging stress in the form of gamma radiation for cables located within containment.

According to plant construction permits granted between 1957 and 1978, as referenced in EPRI TR-103841, crosslinked polyethylene (XLPE) is the most common low-voltage electrical cable insulation material. It can be found inside containment in 90% of U.S. plants. Polyethylene is an example of a polyolefin (hydrocarbon chain formed from carbon molecules containing a carbon double bond, C=C), and the class of XLPE-type polymers are often more broadly referred to as crosslinked polyolefin (XPLO). The majority of the vintage XLPE cables found in nuclear plants today were manufactured by Rockbestos Company and are composed of XLPE formulation with tradename Firewall® III. Nuclear cables with insulation consisting of a variant of this same material under the same trade name is currently available for purchase from RSCC Wire & Cable LLC. Manufacturers of the second and third most commonly found XLPE-insulated cables are Brand-Rex and Raychem, respectively, although Rockbestos cables are found in more plants than the combined total of these other two companies.

Because XLPE cable insulation is commonly found in nuclear power plant containment, XLPE aging from thermal stress, irradiation stress, and the combination of the two is of particular interest for this material. Cables qualified for Class 1E service followed qualification guidelines established in IEEE Std. 323-1974 and IEEE Std. 383-1974 (once these standards were established). Qualification of Rockbestos cable constructions included exposure to thermal conditioning followed by irradiation conditioning to simulate service life before being subjected to simulated loss of coolant accident (LOCA) conditions and post LOCA testing (similar to QR-5805, 1987). Since qualification testing involved sequential exposure to elevated temperature and gamma radiation, the degradation of XLPE under simultaneous thermal and radiation exposure has been identified as in need of further investigation (NUREG/CR-7153, Vol.5).

In collaboration with Iowa State University, Pacific Northwest National Laboratory (PNNL) is investigating the effects of temperature, dose rate, total dose and exposure sequence on measurable properties of RSCC Firewall® III cable insulation. The goal of the work is to identify, within the limit of the factors explored, if differences can be observed between, for instance, cable insulation aged to a common total dose at different dose rates or aged with radiation at temperature versus aged with radiation at room temperature followed by thermal aging.

Simultaneous exposure of XLPE to heat and gamma radiation at three different temperatures, 60 °C, 90 °C, and 115 °C, and at gamma dose rates ranging from 116 to 540 Gy/h for exposure periods up to 25 d has already been completed. Exposure periods at the selected dose rates resulted in total doses from 70 to 32 kGy (7 to 32 Mrad). Sequential aging of XLPE at the same gamma dose rates and total doses at 25 °C followed by exposure at 115 °C for corresponding periods is currently underway. A comprehensive analysis of observable changes in the XLPE material aged at PNNL is ongoing at both Iowa State University and at PNNL. Herein initial percent gel and solvent uptake results are discussed in the context of knowledge gaps related to dose rate and temperature dependence in simultaneous thermal/gamma aging of XLPE. Comparison of simultaneous versus sequential aging and results from comprehensive material analysis will be forthcoming following completion of those efforts.

Materials

The XLPE insulation material investigated here was obtained from Firewall® III Instrumentation Cable with chlorosulfonated polyethylene (CSPE) jacket purchased from RSCC. The cable is product code I46-0021 and is described as multi-conductor shielded (XLPE/CSPE), 90 °C, 600V, Class 1E Nuclear. It has two 16 AWG, 7 strand tin-coated copper conductors. The insulation is 0.64 mm thick and each insulated conductor has a total diameter of 2.8 mm. One of the conductors has grey insulation and the other white. The white XLPE insulation was used for the studies described here.

The black CSPE jacket and aluminum/polyester tape shield were removed to expose the insulated conductors, seen in Figure 1. Tubular “straw” samples were prepared in 100 mm lengths by removal of conductor from the insulation. Samples of ~1 cm length and 0.025 g mass were cut from aged and unaged straws for use in the percent gel and solvent uptake analysis.

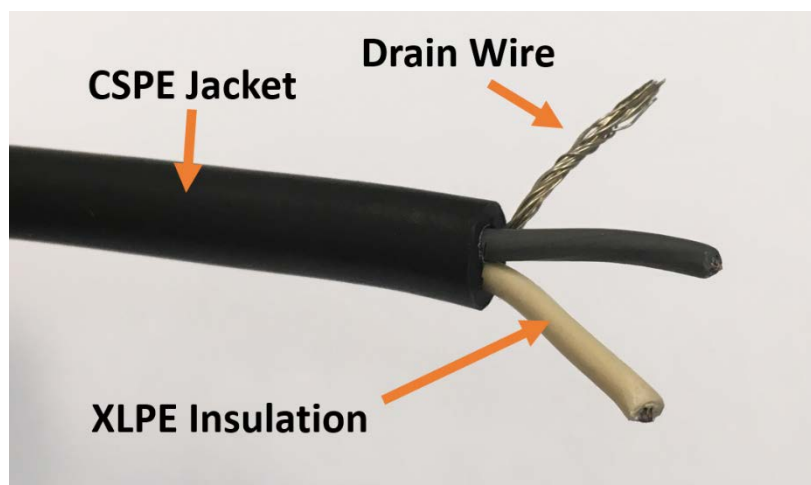


Figure 1. Photo of RSCC Firewall® III Instrumentation Cable (product code I46-0021).

Accelerated Aging

XLPE insulation straw samples were aged at three different temperatures, 60 °C, 90 °C, and 115 °C, in mechanical convection ovens with continuously circulating air. In addition to “thermal only” samples, samples were also aged in a mechanical convection oven placed in a gamma irradiation beam from a Co-60 source in the High Exposure Facility (HEF) of the Radiological Exposures & Metrology (REM) Laboratory at PNNL, as pictured in Figure 2. At each of the three temperatures, samples were exposed to gamma irradiation dose rates from 116 to 540 Gy/h for exposure times of 5 to 25 d. Depending on dose rate, over the maximum exposure of 25 d samples experienced total doses from 69600 Gy (6.96 Mrad) to 324000 Gy (32.4 Mrad).



Figure 2. Mechanical convection oven containing XLPE samples in HEF at PNNL. Co-60 source is positioned directly to the right of samples in the leaded column during irradiation.

Percent Gel & Solvent Uptake Method

The gel fraction (percent gel or “% gel”) of a sample is a relative measurement of the homogeneity of the XLPE polymer network [Celina 1995]. When crosslinking dominates aging in a polymer network, the gel fraction increases. The uptake factor of a sample indicates how easily a sample swells. When polymer chain scission dominates during aging, the uptake factor increases as solvents is absorbed more readily. The gel fraction and uptake factor therefore share an inverse relationship: as gel fraction increases, uptake factor decreases and vice versa.

XLPE samples were preconditioned overnight at room temperature (22 °C) and 44% relative humidity in a closed chamber containing a saturated solution of potassium carbonate prior to testing. The preconditioned samples of approximately 0.025 g were initially massed and loaded into a vial with xylenes (>99%) in a 200:1 solvent to XLPE mass ratio. The solution was heated to 110 °C and held at that temperature for 24 h using the set up pictured in Figure 3. While still hot, the sample was removed from solvent and loaded into an empty, tared vial to determine its solvent-swollen mass. The samples were then dried of solvent at 100 °C in vacuum for 24 h. This method is similar to that used for data reported in SAND2005-7331 except that the authors there used p-xylenes instead xylenes mixture and refluxed samples (p-xylene boiling point is 138.4 °C) rather than incubating them at 110 °C.



Figure 3. Hotplate and timer for sample incubation in percent gel and solvent uptake experiment.

The outputs of the process are sample weights: initial aged sample (W_a), sample swollen by the solvent (W_s), and samples after drying (W_d). Percent gel (% gel) is defined as ratio of dried weight to initial weight. Uptake factor is defined as the ratio of swollen weight to dried weight.

$$\% \text{ Gel} = \frac{W_d}{W_a} \times 100 \qquad \text{Uptake Factor} = \frac{W_s}{W_d}$$

Results

Percent gel was observed to increase both with aging temperature and with dose rate, as seen in the data from 15 d simultaneous exposure in Figure 4.

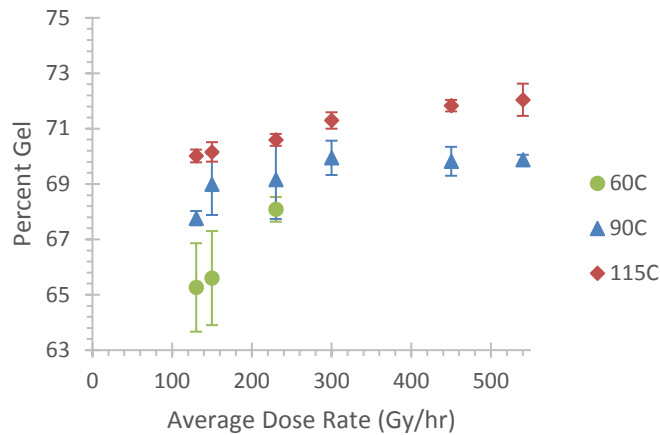


Figure 4. Plot of average dose rate (Gy/h) versus percent gel of sample. All samples were aged for 15 d, at either 60 °C, 90 °C, or 115 °C.

For aging at a given temperature, percent gel was observed to increase with both aging time and with dose rate, as observed in data from the 115 °C simultaneous exposure plotted in Figure 5.

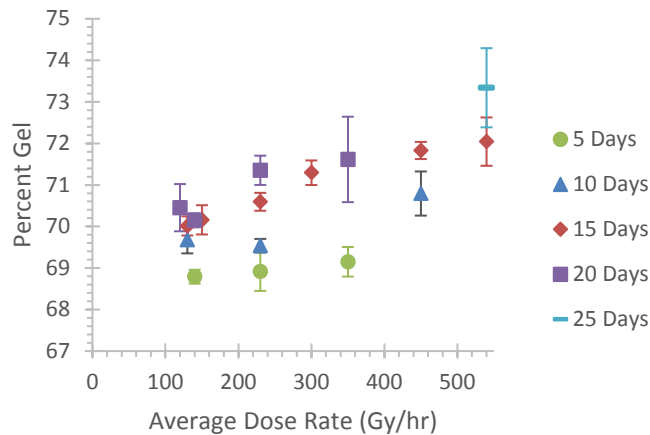


Figure 5. Plot of average dose rate (Gy/h) versus percent gel of sample. All samples were aged at 115 °C for either 5 d, 10 d, 15 d, 20 d or 25 d

Uptake factor decreased with temperature and dose rate, as observed for samples aged for 15 d shown in Figure 6.

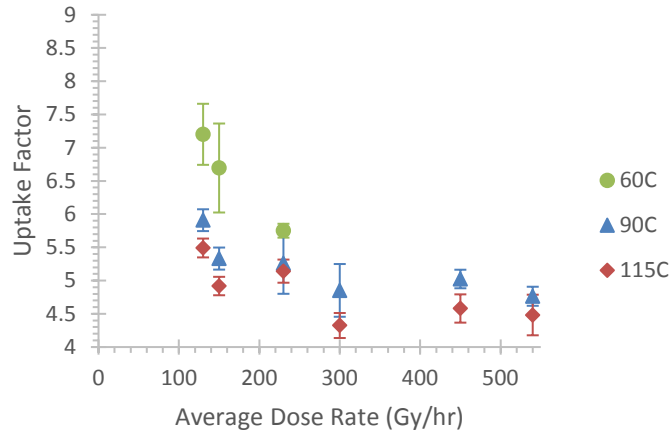


Figure 6. Plot of average dose rate (Gy/h) versus uptake factor of sample. All samples were aged for 15 d at either 60 °C, 90 °C, or 115 °C.

With some scatter in the data, uptake factor is also seen to decrease with exposure time and dose rate for samples simultaneously aged at 115 °C in Figure 7.

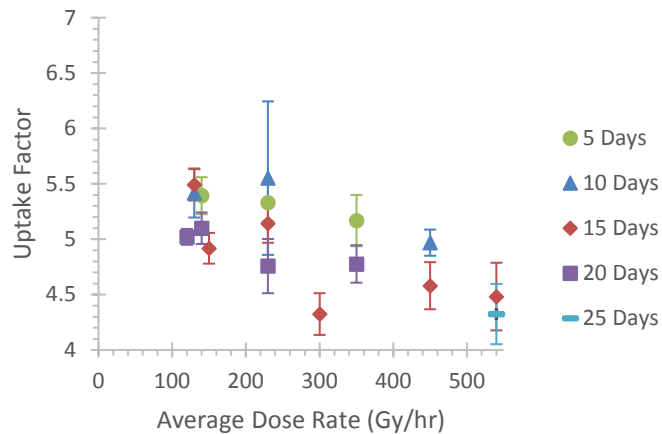


Figure 7. Plot of average dose rate (Gy/h) versus uptake factor of sample. All samples were aged at 115 °C for either 5 d, 10 d, 15 d, 20 d, or 25 d.

Discussion

Bernstein and coworkers report similar percent gel and uptake factor data for XLPE in Sandia’s Cable Repository of Aged Polymer Samples (SCRAPS) database (Addendum to SAND2005-7331). Green-colored XLPE insulation (XLPO-5) from a Rockbestos Firewall® III cable (C-16) (12 AWG 3/C Rockbestos Firewall® III 600V XHHW NEC Type TC Hypalon Jacket) was aged in air at 138 °C with no gamma exposure for up to 250 d. Percent gel and uptake factor were determined by refluxing in p-xylene. Tensile elongation at break (EAB) measurements correlated the percent gel and solvent uptake data to material lifetime curve (from unaged to <<50% EAB). The data plotted below in Figures 8 and 9 indicate

similar magnitude values for percent gel and uptake factor despite the slight differences in method material used, though it is clear that in our 25 d exposure series we are only accessing the early portion of the XLPE lifetime curve, plotted with EAB versus exposure time in Figure 10.

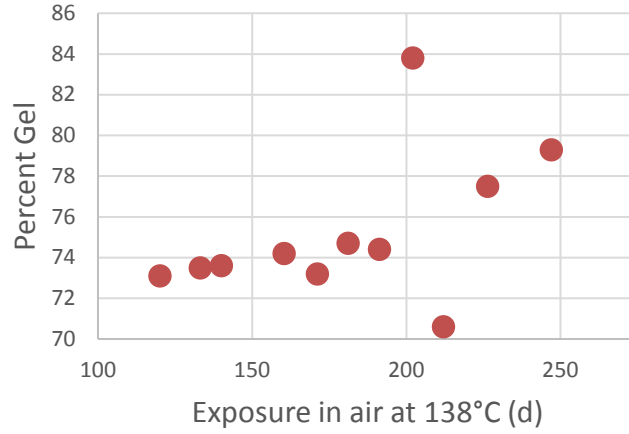


Figure 8. Plot of XLPE percent gel versus exposure time in air at 138 °C (data from SAND2005-7331).

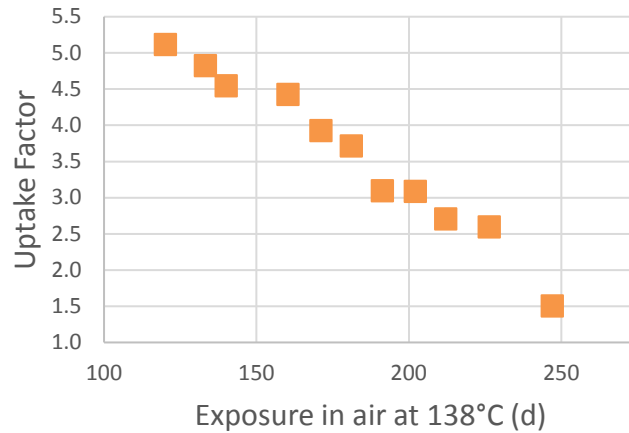


Figure 9. Plot of XLPE uptake factor versus exposure time in air at 138 °C (data from SAND2005-7331).

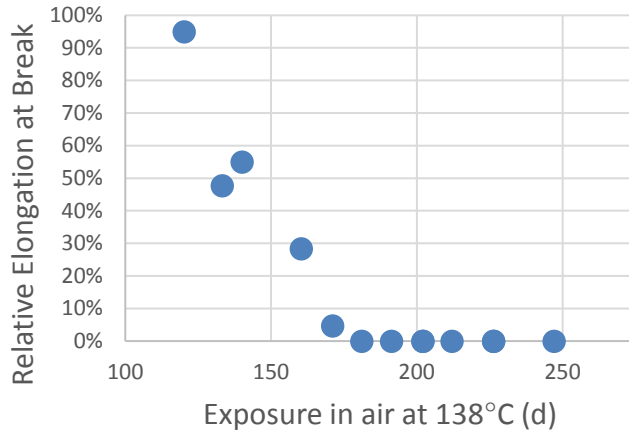


Figure 10. Plot of XLPE relative EAB (ratio of EAB to initial EAB) versus exposure time in air at 138 °C (data from SAND2005-7331).

One of topics highlighted for future studies in the NUREG/CR-7153, Vol. 5 is the so-called inverse temperature effect:

“The observed inverse temperature effect, where polymer degradation occurs more rapidly for constant dose rates as the combined environment temperature is lowered, represents an example in which material aging and lifetime prediction cannot be represented adequately by conventional approaches, such as the Arrhenius methodology.” [NUREG/CR-7153, Vol. 5]

As reported therein, this effect has been observed in both XLPO and ethylene-propylene rubber (EPR) materials. The fact that the effect was not observed in JNES Report SS-0903 is attributed to the fact that the combined radiation-thermal aging performed in that study was carried out at the elevated temperatures of 80-100°C. Evidence of inverse temperature effects are suggested to be more readily observed at combined aging temperatures closer to room temperature.

The inverse temperature effect is illustrated for an XLPO material by Celina et al [Celina 1996] in Figure 11 in which dose to equivalent damage (DED), as defined as total gamma dose required to reduce EAB from its initial value of 310% to the value of 100%, is plotted versus dose rate and temperature. At ~250 Gy/h, DED is seen to increase with exposure temperature from 22 °C to 41 °C to 60 °C. At exposures above 60 °C, however, DED decreases as temperature increases, from 60 °C to 80 °C to 100 °C to 120 °C.

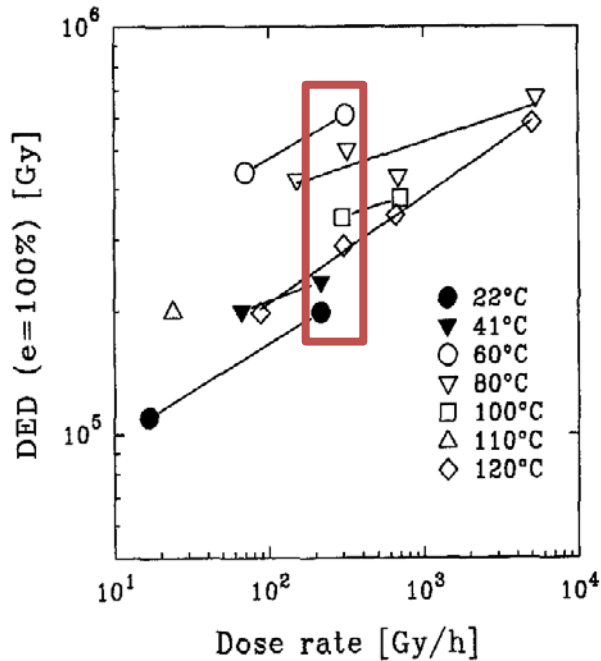


Figure 11. Plot of dose to equivalent damage (DED) at different temperatures and dose rates (figure from Celina 1996). Highlighted are the DED data points at ~250 Gy/h versus aging temperature.

Percent gel and uptake factor for XLPE simultaneously exposed at PNNL to 227 Gy/h gamma dose at 60, 90 and 115 °C for up to 20 days are displayed in Figures 12 and 13.

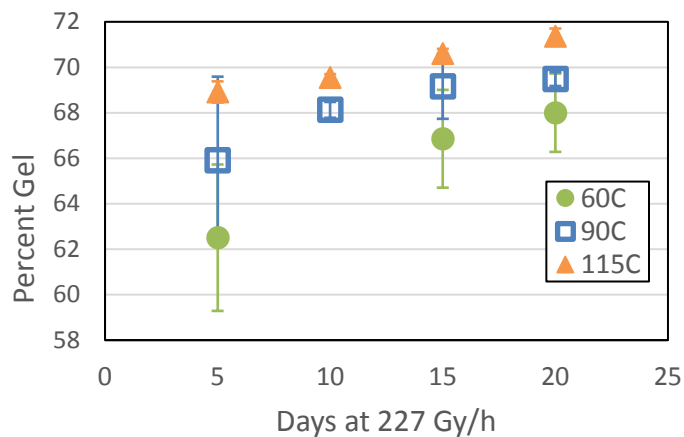


Figure 12. Plot of percent gel versus exposure time in days for XLPE exposed to 227 Gy/h gamma irradiation at 60, 90 and 115 °C.

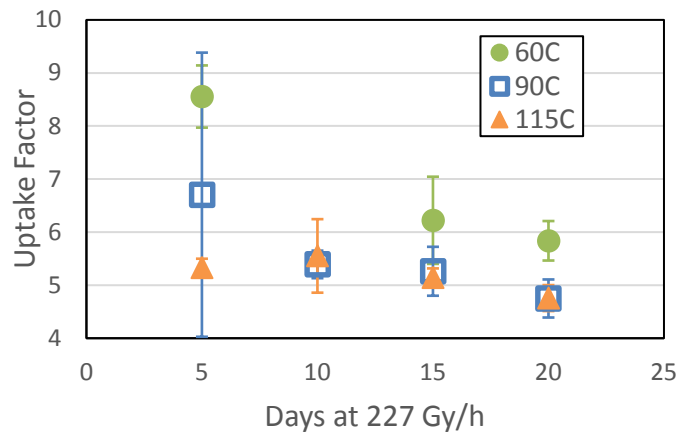


Figure 13. Plot of uptake factor versus exposure time in days for XLPE exposed to 227 Gy/h gamma irradiation at 60, 90 and 115 °C.

These plots indicate that in our experiments percent gel increased and uptake factor decreased with increasing temperature. In this limited data set, XLPE degradation at a constant gamma dose rate was not observed to be more severe at 60°C than at 90°C or 115°C. Aging of identical XLPE samples at identical gamma dose rates is currently underway at 25°C. Percent gel and uptake factor values from these samples will provide more information on this trend. Similarly, characterization data from a comprehensive set of additional techniques as applied to the series of samples aged at different temperatures, doses and dose rates will enable a closer determination of the presence of signs of inverse temperature effects in this example of XLPE insulation from Firewall® III XLPE cable.

Conclusions

Electrical cables used for power, instrumentation and control functions within the containment building of a nuclear power plant can experience both elevated temperatures and gamma irradiation while in service. Since these cables may have several decades of service life, it is important to understand the aging and degradation of cable polymer insulation and jacketing materials to inform condition monitoring strategies and to guide cable repair and replacement decisions. Most historical studies of cable material longevity, including testing to environmentally qualify nuclear cables, investigated effects on material integrity of thermal stress followed by irradiation stress to simulate in plant service. However, in actual service cables may be exposed to both stresses simultaneously. This has led to questions as to how well observed effects from sequential exposure can predict effects from simultaneous exposure. That is, are the degradation mechanisms for cable polymer materials different in the two different scenarios? Prior work has identified the possibility of anomalous aging behavior in XLPE such as the inverse temperature effect in which radiation exposure is more damaging at lower temperatures than at higher temperatures.

This report described results from our ongoing study of the effects of simultaneous thermal and gamma radiation exposure to cable materials. We focus here on cross-linked polyethylene (XLPE) insulation from modern Firewall® III nuclear cables, the class of cable insulation most commonly found within containment in U.S. nuclear power plants. We exposed XLPE insulation samples from commercial nuclear cable to gamma total doses up 32 kGy (32 Mrad) at dose rates from 116 to 540 Gy/h. These radiation exposures were performed while simultaneously heating the samples at 60, 90, or 115 °C. Cross-linking and scission of the polymer molecular chains in the XLPE insulation material with exposure was tracked using a solvent swelling method and the ‘percent gel’ and ‘uptake factor’ metrics.

In this work, percent gel of the aged XLPE samples was seen to increase with exposure temperature and uptake factor was seen to decrease with exposure temperature. Evidence for inverse temperature effects, in which radiation-induced damage is higher at lower temperature, was not observed for the materials and conditions considered here. Characterization of the effects of similar exposure of XLPE samples at 25 °C that is currently underway will provide additional indication of evidence to support understanding of the importance of inverse temperature effects for this material. This work directly addresses identified knowledge gaps in the aging and degradation of cable materials and cable systems related to simultaneous thermal and radiation aging including inverse temperature effects and dose rate effects.

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